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VALENTI, CRAIG FRANK
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WITH APPLICATION TO LIGHT SCATTERING.

CITY UNIVERSITY OF NEW YORK, PH.D., 1978

University Microfilms International 300 N. ZEEB ROAD, ANN ARBOR, MI 48106

A CONTINUUM THEORY OF MAGNETIC INSULATORS WITH APPLICATION TO LIGHT SCATTERING

by

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A dissertation submitted to the Graduate Faculty in Physics in partial fulfillment of the requirements for the degree of Doctor of Philosophy, The City University of New York.

1978

This manuscript has been read and accepted for the Graduate Faculty in Physics in satisfaction of the dissertation requirement for the degree of Doctor of Philosophy.

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ABSTRACT

Adviser: Professor Melvin Lax.

A consistent classical Lagrangian and Hamiltonian non-relativistic theory of a rigid non-conducting magnetic continuum in a magnetic field is presented. The theory starts with a Lagrangian density that describes the system. The Lagrangian equation of motion for the magnetization is shown to be that which generalizes Larmor's equation to continua, namely, $d\vec{m}/dt = \gamma\vec{m}\times\vec{B}^{eff}$. The transition to the Hamiltonian which describes the system is effected by means of Dirac's theory for systems with constraints. The Hamiltonian equation of motion for the time rate of change of the magnetization is identical with that obtained from the Lagrangian as it must be. Moreover, we calculate the generalized Poisson brackets (known as Dirac brackets) of the magnetization components and find $\{m_i(\vec{z},t),m_j(\vec{z}',t)\}^* = \gamma\epsilon_{ijK}m_K\ \delta^3(\vec{z}-\vec{z}')$.

Using the above results as a guide, a more complete macroscopic Lagrangian theory of linear and nonlinear electrodynamics has been constructed for an anisotropic magnetic dielectric possessing acoustic, ionic, electronic, magnetic or spin, and other internal excitations. This theory is applicable to a wide range of materials, namely insulators which are ferromagnetic, antiferromagnetic or ferrimagnetic.

The theory starts with a Lorentzian microscopic formulation in terms of massive point charges possessing an intrinsic spin and moving in a vacuum. This is converted to a long-wavelength macroscopic theory by taking the continuum limit. A Lagrangian is constructed from the vacuum electromagnetic Lagrangian, the usual interaction between the matter's charge-current-spin and the electromagnetic field, a kinetic energy of the matter's motion, a matter stored energy, and a term which accounts for the angular momentum of the spin and electron orbital motion generating the magnetization. This last term does not manifestly exhibit rotational invariance.

The stored energy of the matter must be invariant under arbitrary body rotations, displacements, spatial reflections and time reversal. It is therefore a function of basic invariants, namely: the finite strain tensor, body components of the internal coordinates associated with particle motion and body components of the sublattice magnetizations and their gradients. The stored energy is expanded as a polynomial in these basic invariants with coefficients, called material descriptors, which are restricted in form by the crystal magnetic space group symmetry. Effective local field effects and their possible absorption into the stored energy are considered. A discussion of the natural state of the magnetic crystal is also included.

Equations of motion for the electromagnetic field, the acoustic field, the internal excitations and the sublattice magnetizations follow deductively from the Lagrangian. Conservation laws of linear momentum, angular momentum and energy are formulated using the Lagrangian and/or the resultant equations of motion. These conservation laws are a direct consequence of the invariance of the total Lagrangian under the symmetry operations mentioned above with one exception: Angular momentum conservation for this system is not a result of manifest rotational invariance of the Lagrangian. It is also shown that even though angular momentum is conserved, the stress tensor for this system is asymmetric.

The equations of motion are linearized, and the general theory is applied to the problem of Raman scattering by magnons or the magnon-phonon mixed modes in anisotropic magnetic crystals. This requires a solution of the electric field wave equation which is accomplished by means of a Green's function technique. Proper consideration is given to the noncollinearity of propagation and Poynting vectors which is due to the optical anisotropy. Moreover, surface correction factors due to solid angle expansion, effective scattering volume demagnification and transmissivity are accounted for. The Raman scattering efficiency is evaluated by means of the fluctuation-dissipation theorem.

ACKNOWLEDGEMENTS

The author wishes first to thank Professor Melvin Lax without whose guidance and encouragement this work would hardly have been possible. Thanks also go to Mrs. Joan Lemberger for her kindness in providing secretarial assistance. Finally, but not least of all, the author wishes to thank his wife, Helen, for her patience and understanding, and I join her in silently shouting "hurrah"!

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I. INTRODUCTION

Some classical and quantum theories of ferromagnetic systems are based on the Hamiltonian formalism and either assume the magnetic torque equation or derive it on the basis of assumed commutator or Poisson bracket relations between the spin components.[1-4] Nodvik[5] has presented a classical Hamiltonian theory of a single particle with spin which possesses the expected Poisson brackets and magnetic torque equation. However, it does not appear that his formalism can easily be generalized to continuous media. The work of Tiersten and Tsai[6] is indicative of the conservation law approach from which equations of motion are obtained from a construction of assumed conservation laws. A variational approach to the problem of ferromagnetic media which takes account of the spin angular momentum in the variational principle through a virtual work term not present in the Lagrangian, has been performed by Maugin and Eringen.[7] A more recent approach to the problem using the elegant mathematical formalism of the principle of virtual power, which appears to yield better results, has been published by Maugin.[8] Gilbert[9] and Brown[10] make the analogy of a spinning top to the spin and introduce Euler angle coordinates into the problem. In the Lagrangian they construct, two moments of inertia are set equal to zero. Kittel[11] also performs a classical calculation with a Lagrangian valid only for static magnetic fields. The quantum mechanical derivation of the magnetic torque equation for a ferromagnetic medium assuming spin disturbances of long wavelength has been done in the classic paper of Herring and Kittel.[12]

It is the purpose of Chapter II to give a consistent classical formulation of a rigid magnetic continuum, and to shed light on how one can make the transition from a classical theory to a quantum theory. The formalism is based on a Lagrangian density which has previously not been considered. This Lagrangian density is termed singular in that the generalized velocities cannot be expressed uniquely in terms of the canonical momentum densities. This is due to constraints imposed by the form of the Lagrangian alone.

The question also arises as to what the generalized coordinates, corresponding to the magnetic or spin degrees of freedom in a magnetic medium are. The answer to this question is important in writing down the canonical stress tensor, since that quantity is defined in terms of the Lagrangian density and all the generalized coordinates. It appears that the choice of the magnetization components and their time derivatives as generalized coordinates and velocities is consistent. This is the primary result of Chapter

II, and it motivates the derivation of the stress tensor and conservation laws in Chapter III which describe an elastic magnetic dielectric crystal having any symmetry and degree of anisotropy.

The Hamiltonian of the rigid system is obtained by means of Dirac's[13-18] generalized dynamics when a singular Lagrangian is involved. Dirac was the first to show that the algebra of Poisson brackets determines a division of constraints into two classes: the so-called first class constraints and second class ones. The first class constraints are those that have zero Poisson brackets with all other constraints in the subspace of phase space in which the constraints hold; constraints which are not first class are by definition second class. Dirac also showed how to redefine the Poisson brackets in such a way that all new redefined brackets (so-called Dirac brackets) of second class constraints are zero. It is the Dirac brackets to which one must apply the standard rules in going over to the quantum theory.

Other important work in the problem of developing a consistent classical Hamiltonian dynamics when there is a singular Lagrangian describing a system has been done by many others[19-25], notably Bergmann. The interested reader is also referred to a recent and very comprehensive review by Hansen et al.[26]

Chapter II will be organized as follows: in Section

2.1, we will give the Lagrangian density and derive the magnetic torque equation. In Section 2.2 we will give a brief review of Dirac's method. In Section 2.3, we will transform to the Hamiltonian density and rederive the magnetic torque equation, also obtaining the Dirac brackets between components of the magnetization.

Recently Lax and Nelson developed an ab initio Lagrangian theory describing the interaction of the electromagnetic field with an elastic dielectric.[27] The theory can be used to describe crystals having any symmetry and degree of anisotropy, having any number of particles (ions and electrons) per unit cell, and having nonlinearities of any order in their constitutive relations.

The classical approach of this theory is well justified and has a wide range of applicability in linear and nonlinear optics and acoustics. Nonlinear interactions of light and sound fields can be formulated without quantization of the fields when energies are large compared to the quantum. Ionic motions can be treated classically when driven nonlinear processes are being considered. Moreover, optical absorption associated with excitons has been studied with a classical treatment.[28,29] It is also possible to treat Raman and Brillouin scattering in a classical fashion.[30]

Application of the theory to the photoelastic interaction[31] led to the prediction that the independent elastic variable relevant to this interaction was the displacement gradient, not the strain as long believed. Application of the general theory to acoustically induced optical harmonic generation[32] demonstrated the usefulness of the theory in predicting indirect contributions to the overall effect. The general theory has also been used to describe the elasticity and piezoelectricity in pyroelectrics,[33,34] materials possessing a spontaneous electric dipole moment.

In Chapter III the theory is extended to materials possessing a spontaneous magnetic dipole moment generated by intrinsic spin and electron orbital motion. It has long been established[35] that a classical treatment of magnetic properties is sufficient in the long wavelength continuum limit. Once the general theory is extended, it will be possible to study electromagnetic and acoustic waves in ferromagnets, antiferromagnets and ferrimagnets. Moreover, it could be determined whether the spontaneous magnetic moment or field will lead to indirect contributions to the elasticity, magnetoelasticity, piezoelectricity, magnetoelectricity or other phenomena. It would also be useful to ascertain whether rotations, in contrast to strains, would play a role in any such indirect effects. Toward this end it is advantageous to set up the theory in a general way that includes all levels of nonlinearity.

For simplicity we will omit wave vector dispersion effects such as optical activity which are related to electric polarization gradients. However, dispersive effects related to magnetization gradients and displacement gradients (strain) are retained. We also exclude pyroelectric phenomena, energy dissipation mechanisms and all thermal phenomena from the present discussion.

The concept of rotational invariance developed by Toupin[36] in his static and dynamic theories of the elastic dielectric, was first applied to magnetoelasticity by Vlasov and Ishmukhametov[37], Brown[38] and Tiersten.[39] Vlasov and Ishmukhametov derive equations of motion describing the dynamic behavior of magnetoelastic anisotropic media from a Lagrangian, and show the rotation of volume elements of the media accompanying elastic deformations should be taken into account. Brown uses a variational principle to treat the static case, and Tiersten uses a continuum model and the notion of a quasi-static magnetic field to treat the dynamic case in the presence of dissipation and thermal effects. The experimental results and analyses for yttrium iron garnet (YIG) by Eastman[40] and for the antiferromagnet MnF_2 by Melcher[41] were the first to show that a rotationally invariant nonlinear description of magnetoelasticity was correct while the original linear theories of magnetoelasticity of Kittel[42], Schlomann[43], and Akhiezer, Bariakhtar and Peletminskii[44] based on the infinitesimal magnetostrictive theory of Becker and Doring[45] were in error. The rotationally invariant description takes account of the fact that the magnetization may couple to the elastic deformations not only through the strain but also through the rotational part of the deformations.

Recent work on rotational effects and the propagation of acoustic waves in elastic ferromagnets and antiferromagnets has been done by R. L. Melcher.[46] Like Akhiezer et al, Melcher assumes a Hamiltonian, assumes the torque equation for the magnetic moment and assumes the conservation laws. Moreover, the energy density is equated to the Hamiltonian density. Only the interaction of acoustic waves with magnetic spin waves[47] is considered; no allowance is made for other internal excitations.

Dixon and Eringen[48] employ a conservation law approach from which they obtain field equations, jump conditions and constitutive equations for a polar elastic dielectric subject to deformations and electromagnetic fields. In the derivation of the above equations, a distributed Lorentz force for each volume element is assumed, and Farraday's, Ampere's and Gauss' laws are applied. No intrinsic spin is included in their development, but the magnetic dipole moment due to the motion of the bound charges is considered.

Maugin and Eringen[49] formulate a variational approach to the study of elastic solids in which the magnetization is constant in magnitude. However, electric fields, currents, charges and polarizations (the quasi-magnetostatic approach) are ignored, and therefore, their theory is not a fully dynamical theory. This is equivalent to saying that the velocity of dynamical phenomena is small in comparison to the propagation velocity of electromagnetic perturbations; and that boundary or jump conditions can be written for surfaces which may be considered as being stationary. No internal coordinates (extra degrees of freedom of structural origin) are allowed for, and the Lagrangian used is not the total one but must be supplemented with an expression for the virtual work of body and surface loads. Account is taken of the spin angular momentum in the variational principle through a virtual work term not derivable from any term in the Lagrangian. More recently, Maugin[50] has employed the elegant formalism of the principle of virtual power to obtain what appears to be better results.

The conservation law approach is used by Tiersten and Tsai[51] to obtain the differential equations and boundary conditions describing a finitely deformable, polarizable, and magnetizable, heat conducting continuum in interaction with the electromagnetic field. Their model consists of an electronic charge and spin continuum coupled to a lattice continuum, which itself consists of two interpenetrating ionic continua. A similar model without spin for the case of the heat conducting elastic dielectric had been introduced earlier by Tiersten.[52] An assumption of the model is that stresses are present not at all ionic continua boundaries, but only at those separating elements of the same ionic continuum. Moreover, for the case of more than one spin continuum, it is assumed that the magnetic exchange interaction occurs only between elements of the same spin continuum. Our model differs in this one important aspect: it takes into account all particle and spin continua interactions. The results we obtain appear fundamentally different from those of Tiersten and Tsai. Moreover, the underlying description seems closer to the actual physical situation.

Wong and Grindlay[53] also use a conservation law approach in writing down the equations of motion describing an elastic dielectric material. Currents are ignored, but the extremely important concept of a stored energy constructed with due consideration of symmetry requirements is used. However, the contention that the stress tensor must be symmetric because angular momentum is conserved is at odds with our findings.

In contrast to Tiersten and Tsai and many of the other treatments, we begin with a microscopic discrete particle model before taking the continuum limit. This aids in constructing the proper forms of the interactions. Moreover, in our Lagrangian approach only the Lagrangian has to be constructed; all other quantities such as body forces and work done on the matter by the field, etc., follow deductively. There is no need to assume the conservation laws; they follow automatically and naturally from the Lagrangian or its resultant equations of motion. There is no need to assume the Maxwell equations; these follow naturally as equations of motion from the Lagrangian using the electromagnetic scalar and vector potentials as canonical coordinates. Another advantage to our theory is that we are not limited to any specified number of particles per unit cell. This enables us to account for all material resonances. We have also addressed ourselves to the problem of the local fields, and have made an attempt to account for them. There are widespread discrepancies in the results of those referred to. Our results are no exception, but we believe our theory is valid, and our procedures less likely to incur errors.

Chapter III is organized as follows: in Section 3.1 we write down a microscopic Lagrangian. In Section 3.2, we pass to the continuum limit by replacing the discrete coordinates with a set of functions of the continuous material variable $\vec{X}$. The set of functions of the position coordinates are then transformed to a set of internal coordinates. With the construction in Section 3.3 of the stored energy as invariant under arbitrary body rotations, displacements, translations, spatial reflections and time reversal, we have completed the transition to a macroscopic Lagrangian. In Section 3.4, we obtain the Maxwell-Lorentz electromagnetic equations, the center of mass equation of motion in Section 3.5. In Section 3.6 we examine local field effects and find that they can be absorbed into the stored energy. The internal motion equations are obtained from the Lagrangian in Section 3.7. A non-rotationally invariant term accounting for the angular momentum of the spin and electron orbital motion is added to the Lagrangian in Section 3.8, and the magnetization equation of motion is obtained. Conservation of linear momentum and the stress tensor are formulated in Section 3.9. The uniqueness of our stress tensor is established in Section 3.10. The asymmetry of the stress tensor for a magnetic dielectric is proven in Section 3.11. Conservation of angular momentum and conservation of energy are presented in Section 3.12 and Section 3.13 respectively.

In Chapter IV the equations of motion are linearized in the dynamical variables, and general expressions for the nonlinear interactions are ascertained. These results are a preliminary to any interative technique for the solution of the nonlinear equations of motion. In this linearization procedure, we follow very closely Lax and Nelson[54].

Chapter IV is organized as follows: In Section 4.1, expressions which relate derivatives in the spatial frame to derivatives in the material frame are derived. In Section 4.2 the particle equations of motion are rederived valid to all orders as a preliminary to the linearization of the center of mass and the internal coordinate equations of motion in Section 4.3 and Section 4.4, respectively. In Section 4.5, the magnetic equation of motion is linearized.

In Chapter V an application of the theory is made to the problem of the nonlinear interaction of light at optical frequencies with magnon-phonon-photon mixed modes which may be referred to as magneto-polaritons. The elastic degree of freedom (acoustic phonons) will not enter into this problem because of the higher frequencies involved.

Raman scattering by spin waves or magnons was first suggested by Bass and Kaganov[55], and by Elliot and Loudon[56], and has been further analyzed by Shen and Bloembergen[57] and also Fleury and Loudon[58]. These latter authors have concluded that one-magnon scattering proceeds through a mechanism consisting of an indirect electric-dipole coupling via the spin-orbit interaction. They also propose an "exchange scattering" mechanism for two-magnon scattering which can be very strong in some antiferromagnets. Experimental results for one-and two-magnon scattering processes in the antiferromagnets FeF_2, MnF_2 and NiF_2 have been reported by Fleury et al.[59,60,61] The coupling of magnons and photons in antiferromagnets to form mixed modes called "magnitons" has been discussed previously.[62,63]

In contrast to the Fleury-Loudon and Shen-Bloembergen treatments, ours is entirely classical. Moreover, we will only consider Raman scattering by a single magneto-polariton. The classical treatment models very closely that of Lax and Nelson's for the polariton.[64] An electric field wave equation driven by a nonlinear source is derived for the case of a magnetic media. Our theory enables us to obtain expressions for the linear and nonlinear polarizations and magnetizations. The linear expressions or constitutive relations are used in the linear portion of the wave equation, while the nonlinear expressions are just the basis for the driving nonlinear source terms in the wave equation. This wave equation is solved using a Green's function technique developed by Lax and Nelson.[65] This solution is then used to find an expression for the scattered power inside the crystal. The detected power outside the crystal is then calculated from that inside with due respect to surface correction factors:

1 The surface of the crystal acts as an optical instrument that expands the solid angle of the scattered beam.

2 The detected power is limited by the solid angle presented by the detector to the scattered beam.

3 The surface also acts to decrease the apparent size of the source volume V_s.

The Raman scattering efficiency which is the ratio of scattered to incident powers outside the crystal will contain correlation functions. These correlation functions are calculated by means of the fluctuation-dissipation theorem introduced into nonlinear optics by Butcher and Ogg[66] and applied to the polariton problem by Barker and Loudon[67] and Lax and Nelson.[68]

Chapter V is organized as follows: in Section 5.1, some simplifying assumptions and their justifications are given.

In Section 5.2, the linear constitutive relations for the polarization and magnetization are derived. The driven wave equation for the electric field is developed in Section 5.3, and the inside Green's function technique for its solution is presented in Section 5.4 In Section 5.5, a brief digression on surface corrections is made. Expressions for the scattered power and the Raman efficiency in terms of correlation functions between components of the nonlinear polarization are determined in Section 5.6. A brief treatment of the fluctuation-dissipation theorem is presented in Section 5.7. In Section 5.8, the nonlinear polarization appropriate to a magnetic dielectric is determined. The correlation functions between components of the nonlinear polarization are evaluated with the help of the fluctuation-dissipation theorem. With the correlation functions evaluated, the Raman scattering efficiency is determined.

This thesis ends with Chapter VI, a summary and discussion of the principal results.

II. THE RIGID MAGNETIC CONTINUUM

2.1 The Lagrangian Formalism

We assume that we are dealing with a magnetic continuum which is obtained from a crystal lattice of one sublattice per unit cell by taking the appropriate long wavelength limit. Each point of the continuum is described by a magnetization vector whose magnitude remains constant:

$$|\vec{m}(\vec{z},t)|^2 = 1 \quad , \tag{2.1.1}$$

where we have normalized to unit length for simplicity. We find it necessary to account for the angular momenta of the intrinsic spin and electron orbital motion which generate the magnetization. We assume this can be done by including into the Lagrangian density a gyroscopic term[69] of the form

$$\hat{K} = \frac{1}{3\gamma} \sum_J \frac{(\vec{m} \cdot \hat{e}^J)\,[\dot{\vec{m}} \cdot (\hat{e}^J \times \vec{m})]}{|\vec{m}|^2 - (\vec{m} \cdot \hat{e}^J)^2} \quad . \tag{2.1.2}$$

γ is identified as the gyromagnetic ratio (assumed here to be isotropic), and $\hat{e}^J, J = 1,2,3$, is a set of unit orthogonal cartesian vectors which are "external". By this we mean they are fixed in space and their orientation with respect to the inertial frame of reference is arbitrary. Of course, manifest rotational invariance of the Lagrangian will be lost by the inclusion of these arbitrary vectors. However, as we will show, the results of our theory will be independent of these vectors. In passing, it is interesting to note that the kinetic energy term of the Doering-Gilbert Lagrangian[70,71] may be obtained from Eq. (2.1.2) by removing the summation over J and factor of 1/3 and choosing the resultant single external vector to be oriented along the z axis.

The use of external vectors is not altogether unknown in quantum field theory. For example, Zwanziger[72] in his quantum field theory of electric and magnetic charges uses a Lagrangian which depends on an arbitarily fixed four vector and thus loses manifest Lorentz invariance. However, invariance is regained under the restriction that values of the combination of electric and magnetic charge $(e_n g_m - g_n e_m)$ be quantized. He, moreover, shows that angular momentum may be conserved even though the Lagrangian itself might not be rotationally invariant.

As the basis for our formalism, we use the Lagrangian density

$$\hat{L} = \hat{K} + \vec{m} \cdot \vec{B} - V + \lambda[|\vec{m}|^2 - 1] \quad , \tag{2.1.3}$$

where $\vec{B}$ is the magnetic field and the constraint, Eq. (2.1.1), is introduced by means of the undetermined multiplier λ. V is the stored or potential energy per unit volume which in the long-wavelength limit is taken to be a function of the magnetization components m_i and their first spatial derivatives $m_{i,j} \equiv \partial m_i/\partial z_j$. This stored energy phenomenologically accounts for interactions of quantum mechanical origin not expressible in terms of electromagnetic interactions. These include, for example, the exchange and magnetic anisotropy interactions and the spin orbit coupling.

The stored energy of the matter must be invariant under arbitrary body rotations, displacements, spatial reflections and time reversal. It is generally assumed that it can be expanded as a polynomial in m_i and $m_{i,j}$ with coefficients which are restricted in form by the crystal magnetic space group symmetry. In this chapter we are not concerned with the exact form of the stored energy. All that need be assumed is its functional dependence on m_i and $m_{i,j}$, and its rotational invariance. The proper construction of the stored energy for the more complicated case of an elastic anisotropic magnetic dielectric possessing accoustic, ionic, electronic, magnetic or spin, and other internal excitations has been done in Chapter III.

The Lagrangian density given in Eq. (2.1.3) is of course not the total Lagrangian density of the system. Interaction and field Lagrangian densities of the form[73] (rationalized MKS)

$$\hat{L}_I = \vec{j}(\vec{z},t) \cdot \vec{A}(\vec{z},t) - q(\vec{z},t)\Phi(\vec{z},t) \quad , \qquad (2.1.4)$$

$$\hat{L}_F = \frac{1}{2}\epsilon_0 \; [\vec{E}^2(\vec{z},t) - c^2 \; \vec{B}^2(\vec{z},t)] \quad , \qquad (2.1.5)$$

should be included. Here the electric and magnetic field vectors $\vec{E}$ and $\vec{B}$ are defined in terms of the scalar and vector potentials Φ and $\vec{A}$ by means of

$$\vec{E}(\vec{z},t) = -\nabla\Phi(\vec{z},t) - \frac{\partial\vec{A}(\vec{z},t)}{\partial t} \quad , \qquad (2.1.6)$$

$$\vec{B}(\vec{z},t) = \nabla \times \vec{A}(\vec{z},t) \quad , \qquad (2.1.7)$$

and $\vec{j}$ and $\vec{q}$ represent the current and charge densities respectively. However, our primary concern in this chapter being the magnetic or spin degrees of freedom represented by the magnetization, we will for simplicity not consider the interaction and field Lagrangian densities given by Eqs. (2.1.4) and (2.1.5) as they do not couple in to $\vec{m}(\vec{z},t)$.

When a first order space derivative is present in the Lagrangian density, the Lagrange equation of motion for a generalized coordinate q_i becomes

$$\frac{d}{dt}\frac{\partial \hat{L}}{\partial \dot{q}_i} = \frac{\partial \hat{L}}{\partial q_i} - \frac{\partial}{\partial z_j}\frac{\partial \hat{L}}{\partial q_{i,j}} \equiv \frac{\delta L}{\delta q_i} \quad , \qquad (2.1.8)$$

where $L = \int \hat{L} d^3\vec{z}$. Here $\delta/\delta q_i$ is known as a functional derivative. We choose the magnetization components $m_i(\vec{z},t)$ as generalized coordinates for the Lagrange equation of motion, Eq. (2.1.8). Using Eqs. (2.1.3) and (2.1.8) we obtain the equation of motion:

$$\frac{d}{dt}\frac{\partial \hat{K}}{\partial \dot{\vec{m}}} - \frac{\partial \hat{K}}{\partial \vec{m}} - 2\,\lambda\,\vec{m} = \vec{B}^{eff} \quad , \qquad (2.1.9)$$

where the effective magnetic field is defined by

$$\vec{B}^{eff} \equiv \vec{B}(\vec{z},t) - \left(\frac{\partial V}{\partial \vec{m}} - \frac{\partial}{\partial z_j}\frac{\partial V}{\partial \vec{m}_{,j}}\right) \quad . \qquad (2.1.10)$$

Multiplying Eq. (2.1.9) by the gyromagnetic ratio γ and taking the vector cross product with $\vec{m}$, we obtain after some nontrivial algebra and use of Eq. (2.1.1) in the form $\vec{m}\cdot\dot{\vec{m}} = 0$, the equation

$$\frac{1}{3}\sum_J \frac{(\dot{\vec{m}}\cdot\hat{e}^J)\,\vec{m}\times(\hat{e}^J\times\vec{m}) - [\vec{m}\cdot(\dot{\vec{m}}\times\hat{e}^J)]\,(\vec{m}\times\hat{e}^J)}{|\vec{m}|^2 - (\vec{m}\cdot\hat{e}^J)^2}$$

$$= \gamma\vec{m}\times\vec{B}^{eff} \quad . \qquad (2.1.11)$$

The components of the arbitrary external vectors $\hat{e}^J_k (J = 1,2,3)$ may be represented by an orthogonal matrix

$$e_k^J = S_{kJ} \quad . \tag{2.1.12}$$

Eq. (2.1.11) may then be recast into the matrix equation

$$A_{ij}\, \dot{m}_j = \gamma(\vec{m} \times \vec{B}^{eff})_i \quad , \tag{2.1.13}$$

where

$$A_{ij} = \frac{1}{3} \sum_J \frac{[|\vec{m}|^2\ S_{iJ} - m_k\ S_{kJ}\ m_i]\ S_{jJ} + \epsilon_{ika}\ S_{kJ}\ m_a\ \epsilon_{jcb}\ S_{cJ}\ m_b}{|\vec{m}|^2 - (m_l S_{lJ})^2} \quad . \tag{2.1.14}$$

The Einstein convention of summation over repeated indices is utilized here.

It is possible to express A_{ij} as a sum of two parts:

$$A_{ij} = \delta_{ij} + V_i\ m_j \quad , \tag{2.1.15}$$

where

$$V_1 = \frac{1}{3} \sum_J \frac{m_2\ S_{1J}\ S_{2J} - m_1(S_{2J}\ S_{2J} + S_{3J}\ S_{3J}) + m_3\ S_{1J}\ S_{3J}}{|\vec{m}|^2 - (m_k\ S_{kJ})^2} \quad , \tag{2.1.16a}$$

$$V_2 = \frac{1}{3} \sum_J \frac{m_1\ S_{1J}\ S_{2J} - m_2(S_{1J}\ S_{1J} + S_{3J}\ S_{3J}) + m_3\ S_{2J}\ S_{3J}}{|\vec{m}|^2 - (m_k\ S_{kJ})^2} \quad , \tag{2.1.16b}$$

$$V_3 = \frac{1}{3} \sum_J \frac{m_1\ S_{1J}\ S_{3J} - m_3(S_{1J}\ S_{1J} + S_{2J}\ S_{2J}) + m_2\ S_{2J}\ S_{3J}}{|\vec{m}|^2 - (m_k\ S_{kJ})^2} \quad .$$

(2.1.16c)

In view of the constraint given by Eq. (2.1.1), $\vec{m}$ is perpendicular to $\dot{\vec{m}}$. Thus, the equation of motion, Eq. (2.1.13), with the help of Eq. (2.1.15) simplifies to

$$\dot{\vec{m}} = \gamma(\vec{m} \times \vec{B}^{eff}) \quad . \tag{2.1.17}$$

This is just the generalization of Larmor's precession equation to continua. We also note that the result is independent of the external vectors $\hat{e}^J (J = 1,2,3)$.

2.2 Dirac's Theory of Systems with Constraints

A. Singular Lagrangians

Given a mechanical system (of N degrees of freedom) with a Lagrangian L,

$$L = L(q,\dot{q}) \quad , \tag{2.2.1}$$

one defines the conjugate momenta by

$$p_n = \frac{\partial L}{\partial \dot{q}_n} \qquad (n = 1,...N) \quad . \tag{2.2.2}$$

We shall dwell on the case when the expressions $\partial L/\partial \dot{q}_n$ are not independent functions of $\dot{q}_n$. Eliminating the $\dot{q}$'s one obtains a certain number of independent constraints (called primary constraints)

$$\varphi_m(q,p) = 0 \qquad (m = 1,2,\ldots k) \quad . \qquad (2.2.3)$$

Thus, k of the p's are not independent. Solving Eq. (2.2.3) for k such p's in terms of the rest we get

$$p_\alpha = \Psi_\alpha(q,p_i) \qquad (\alpha = 1,2,\ldots k) \quad , \qquad (2.2.4)$$

where the p_i's (i = k + 1,N) are independent.

B. Equations of Motion

We first introduce several important definitions. Define the phase space Γ as a set whose elements are ordered 2N-tuples $(q_1,\ldots q_N,p_1,\ldots p_N)$. Then, introduce the submanifold $\overline{M}$ in Γ which, by definition, is the subset of Γ for which the constraints, Eq. (2.2.3), hold.

The Poisson bracket for two functions f and g of the q's and p's is defined by

$$\{f,g\} = \sum_{n=1}^{N} \left(\frac{\partial f}{\partial q_n}\frac{\partial g}{\partial p_n} - \frac{\partial f}{\partial p_n}\frac{\partial g}{\partial q_n}\right) \quad . \qquad (2.2.5)$$

It is convenient to introduce the "total" Hamiltonian H' in the following way:

$$H' = H + v_m \varphi_m = p_n \dot{q}_n - L + v_m \varphi_m \quad , \qquad (2.2.6)$$

where the v_m are arbitrary Lagrange multipliers. [If additional ("secondary") constraints are found later, Eq. (2.2.6) will have to be modified to include them.] In the standard fashion one is led to the equations of motion

$$\dot{p}_n \Big|_{\bar{M}} = \{p_n, H'\} \Big|_{\bar{M}} = -\frac{\partial H}{\partial q_n} - v_m \frac{\partial \varphi_m}{\partial q_n} \quad ,$$

$$\dot{q}_n \Big|_{\bar{M}} = \{q_n, H'\} \Big|_{\bar{M}} = \frac{\partial H}{\partial p_n} + v_m \frac{\partial \varphi_m}{\partial p_n} \quad , \qquad (2.2.7)$$

where our notation is intended to emphasize that these equations hold in the submanifold $\bar{M}$. For a function g of p_n and q_n we find the equation of motion

$$\dot{g} \Big|_{\bar{M}} = \{g, H'\} \Big|_{\bar{M}} \quad . \qquad (2.2.8)$$

Hence, H' as given by Eq. (2.2.6) is the generator of time translation.

C. First-class and second-class constraints

The constraints φ_m must remain zero at all times, which implies:

$$\dot{\varphi}_m \Big|_{\bar{M}} = [\{\varphi_m, H\} + v_n\{\varphi_m, \varphi_n\}] \Big|_{\bar{M}} = 0 \quad . \qquad (2.2.9)$$

Excluding the case when these equations are contradictory either among themselves or with Eq. (2.2.3), the equations obtained may be (a) a trivial identity, (b) independent of the v's, (c) may involve some of the v's.

In case they are of type (b), they represent new constraints (called secondary constraints) and may be written in the form

$$\rho_i(q,p) = 0 \quad . \tag{2.2.10}$$

We can continue this process of generating secondary constraints until we arrive at the point when no more independent equations of type (b) are produced. This process must terminate since the original number of field variables is finite. After eliminating as many v's as possible from (c) type equations, we can use the remaining equations to solve for some (or all) of the v's.

Let us denote by M the subspace of phase space in which all constraints hold (i.e. both primary and secondary ones). We shall assume the irreducibility of all constraints with respect to M, i.e. any function of q's and p's vanishing in M must be expressible as a linear function of the φ's and the ρ's with functions of q's and p's as coefficients. We thus have in particular

$$H' = H(q,p) + v_1\,(q,p)\;\Psi_1\,(q,p) \;, \tag{2.2.11}$$

where we have denoted by a common symbol $\Psi_l(q,p)$ all the constraints, i.e. $(\Psi_l) = ((\varphi_m),(\rho_i))$.

By definition, a <u>first-class</u> constraint φ_a (secondary or primary) satisfies

$$\{\varphi_a, \Psi_l\} \Big|_M = 0 \qquad (2.2.12)$$

for all Ψ_l, and thus in view of our irreducibility hypothesis the Poisson brackets must be functions of the complete set of constraints Ψ_l. Since first-class constraints generate infinitesimal contact transformations (that do not affect the physical state of the system), we may expect a linear Poisson bracket relationship characteristic for generators of a Lie group:

$$\{\varphi_a,\Psi_l\} = \lambda_{alk}\, \Psi_k \quad . \qquad (2.2.13)$$

Such transformations are termed gauge transformations, and there exists a certain amount of freedom in choosing explicit forms for each gauge.

We call a constraint θ_a <u>second-class</u> if it is not first-class; i.e. it has a non-vanishing Poisson bracket with at least one other constraint.

D. Dirac brackets and quantization

A naive transition to quantum theory would consist in imposing the constraints as conditions on the quantum state vectors and replacing standard Poisson brackets by "$-i$" times commutators. But then if

$$\Psi_1 | a \rangle = 0, \quad \Psi_2 | a \rangle = 0 \quad , \tag{2.2.14}$$

we find

$$[\Psi_2, \Psi_1] | a \rangle = 0 \quad , \tag{2.2.15}$$

which corresponds to a classical equation

$$\{\Psi_2, \Psi_1\} \Big|_M = 0 \quad . \tag{2.2.16}$$

Thus, for the naive passage to quantum theory to be possible, all constraints must be first-class. In case a mechanical system has second-class constraints, θ_a, the remedy consists in redefining the Poisson brackets in a suitable manner:

$$\{\xi, \eta\}^* = \{\xi, \eta\} - \{\xi, \theta_a\} C_{ab} \{\theta_b, \eta\} \quad , \tag{2.2.17}$$

where C_{ab} is defined by

$$C_{ab}\{\theta_b, \theta_c\} = \delta_{ac} \quad . \tag{2.2.18}$$

The new bracket, $\{\xi, \eta\}^*$, is called a Dirac bracket, while the brackets on the right-hand side are standard Poisson brackets. It can be shown that the new brackets have all the standard properties of Poisson brackets. Note that if either ξ or η is a first class constraint, the Dirac bracket reduces to the Poisson bracket because of the definition, Eq. (2.2.12).

As a consequence of the definitions, Eqs. (2.2.17) and (2.2.18), we find that the Dirac bracket of any dynamical quantity ξ with a second class constraint vanishes:

$$\{\xi,\theta_a\}^* = \{\xi,\theta_a\} - \{\xi,\theta_b\} C_{bc}\{\theta_c,\theta_a\}$$

$$= \{\xi,\theta_a\} - \{\xi,\theta_b\}\delta_{ba} = 0 \quad . \qquad (2.2.19)$$

The consistency condition Eq. (2.2.9) and the definition of the Dirac bracket, Eq. (2.2.17), imply that the new bracket may also be used to yield the Hamiltonian equations of motion:

$$\{g,H'\}^* \Big|_M = \{g,H'\} \Big|_M = \dot{g} \Big|_M \quad . \qquad (2.2.20)$$

The passage to quantum theory can now be made by replacing the new brackets by "$-i$" times commutators. Then, in quantum theory, we can take the second class constraints $\theta_a = 0$ to hold as operator equations without any contradiction, since in view of Eq. (2.2.19) $[\theta_a,\xi] = 0$ for any operator ξ. The first class constraints, φ_a, can continue to be handled as in Eq. (2.2.14) as conditions on the state vector. Alternatively, gauge conditions (to be considered as new constraints) can be imposed so as to convert first class constraints into second class constraints. The latter can then be handled by the Dirac bracket procedure.

In summary, once we agree to use Dirac brackets, we have effectively taken into account certain dependencies among the field variables as reflected by the second class constraints. Thus, we can ignore these constraints or equivalently, take the second class constraints $\theta_a = 0$ to hold as operator equations (this is sometimes referred to as being "strongly" equal to zero). We cannot ignore the first class constraints φ_a. The dependencies among the field variables reflected by these constraints must be accounted for. In quantum theory this can be done by imposing the first class constraints as conditions on the state vectors, $\varphi_a \mid a \rangle = 0$ (this is sometimes referred to as being "weakly" equal to zero). The first class constraints could also be accounted for by introducing new constraints called "gauge conditions" which effectively convert <u>all</u> constraints into second class constraints from which new Dirac brackets can be determined before going over to quantum theory.

E. Generalization to Field Theory

The generalization of this formalism to field theory, i.e. a mechanical system with a continuously infinite number of degrees of freedom, presents no difficulty. The important things to remember is that p_n is to be considered as a conjugate momentum "density" defined by the functional derivative of a Lagrangian

$$p_n \equiv \frac{\delta L}{\delta \dot{q}_n} = \frac{\partial \hat{L}}{\partial \dot{q}_n} \quad , \tag{2.2.21}$$

and the Poisson bracket of two functions f and g of the q's and p's, not depending explicitly on time, is defined by

$$\{f,g\} = \int \sum_{n=1}^{N} \left(\frac{\delta f}{\delta q_n}\frac{\delta g}{\delta p_n} - \frac{\delta f}{\delta p_n}\frac{\delta g}{\delta q_n}\right) d^3\vec{z} \quad . \tag{2.2.22}$$

Here f and g are assumed to be expressible in terms of density functions:

$$f = \int F \, d^3\vec{z} \quad ,$$

$$g = \int G \, d^3\vec{z} \quad . \tag{2.2.23}$$

As a result of Eqs. (2.2.22) and (2.2.23) we have

$$\{q_i(\vec{z},t), p_j(\vec{z}',t)\} = \delta_{ij}\delta^3(\vec{z}-\vec{z}') \quad ,$$

$$\{\frac{\partial q_i}{\partial z_j}(\vec{z},t), p_k(\vec{z}',t)\} = \{q_i(\vec{z}',t), \frac{\partial p_k}{\partial z_j}(\vec{z},t)\} = \delta_{ik}\frac{\partial}{\partial z_j}\delta^3(\vec{z}-\vec{z}') \quad ,$$

$$\{q_i(\vec{z},t),q_j(\vec{z}',t)\} = \{p_i(\vec{z},t),p_j(\vec{z}',t)\} = 0 \quad . \tag{2.2.24}$$

Finally, the generalizations of Eqs. (2.2.17) and (2.2.18) to continuous systems are

$$\{\xi(\vec{z}),\eta(\vec{z}')\}^* = \{\xi(\vec{z}),\eta(\vec{z}')\} - \iint d\vec{x}\, d\vec{y}\, \{\xi(\vec{z}),\theta_a(\vec{x})\}\, C_{ab}(\vec{x},\vec{y})\, \{\theta_b(\vec{y}),\eta(\vec{z}')\}, \tag{2.2.25}$$

and

$$C_{ab}(\vec{x},\vec{y}) = \{\theta_a(\vec{x}),\ \theta_b(\vec{y})\}^{-1} \quad , \tag{2.2.26}$$

where the inverse is defined by the condition

$$\iint d\vec{x}'\, d\vec{y}\, C_{ij}\, (\vec{x},\vec{y})\{\theta_j(\vec{y}),\ \theta_k(\vec{x}')\} = \delta_{ik} \quad . \tag{2.2.27}$$

2.3 The Hamiltonian Formalism

The first task is to find all the constraints of our system, and whether they are first-class or second-class. The momentum density conjugate to m_i is given, with the help of Eqs. (2.1.2) and (2.1.3), by

$$p_i = \frac{\delta\hat{L}}{\delta\dot{m}_i} = \frac{1}{3\gamma}\sum_J \frac{(m_e\ S_{eJ})\ \epsilon_{ijk}\ S_{jJ}\ m_k}{|\vec{m}|^2 - (m_a\ S_{aJ})^2} \quad , \tag{2.3.1}$$

and that conjugate to λ is given by

$$P = \frac{\partial \hat{L}}{\partial \dot{\lambda}} = 0 \quad . \tag{2.3.2}$$

(It should be remarked that the choice of λ as a dynamical variable is somewhat artificial, but is needed in order for the Dirac procedure to be applied in a strict fashion. However, the same results yielded by this method could be obtained by imposing $|\vec{m}|^2 - 1 = 0$ as a second class constraint which does not arise from the form of the Lagrangian.)

Thus, there are four primary constraints:

$$\varphi_0 \equiv P = 0 \quad , \tag{2.3.3}$$

$$\theta_i \equiv p_i - \frac{1}{3\gamma} \sum_J \frac{(m_e\ S_{eJ})\,\epsilon_{ijk}\ S_{jJ}\ m_k}{|\vec{m}|^2 - (m_a\ S_{aJ})^2} = 0, i = 1,2,3. \tag{2.3.4}$$

(Here we have anticipated that constraint Eq. (2.3.3) is of the first class type, while constraints Eq. (2.3.4) are of the second class type.)

The Hamiltonian density for the system is given through Eqs. (2.1.2), (2.1.3), (2.2.6) and (2.3.1) as

$$\hat{H}' = \hat{H} + \sum_{j=1}^{3} v_j\ \theta_j + v_0\ \varphi_0$$

$$= \dot{m}_i p_i + \dot{\lambda} P - \hat{L} + \sum_{j=1}^{3} v_j \theta_j + v_0 \varphi_0$$

$$= -\vec{m} \cdot \vec{B} + V - \lambda[|\vec{m}|^2 - 1] + \dot{\lambda} P + \sum_{j=1}^{3} v_j \theta_j + v_0 \varphi_0 \quad . \tag{2.3.5}$$

Note that because $\dot{\vec{m}}$ entered only linearly in $\hat{L}$, $\hat{H}$ is independent of $\vec{p}$. The consistency condition Eq. (2.2.9) combined with Eq. (2.3.5) yields

$$\dot{\varphi}_0 = \{\varphi_0, \int \hat{H}' d^3\vec{z}\} = \{ P, \int \hat{H}' d^3z \} = |\vec{m}|^2 - 1 = 0 \quad , \tag{2.3.6}$$

since P has vanishing Poisson brackets with everything except λ. (Note: the constraints may be imposed <u>after</u> the Poisson bracket has been calculated; thus, $\{P,\dot{\lambda}\}P = 0$.) A secondary constraint has therefore been generated, and is given by

$$\theta_4 = |\vec{m}|^2 - 1 = 0 \quad . \tag{2.3.7}$$

(Again we have anticipated that this is a second class constraint.)

Supplementing the Hamiltonian density Eq. (2.3.5) with the new constraint θ_4 and again invoking the consistency condition Eq. (2.2.9), we obtain

$$\dot{\theta}_i = \{\theta_i, \int \hat{H} d^3\vec{z}\} + \sum_{j=1}^{4} v_j \{\theta_i, \int \theta_j d^3\vec{z}\} = 0 \quad . \tag{2.3.8}$$

For i = 1,2,3, it should be noted that

$$\{\theta_i, \int \hat{H}\, d^3\vec{z}\} = B_i^{eff} + 2\lambda m_i \ , \qquad (2.3.9)$$

which follows from consideration of Eqs. (2.1.10), (2.2.24), (2.3.4) and (2.3.5). Moreover,

$$\{\theta_4, \int \hat{H}\, d^3\vec{z}\} = 0 \qquad (2.3.10)$$

follows from the fact that $\hat{H}$ is independent of the conjugate momenta p_i.

Equation (2.3.8) yields four simultaneous equations which may be solved for the unknown multipliers v_1, v_2, v_3 and v_4. These equations may be written in the form

$$B_i^{eff} + 2\lambda m_i + \sum_{j=1}^{4} v_j\, \theta_{ij} = 0, \ i = 1,2,3 \qquad (2.3.11)$$

$$\sum_{j=1}^{4} v_j\, \theta_{4j} = 0 \quad , \qquad (2.3.12)$$

where the matrix θ_{ij} has been defined as

$$\theta_{ij} \equiv \{\theta_i, \int \theta_j\, d^3\vec{z}\} \quad . \qquad (2.3.13)$$

We have exhausted all possibilities of generating any new constraints, and can now identify, as we had anticipated, the constraints θ_i, i = 1,2,3,4 as second class since they do not have vanishing brackets among themselves. On the other hand the constraint φ_0 of Eq. (2.3.3) is of first class type since its bracket with all the other constraints vanishes, and we are free to choose the "gauge" $\lambda = 0$ without changing the dynamics. Hereafter, we will completely drop all reference to λ and its conjugate momentum P as these variables will not contribute anything to the physics.

Now the equation of motion for $\dot{m}_i$ is given by

$$\dot{m}_i = \{m_i, \int \hat{H}\, d^3\vec{z}\} + \sum_{j=1}^{4} v_j \{m_i, \int \theta_j\, d^3\vec{z}\} = v_i \quad , \qquad (2.3.14)$$

where the first term on the right hand side vanishes because $\hat{H}$ is independent of the p's. The unknown multipliers v_1, v_2, v_3 and v_4 are determined by Eqs. (2.3.11) and (2.3.12). This calculation is presented in Appendix A. The results of (A.6) yield the precession equation

$$\dot{\vec{m}} = \gamma \vec{m} \times \vec{B}^{eff} \quad , \qquad (2.3.15)$$

in agreement with the result of the Lagrangian formalism.

We now turn to a calculation of the Dirac brackets of the magnetization components. Equations (2.2.24) and (2.2.25), (2.3.4) and (2.3.7) yield the Dirac bracket

$$\{m_i(\vec{z}), m_j(\vec{z}')\}^* = + \iint d\vec{x}\, d\vec{y}\, \delta_{ia}\, \delta^3(\vec{z} - \vec{x})\, C_{ab}(\vec{x},\vec{y})\, \delta_{bj}\, \delta^3(\vec{y} - \vec{z}')$$

$$= C_{ij}(\vec{z},\vec{z}') \quad (i,j = 1,2,3) \quad . \tag{2.3.16}$$

The calculation of the matrix C_{ij} proceeds by noting that

$$\theta_{ij}(\vec{z}) \equiv \{\theta_i(\vec{z}), \int \theta_j(\vec{z}')\, d\vec{z}'\}$$

$$= \int d\vec{z}'\{\theta_i(\vec{z}), \theta_j(\vec{z}')\} \quad . \tag{2.3.17}$$

Thus, the bracket between two second class constraints is expressible in terms of the previously defined matrix $\theta_{ij}(\vec{z})$ of Eq. (2.3.13):

$$\{\theta_i(\vec{z}), \theta_j(\vec{z}')\} = \theta_{ij}(\vec{z})\ \delta^3(\vec{z}-\vec{z}') \quad . \tag{2.3.18}$$

The integral equation defining the matrix C_{ij}, Eq. (2.2.27), is now expressible with the aid of Eq. (2.3.18) as

$$\int d\vec{y}\ C_{ij}(\vec{x},\vec{y})\ \theta_{jk}(\vec{y}) = \delta_{ik} \quad . \tag{2.3.19}$$

By inspection, the solution for the matrix C_{ij} is

$$C_{ij}(\vec{x},\vec{y}) = \theta^{-1}_{ij}(\vec{x})\ \delta^3(\vec{x} - \vec{y}) \quad . \tag{2.3.20}$$

The matrix inverse, θ^{-1}_{ij}, has been calculated in Appendix A and appears as Eqs. (A.7) - (A.10). From this and Eqs.

(2.3.16) and (2.3.20), the Dirac brackets are given by

$$\{m_i(\vec{z},t), m_j(\vec{z}',t)\}^* = \gamma\ \epsilon_{ijk}\ m_k(\vec{z},t)\ \delta^3(\vec{z}-\vec{z}')\ . \qquad (2.3.21)$$

The passage to quantum theory could now be made by replacing the above Dirac brackets by $-i$ times commutators. The second class constraints can be strongly (as operators) set equal to zero, and the Hamiltonian to be quantized would then be given by

$$H = -\vec{m}\cdot\vec{B} + V\ . \qquad (2.3.22)$$

This Hamiltonian is essentially the well known starting point for spin wave or magnon theory. The usual precession equation, (2.3.15), follows from Eq. (2.2.20) when the Dirac brackets, Eq. (2.3.21), are used with the Hamiltonian of Eq. (2.3.22). The preceeding derivation, Eq. (2.3.14), obtained the same result without explicit use of the Dirac bracket formalism.

The application of the above theory to the case of a single particle possessing spin and translational degrees of freedoms is simple and straightforward. The application to an elastic magnetic dielectric having more than one particle per unit cell will appear in Chapter III.

III. THE ELASTIC MAGNETIC CONTINUUM

3.1 The Microscopic Lagrangian

We consider a crystal as a mechanical system consisting of a set of point particles situated in a vacuum. Each particle is located at the position $\vec{x}^{n\alpha}$, and has fixed charge e^{α}, fixed mass m^{α} and may have a permanent and, therefore, spontaneous magnetic dipole moment $\vec{\mu}^{n\alpha}$ which arises from the intrinsic spin and electron orbital motion. The notation follows that of ref. 27. The index n has three integer components that name the primitive cell n, and α labels the type of particle or sublattice. The particles are acted upon by mechanical forces (of quantum mechanical origin) derivable from a potential energy $V(\{\vec{x}^{n\alpha},\vec{\mu}^{n\alpha}\})$ where $\{\vec{x}^{n\alpha},\vec{\mu}^{n\alpha}\}$ denotes the set of all particle positions and their respective spin magnetic moments. The particles are also subject to forces of electromagnetic origin which arise from electric and magnetic fields, either externally applied or produced by the charges and magnetic dipoles themselves.

The total Lagrangian of the system has three parts

$$L = L_P + L_F + L_I \quad , \tag{3.1.1}$$

which consists of the nonrelativistic particle Lagrangian having the form

$$L_P = \frac{1}{2} \sum_{n\alpha} m^{\alpha} (\dot{\vec{x}}^{n\alpha})^2 - V(\{\vec{x}^{n\alpha}, \vec{\mu}^{n\alpha}\}) \quad , \tag{3.1.2}$$

the field Lagrangian expressed in rationalized MKS units of the form[73]

$$L_F = \int \hat{L}_F \, d\vec{z} = \frac{1}{2} \epsilon_0 \int [\vec{E}^2(\vec{z},t) - c^2 \vec{B}^2(\vec{z},t)] \, d\vec{z} \; , \tag{3.1.3}$$

and the electromagnetic field-particle interaction Lagrangian[73] of the form

$$L_I = \int \hat{L}_{FI} \, d\vec{z} = \sum_{n\alpha} e^{\alpha} [\dot{\vec{x}}^{n\alpha}(t) \cdot \vec{A}(\vec{x}^{n\alpha}(t),t) - \Phi(\vec{x}^{n\alpha}(t),t)]$$

$$+ \sum_{n\alpha} \vec{\mu}^{n\alpha}(t) \cdot \vec{B}(\vec{x}^{n\alpha}(t),t) \quad . \tag{3.1.4}$$

Here $d\vec{z} = dz_1 \, dz_2 \, dz_3$ is a volume element in the laboratory coordinate system, and the electric and magnetic field vectors $\vec{E}$ and $\vec{B}$ are defined in terms of the scalar and vector potentials Φ and $\vec{A}$ by means of

$$\vec{E}(\vec{z},t) = -\nabla \, \Phi(\vec{z},t) - \frac{\partial \vec{A}(\vec{z},t)}{\partial t} \quad , \tag{3.1.5}$$

$$\vec{B}(\vec{z},t) = \nabla \times \vec{A}(\vec{z},t) \quad . \tag{3.1.6}$$

3.2 The Continuum Limit and the Macroscopic Lagrangian

Since we will be interested in the interaction of visible light or of long-wavelength electromagnetic radiation, ultrasonic waves, and spin waves, that is, phenomena with wavelength much larger than the dimension of the primitive cell, the theory can be a long wavelength or macroscopic theory. The passage to the continuum limit is accomplished by replacing the index n, which names the cell, with a continuous variable $\vec{X}$ while retaining the particle type index α as a sublattice index. The variable $\vec{X}$ is called the material coordinate because it rides with the mass point. Thus, we make the replacement

$$\vec{x}^{n\alpha}(t) \to \vec{x}^{\alpha}(\vec{X},t) \quad , \tag{3.2.1}$$

$$\vec{\mu}^{n\alpha}(t) \to \vec{\mu}^{\alpha}(\vec{X},t) \quad , \tag{3.2.2}$$

of a set of discrete positions and magnetic dipoles by a set of functions of the continuous variable $\vec{X}$. x_i^α represents the i-th component of the position of sublattice α expressed in a Cartesian frame called the spatial frame, and X_A represents the A-th Cartesian component of the name $\vec{X}$ in what is called the material frame. Upper case Latin letters will denote components in the material coordinate system and lower case Latin letters will denote components in the spatial system. This notation follows that of Truesdell.[74] In the continuum limit cell sums become integrals over the range of the material coordinate:

$$\sum_n F(\vec{x}^{n\alpha}(t)) \to \frac{1}{\Omega_0} \int F(\vec{x}^{\alpha}(\vec{X},t))\, d\vec{X} \quad , \tag{3.2.3}$$

where Ω_0 is the undeformed volume of the primitive unit cell.

A center of mass position is defined by

$$\vec{x}(\vec{X},t) = \sum_{\alpha=1}^{N} \rho^{\alpha}\, \vec{x}^{\alpha}(\vec{X},t)/\rho^{0} \quad , \tag{3.2.4}$$

$$\rho^{0} \equiv \sum_{\alpha=1}^{N} \rho^{\alpha} \equiv \sum_{\alpha=1}^{N} m^{\alpha}/\Omega_0 \quad , \tag{3.2.5}$$

and a set $\alpha=1,\ldots,N$ of internal displacements[75] is introduced

$$\vec{u}^{\alpha}(\vec{X},t) \equiv \vec{x}^{\alpha}(\vec{X},t) - \vec{x}(\vec{X},t) \quad . \tag{3.2.6}$$

The distinction between the material coordinate $\vec{X}$ of the center of mass and the spatial coordinate $\vec{x}$ of the center of mass will be maintained throughout this chapter. In calculations, however, it will be convenient to make the spatial and material frames identical Cartesian ones. The material components X_A of the center of mass may then be regarded as equal to the center of mass position components x_a when the body is in its natural state, that is, a homogeneous, time independent state free from applied external fields or stress. Moreover, it will be assumed that there has been no initial plastic deformation so that both the stress and the strain will be zero in the natural state in a nonferroelastic material.

It will be more convenient to transform the $\vec{x}^{\alpha}$ coordinates to a new set of position coordinates consisting of the center of mass position $\vec{x}(\vec{X},t)$ and internal coordinates $\vec{y}^{\mu}(\vec{X},t)$ $(\mu=1,\ldots,N-1)$ which are displacement invariant. The transformation is defined by

$$\vec{y}^{\mu}(\vec{X},t) \equiv \sum_{\alpha=1}^{N} U^{\mu\alpha}\,\vec{x}^{\alpha}(\vec{X},t) \quad , \tag{3.2.7}$$

$$\vec{x}^{\alpha}(\vec{X},t) \equiv \sum_{\mu=0}^{N-1} V^{\alpha\mu}\,\vec{y}^{\mu}(\vec{X},t) \quad , \tag{3.2.8}$$

where

$$\sum_{\alpha=1}^{N} U^{\mu\alpha}\,V^{\alpha\nu} = \delta^{\mu\nu} \quad , \tag{3.2.9}$$

$$\sum_{\mu=0}^{N-1} V^{\alpha\mu}\,U^{\mu\beta} = \delta^{\alpha\beta} \quad , \tag{3.2.10}$$

$$\vec{y}^{0}(\vec{X},t) \equiv \vec{x}(\vec{X},t) \quad . \tag{3.2.11}$$

Displacement invariance of $\vec{y}^{\mu}(\mu\neq 0)$ and its lack for $\mu=0$

require

$$\sum_{\alpha=1}^{N} U^{\mu\alpha} = \delta^{\mu 0} \quad . \tag{3.2.12}$$

An alternative statement of displacement invariance derived from Eq. (3.2.10) would be

$$V^{\alpha 0} = 1 \quad . \tag{3.2.13}$$

Equations (3.2.4), (3.2.7) and (3.2.11) together imply

$$U^{0\alpha} = \rho^{\alpha}/\rho^{0} \quad . \tag{3.2.14}$$

Substitutions of Eq. (3.2.14) into Eq. (3.2.9) yields

$$\sum_{\alpha=1}^{N} \rho^{\alpha} V^{\alpha\nu}/\rho^{0} = \delta^{0\nu} \quad . \tag{3.2.15}$$

The diagonality of the kinetic energy can be retained by requiring

$$\sum_{\alpha=1}^{N} \rho^{\alpha} V^{\alpha\mu} V^{\alpha\nu} = m^{\mu} \delta^{\mu\nu} , \; m^{0} \equiv \rho^{0} \quad . \tag{3.2.16}$$

Then we have

$$\frac{1}{2} \sum_{\alpha=1}^{N} \rho^{\alpha} [\dot{x}^{\alpha}]^{2} = \frac{1}{2} \sum_{\mu=0}^{N-1} m^{\mu} [\dot{y}^{\mu}]^{2} \quad . \tag{3.2.17}$$

If we multiply Eq. (3.2.16) by $U^{\nu\beta}$, sum over ν, and use Eq. (3.2.10), we obtain a connection between the transformation and its inverse:

$$\rho^{\beta} V^{\beta\mu} = m^{\mu} U^{\mu\beta} \quad . \qquad (3.2.18)$$

The time derivatives occurring in Eq. (3.2.17), taken holding the material coordinate $\vec{X}$ fixed, are called material time derivatives. This is in contrast to the spatial frame time derivative where the spatial frame coordinate $\vec{z}$ (or $\vec{x}$ if in matter) is held fixed. A material time derivative is related to its corresponding spatial frame time derivative by

$$\frac{dF(\vec{X},t)}{dt} = \dot{F} = \frac{\partial\hat{F}(\vec{x},t)}{\partial t} + \dot{x}_i \frac{\partial\hat{F}(\vec{x},t)}{\partial x_i} \quad . \qquad (3.2.19)$$

The total continuum Lagrangian can be expressed in terms of a Lagrangian density referred to either the spatial coordinate system or the material coordinate system:

$$L = \int \hat{\mathcal{L}}^S \, dv = \int \hat{\mathcal{L}}^M \, dV \quad . \qquad (3.2.20)$$

Here dv and dV are volume elements in the spatial and material frames respectively. As previously noted in Section 3.1, the total Lagrangian density consists of the sum of three Lagrangian densities describing the field, the field-matter interaction and the matter:

$$\hat{L}^j = \hat{L}_F^j + \hat{L}_I^j + \hat{L}_M^j \;, (j = S,M) \quad . \qquad (3.2.21)$$

The field Lagrangian in the spatial frame has already been noted in Eq. (3.1.3). Using Eqs. (3.1.4), (3.2.2) and (3.2.3), the continuum interaction Lagrangian is given by

$$\hat{L}_I^M = \sum_\alpha q^\alpha \, [\dot{\vec{x}}^\alpha(\vec{X},t) \cdot \vec{A}(\vec{x}^\alpha(\vec{X},t),t) - \Phi(\vec{x}^\alpha(\vec{X},t),t)]$$

$$+ \sum_\alpha \vec{m}^{T\alpha}(\vec{X},t) \cdot \vec{B}(\vec{x}^\alpha(\vec{X},t)t) \quad , \qquad (3.2.22)$$

or

$$\hat{L}_I^S = \vec{j}(\vec{z},t) \cdot \vec{A}(\vec{z},t) - q(\vec{z},t) \; \Phi(\vec{z},t) + \vec{m}(\vec{z},t) \cdot \vec{B}(\vec{z},t) \quad , \qquad (3.2.23)$$

where the charge and current densities associated with the ionic motion are given by

$$q(\vec{z},t) = \sum_\alpha q^\alpha \int \delta(\vec{z} - \vec{x}^\alpha(\vec{X},t)) \; dV \; , \qquad (3.2.24)$$

$$\vec{j}(\vec{z},t) = \sum_\alpha q^\alpha \int \dot{\vec{x}}^\alpha(\vec{X},t) \; \delta(\vec{z} - \vec{x}^\alpha(\vec{X},t)) \; dV \; , \qquad (3.2.25)$$

and the spin and electronic contribution to the magnetization is given by

$$\vec{m}(\vec{z},t) = \int \sum_{\alpha} \vec{m}^{T\alpha}(\vec{X},t)\ \delta(\vec{z} - \vec{x}^{\alpha}(\vec{X},t))\ dV \ , \qquad (3.2.26)$$

where

$$q^{\alpha} \equiv e^{\alpha}/\Omega_0 \ , \qquad (3.2.27)$$

and

$$\vec{m}^{T\alpha} \equiv \vec{\mu}^{\alpha}(\vec{X},t)/\Omega_0 \qquad (3.2.28)$$

is the effective magnetic moment density of sublattice α from spin and orbital contributions. The superscript T on $\vec{m}^{T\alpha}$, standing for total, is used to indicate that the sublattice magnetizations may possess a constant or spontaneous part $\vec{m}^{S\alpha}$ in addition to a part $\vec{m}^{\alpha}$ which may vary because of some external influence.

In either of the forms (3.2.22) and (3.2.23), the interaction Lagrangian contains multipoles of all order. We will specialize to the case of the magnetic dielectric material in which the free charge is zero:

$$\sum_{\alpha} q^{\alpha} = 0 \ . \qquad (3.2.29)$$

The ionic magnetic dipole term will be retained but electric quadrupole, and higher order terms will be dropped. This is accomplished by expanding the functions of $\vec{x}^{\alpha}$ about $\vec{x}$ using

Eq. (3.2.6) and retaining all terms to first order in the small quantities $\{\vec{u}^{\alpha}\}$ or their equivalent $\{\vec{y}^{\mu}\}$ ($\mu \neq 0$). We, moreover, use the fact that a total time (or space) derivative can be discarded from the Lagrangian since it cannot affect the equations of motion, and find that the interaction Lagrangian to dipole order is given in the material frame by

$$\hat{L}_I^M = \sum_{\mu}{}' q^{\mu}\, \vec{y}^{\mu}(\vec{X},t) \cdot \hat{E}(\vec{x}(\vec{X},t),t)$$

$$+ \frac{1}{2} \sum_{\mu,\nu}{}' q^{\mu\nu}(\vec{y}^{\mu} \times \dot{\vec{y}}^{\nu}) \cdot \vec{B}(\vec{x}(\vec{X},t),t)$$

$$+ \sum_{\alpha} \vec{m}^{T\alpha}(\vec{X},t) \cdot \vec{B}(\vec{x}(\vec{X},t),t)$$

$$+ \sum_{\mu}{}' \vec{m}^{T\mu}(\vec{X},t) \cdot (\vec{y}^{\mu}(\vec{X},t) \cdot \nabla)\, \vec{B}(\vec{x}(\vec{X},t),t) \quad , \tag{3.2.30}$$

while in the spatial frame we find

$$\hat{L}_I^S = \sum_{\mu}{}' q^{\mu} [\vec{y}^{\mu}(\vec{X},t)/J(\vec{X},t)]_{\vec{z}=\vec{x}(\vec{X},t)} \cdot \hat{E}(\vec{z},t)$$

$$+ \vec{M}(\vec{z},t) \cdot \vec{B}(\vec{z},t) + \underline{N} : \nabla\, \vec{B}(\vec{z},t) \quad , \tag{3.2.31}$$

where

$$q^{\mu} \equiv \sum_{\alpha} q^{\alpha}\, v^{\alpha\mu} \;, \; q^{\mu\nu} \equiv \sum_{\alpha} q^{\alpha}\, v^{\alpha\mu}\, v^{\alpha\nu} \;, \tag{3.2.32}$$

$$\hat{E} \equiv \vec{E} + \dot{\vec{x}} \times \vec{B} \quad , \tag{3.2.33}$$

$$\vec{m}^{T\mu} \equiv \sum_{\alpha} \vec{m}^{T\alpha} V^{\alpha\mu} \quad , \tag{3.2.34}$$

$$\vec{M}(\vec{z},t) \equiv [\{\sum_{\alpha} \vec{m}^{T\alpha}(\vec{X},t)$$

$$+ \frac{1}{2} \sum_{\mu,\nu}{}' q^{\mu\nu}(\vec{y}^{\mu} \times \dot{\vec{y}}^{\nu})\} \,/J(\vec{X},t)]_{\vec{z}=\vec{x}(\vec{X},t)} \quad , \tag{3.2.35}$$

$$\underline{N}(\vec{z},t) \equiv \sum_{\alpha}[\vec{m}^{T\alpha}(\vec{X},t)\ \vec{u}^{\alpha}(\vec{X},t)/J(\vec{X},t)]_{\vec{z}\,=\,\vec{x}(\vec{X},t)}$$

$$= \sum_{\mu}{}' \ [\vec{m}^{T\mu}(\vec{X},t)\ \vec{y}^{\mu}(\vec{X},t)/J(\vec{X},t)]_{\vec{z}\,=\,\vec{x}(\vec{X},t)} \quad . \tag{3.2.36}$$

The expression for q^{μ}, the effective charge associated with the μ-th degree of freedom and neutrality of the unit cell as given by Eq. (3.2.29) implies that $q^{0} = 0$. A primed sum indicates in the above equations that the $\mu = 0$ term is to be omitted. The Jacobian of the transformation from the $\vec{X}$ frame to the $\vec{x}$ frame is

$$J(\vec{X},t) \equiv \det \partial x_i/\partial X_A \equiv \det x_{i,A} \quad , \tag{3.2.37}$$

where the comma denotes differentiation in the last expression. Note that in a magnetic material the dipole approximation requires the retention of the spin quadrupole moment $\underline{N}$. The : notation in Eq. (3.2.31) indicates a contraction on two indices.

The matter Lagrangian consists of the kinetic energy minus the stored energy. With the aid of Eqs. (3.2.11), (3.2.16) and (3.2.17) this becomes

$$\hat{L}_M^M = (\rho^0/2)(\dot{\vec{x}})^2 + \sum_{\mu}{}' (m^\mu/2)\ (\dot{\vec{y}}^\mu)^2 - \rho^0 \Sigma \ , \qquad (3.2.38)$$

where Σ is the stored energy per unit undeformed mass of the crystal. The exact form that Σ may take will be discussed in Section 3.3.

The Lagrange equations can be written in either the spatial or material frame depending on which Lagrangian density in Eq. (3.2.21) is used. When second order space derivatives are present in the Lagrangian density, the Lagrange equation of motion in the spatial frame for a generalized coordinate q_i is

$$\frac{\partial}{\partial t}\frac{\partial \hat{L}^S}{\partial(\partial q_i/\partial t)} = \frac{\partial \hat{L}^S}{\partial q_i} - \frac{\partial}{\partial z_j}\frac{\partial \hat{L}^S}{\partial q_{i,j}} + \frac{\partial^2}{\partial z_j \partial z_k}\frac{\partial \hat{L}^S}{\partial q_{i,jk}} \ , \qquad (3.2.39)$$

where $\vec{z}$ is the independent variable. In the material frame the Lagrangian equation for the generalized coordinate q_i is

$$\frac{d}{dt}\frac{\partial \hat{L}^M}{\partial \dot{q}_i} = \frac{\partial \hat{L}^M}{\partial q_i} - \frac{\partial}{\partial X_A}\frac{\partial \hat{L}^M}{\partial q_{i,A}} + \frac{\partial^2}{\partial X_A \partial X_B}\frac{\partial \hat{L}^M}{\partial q_{i,AB}} \ , \qquad (3.2.40)$$

where $\vec{X}$ is the independent variable.

3.3 The Stored Energy

The form that Σ may take consistent with the conservation laws has been discussed in detail previously for dielectric and pyroelectric materials.[76] These same arguments hold for the case of an elastic magnetic dielectric along with the additional symmetry of invariance of the stored energy under time reversal. We summarize these arguments here.

In general Σ could be a function of all the degrees of freedom, $\vec{x}$, $\vec{y}^{\mu}(\mu=1,\ldots,N-1)$, $\vec{m}^{T\alpha}(\alpha=1,\ldots,N)$, their first and higher derivatives with respect to $\vec{X}$ and $\vec{X}$ itself. Since in the long wavelength limit, derivatives of higher order than the first make progressively smaller contributions to the energy, these will be neglected. Moreover, we will omit wave vector dispersion effects, such as optical or acoustic activity,[77] which are lowest order corrections to the long wavelength limit. This requires that we exclude the dependence of Σ upon the derivatives $\vec{y}^{\mu}_{,A} \equiv \partial\vec{y}^{\mu}/\partial X_A$ $(\mu=1,\ldots,N-1)$. We will retain derivatives of the magnetization $\vec{m}^{T\alpha}_{,A}$. The following considerations will also restrict the choice, the construction, and the combination of the relevant variables upon which Σ will depend:

A. Invariance Requirements

(1) Momentum conservation requires invariance of $\sum$ with respect to uniform displacements in the spatial coordinate system. Since all $\vec{y}^{\mu}(\mu=1,\ldots,N-1)$ and the first derivative of $\vec{x}$ possess such invariance, $\sum$ may be a function of these variables. The center of mass position $\vec{x}$, however, does not possess such invariance, and so $\sum$ may not be a function of $\vec{x}$.

(2) Angular momentum conservation requires invariance of $\sum$ with respect to uniform rotations in the spatial coordinate system. Such invariance is guaranteed if each of the independent variables of $\sum$ is individually rotationally invariant. A complete set of such variables consist of E_{AB}, $\Lambda^{\mu}_{A}(\mu=1,\ldots,N-1)$, $\Gamma^{T\alpha}_{A}, \Gamma^{\alpha}_{A;B}(\alpha=1,\ldots,N)$, X_A and $\mathrm{sgn}(J)$. The Green finite strain tensor E_{AB} is defined by[78]

$$E_{AB} \equiv (C_{AB} - \delta_{AB})/2 \quad , \tag{3.3.1}$$

$$C_{AB} \equiv x_{i,A}\, x_{i,B} \quad . \tag{3.3.2}$$

Summation over repeated indices is implied. The rotationally invariant internal coordinates Λ^{μ}_{A} are defined by

$$\Lambda^{\mu}_{A} \equiv y^{\mu}_{i}\, R_{iA} \ (\mu=1,\ldots,N-1) \quad , \tag{3.3.3}$$

where R_{iA} is the finite rotation tensor[79] given by

$$R_{iA} \equiv x_{i,B}(C^{-1/2})_{BA} \quad . \tag{3.3.4}$$

The rotationally invariant magnetization and magnetization gradient variables, $\Gamma_A^{T\alpha}$ and $\Gamma_{A;B}^{\alpha}$, are defined by

$$\Gamma_A^{T\alpha} \equiv m_i^{T\alpha} R_{iA} \quad , \tag{3.3.5}$$

$$\Gamma_{A;B}^{\alpha} \equiv m_{i,B}^{\alpha} R_{iA} \quad . \tag{3.3.6}$$

The sgn(J) variable enters the discussion in the following manner. The stored energy depends on the vector variables $\vec{x}_{,1}, \vec{x}_{,2}$, and $\vec{x}_{,3}$. Rotational invariants were formed from these giving us the variables E_{AB}. However, another rotational invariant could be formed by taking the triple product or the determinant of the components of the three vectors $\vec{x}_{,1}, \vec{x}_{,2}$ and $\vec{x}_{,3}$. This invariant is just the Jacobian J of the transformation as defined by Eq. (3.2.37). It is easy to show using the rules of determinantal multiplication that

$$J^2 = \det C_{AB} \quad , \tag{3.3.7}$$

so that J is expressible up to a ± sign in terms of the finite strain tensor E_{AB}. Since J is a pseudoscalar, sgn(J) is ±1 in right and left handed coordinate systems respectively. [The reader is reminded that the parity operator which changes the "handedness" of the coordinate system is quite different from the material inversion operator which inverts the body. That is, parity conservation is a completely distinct question not associated with any material inversion symmetry that a crystal might possess.] It is therefore possible to separate the stored energy into a part invariant under a parity operation, and a part which is not:

$$\Sigma = \Sigma_1 + [\, \mathrm{sgn}(J) \,] \, \Sigma_2 \quad , \qquad (3.3.8)$$

where Σ_1 and Σ_2 are both invariant under a parity transformation of the coordinate system.

(3) Although it is well known that parity is not precisely conserved, we will neglect the small violations of parity conservation for our case. Σ must now be invariant with respect to inversion of the spatial coordinate system. This necessitates the dropping of the small parity non-conserving term $[\, \mathrm{sgn}(J) \,] \, \Sigma_2$ from Eq. (3.3.8). We note, however, that up to this point Σ_1 in Eq. (3.3.8) may contain terms which have a product of sgn(J) with odd power combinations of $\Gamma_A^{T\alpha}$ and $\Gamma_{A;B}^{\alpha}$. The latter magnetic invariants are odd under a parity transformation because of the presence of the finite rotation tensor defined by Eq. (3.3.4).

(4) Σ must be invariant under the operation of time reversal. This requires that the magnetic variables $\Gamma_A^{T\alpha}$ and $\Gamma_{A;B}^{\alpha}$ occur in even power combinations in the power series for Σ. Consideration of this and the statements made concerning parity conservation make it possible to drop sgn(J) from the set of variables for Σ.

(5) We consider only homogeneous crystals, which in the continuum limit requires invariance of $\sum$ with respect to arbitrary translations of the material coordinate system. Thus, X_A must be removed from the set of variables for $\sum$. In the language of conservation laws, homogeneity is equivalent to crystal momentum conservation.

(6) $\sum$ must be invariant under the operations of the space group that describes the symmetry of the crystal under consideration. This will place restrictions on the expansion coefficients that appear in the power series in the relevant variables to be written down shortly.

From the above considerations the functional dependence of $\sum$ may be stated as

$$\sum = \sum (E_{AB}, \Lambda^{\mu}_{A}, \Gamma^{T\alpha}_{A}, \Gamma^{\alpha}_{A;B}) \quad , \qquad (3.3.9)$$

where $\sum$ is understood to be an even function of the magnetic variables $\Gamma^{T\alpha}_{A}$ and $\Gamma^{\alpha}_{A;B}$.

B. Magnetic Space Group and Crystal Symmetry

A magnetic space group is formed from an ordinary space group in the following fashion:[80] Let G represent an ordinary space group. Choose any subgroup H of index 2 in G (by index 2 is meant the order or number of elements in G is twice the order of the subgroup H). Multiply the elements of G-H by the time reversal operation K. Then

$G' = H + K(G-H)$ is a magnetic space group.

The displacements $\{\vec{u}^{\alpha}\}$ are even under time reversal while the magnetizations $\{\vec{m}^{T\alpha}\}$ are odd. The opposite is true under the action of the material inversion operation which reverses all polar vectors but leaves invariant all axial or pseudo vectors. Under a magnetic space group operation S which contains a rotational part represented by the rotation matrix $\underline{S}$, a particle of type α at position $\vec{X}$ is carried by S into a particle of type $S(\alpha)$ at position $\underline{S} \cdot \vec{X} + \vec{v}(S)$. Here $\vec{v}(S)$ is the translational part of the magnetic space group operation S. Thus, the new displacement of a particle of type α at $\vec{X}$ is given by[81]

$$[\, u_i^{\alpha}(\vec{X},t) \,]' = S_{ij} \, u_j^{S^{-1}(\alpha)} \, (\underline{S}^{-1} \cdot \vec{X} - \underline{S}^{-1} \cdot \vec{v},t) \,. \quad (3.3.10)$$

Here S_{ij} is the three dimensional representation of the rotation-reflection part of the magnetic space group element S. This representation of S is understood to be that which is odd under the material inversion operator. The new magnetization of a particle of type α at $\vec{X}$ is given by

$$[\, m_i^{\alpha}(\vec{X},t) \,]' = I(S) \, K(S) \, S_{ij} \, m_j^{S^{-1}(\alpha)} \, (\underline{S}^{-1} \cdot \vec{X} - \underline{S}^{-1} \cdot \vec{v},t) \,. \quad (3.3.11)$$

$I(S) = \pm 1$ according to whether the rotational part of the operation S is proper or improper. $K(S) = \pm 1$ according to whether the operator S is unitary or anti-unitary. That is, K(S) = + 1 if S does not contain the time reversal operator, and −1 if it does. For the sake of clarity we have suppressed the total symbol T over the magnetization in Eq. (3.3.11). By consideration of Eqs. (3.2.7), (3.2.11), (3.2.12), (3.3.1) −(3.3.6), (3.3.10) and (3.3.11) we obtain the transformation properties of the invariants in the form

$$[\Lambda^{\mu}_{A}(\vec{X},t)]' = S_{AB}\ U^{\mu\alpha}\ V^{S^{-1}(\alpha)\nu}\Lambda^{\nu}_{B}(\underline{S}^{-1}\cdot\vec{X}-\underline{S}^{-1}\cdot\vec{v},t)\ , \tag{3.3.12a}$$

$$[E_{AB}(\vec{X},t)]' = S_{AC}\ S_{BD}\ E_{CD}(\underline{S}^{-1}\cdot\vec{X}-\underline{S}^{-1}\cdot\vec{v},t)\ , \tag{3.3.12b}$$

$$[\Gamma^{\alpha}_{A}(\vec{X},t)]' = K(S)\ I(S)\ S_{AB}\ \Gamma^{S^{-1}(\alpha)}_{B}(\underline{S}^{-1}\cdot\vec{X}-\underline{S}^{-1}\cdot\vec{v},t)\ , \tag{3.3.12c}$$

$$[\Gamma^{\alpha}_{A;B}(\vec{X},t)]' = K(S)\ I(S)\ S_{AC}\ S_{BD}\ \Gamma^{S^{-1}(\alpha)}_{C;D}(\underline{S}^{-1}\cdot\vec{X}-\underline{S}^{-1}\cdot\vec{v},t)\ . \tag{3.3.12d}$$

The quantity to be invariant under the magnetic space group operation is the total stored energy integrated over the crystal. That is,

$$\int \Sigma\, d\vec{X} = \int \Sigma'\, d\vec{X} = \int \Sigma'\, d(\underline{S}^{-1}\cdot\vec{X}-\underline{S}^{-1}\cdot\vec{v})\quad , \tag{3.3.13}$$

where Σ' is the stored energy written as a function of the primed variables given in Eqs. (3.3.12). Comparing integrands in Eq. (3.3.13), we obtain the invariance requirement

$$\sum(\Lambda^{\mu}_{A},\ E_{AB},\ \Gamma^{T\alpha}_{A},\ \Gamma^{\alpha}_{A;B})$$

$$= \sum(S_{AB}\ U^{\mu\alpha}\ V^{S^{-1}(\alpha)\nu}\Lambda^{\nu}_{B},\ S_{AC}\ S_{BD}\ E_{CD},$$

$$K(S)\ I(S)\ S_{AB}\ \Gamma^{S^{-1}(\alpha)}_{B},K(S)\ I(S)\ S_{AC}\ S_{BD}\ \Gamma^{S^{-1}(\alpha)}_{C;D}) \ . \qquad (3.3.14)$$

It was previously stated that time reversal invariance required the magnetizations and/or magetization gradients to occur only in even power combinations. Equation (3.3.14) may be written in the alternative form

$$\sum\ (\Lambda^{\mu}_{A},\ E_{AB},\ \Gamma^{T\alpha}_{A},\ \Gamma^{\alpha}_{A;B})$$

$$= \sum\ (S_{AB}\ U^{\mu\alpha}\ V^{S^{-1}(\alpha)\nu}\ \Lambda^{\nu}_{B},\ S_{AC}\ S_{BD}\ E_{CD},$$

$$S_{AB}\ \Gamma^{S^{-1}(\alpha)}_{B},\ S_{AC}\ S_{BD}\ \Gamma^{S^{-1}(\alpha)}_{C;D}\ ) \ . \qquad (3.3.15)$$

In the above, it is understood that the magnetic invariants only appear in even power combinations.

For purposes of discussion we may expand the stored energy in a power series obtaining

$$\rho^{0}\ \sum = {}^{(1000)}H^{\mu}_{A}\ \Lambda^{\mu}_{A} + {}^{(0100)}H_{AB}\ E_{AB} + {}^{(2000)}H^{\mu\nu}_{AB}\ \Lambda^{\mu}_{A}\ \Lambda^{\nu}_{B}$$

$$+ {}^{(0200)}H_{ABCD}\ E_{AB}\ E_{CD} + {}^{(1100)}H^{\mu}_{ABC}\ \Lambda^{\mu}_{A}\ E_{BC}$$

$$+ {}^{(0020)}H^{\alpha\beta}_{AB}\ \Gamma^{T\alpha}_{A}\ \Gamma^{T\beta}_{B}$$

$$+ {}^{(0002)}H^{\alpha\beta}_{ABCD}\ \Gamma^{\alpha}_{A;B}\ \Gamma^{\beta}_{C;D} + {}^{(0011)}H^{\alpha\beta}_{ABC}\ \Gamma^{T\alpha}_{A}\ \Gamma^{\beta}_{B;C}$$

$$+ {}^{(1020)}H^{\mu\alpha\beta}_{ABC}\ \Lambda^{\mu}_{A}\ \Gamma^{T\alpha}_{B}\ \Gamma^{T\beta}_{C} + {}^{(0120)}H^{\alpha\beta}_{ABCD}\ E_{AB}\ \Gamma^{T\alpha}_{C}\ \Gamma^{T\beta}_{D}$$

$$+ {}^{(1011)}H^{\mu\alpha\beta}_{ABCD}\ \Lambda^{\mu}_{A}\ \Gamma^{T\alpha}_{B}\ \Gamma^{\beta}_{C;D} + \ldots \qquad (3.3.16)$$

The H coefficients are called material descriptors[82] since they describe intrinsic properties of the solid under study. The numerical values of these tensors can be calculated only from quantum mechanics. In the present treatment, we are not concerned with such calculations and regard the numerical values of these tensors as being determined by comparison with experiment. Moreover, it is important to realize that by their very nature, the material descriptors cannot be functions of any of the characteristics of any applied influence. Thus, for example, they are frequency and wave vector independent. The frequency dependence of various material tensors (e.g. the piezomagnetic tensor) will arise from the solution of the time dependent dynamical equations of motion which result from the Lagrangian.

In Eq. (3.3.16) the material descriptors contain numerical presuperscripts (k,l,m,n) where k denotes the number of polarization-like factors Λ^{μ}, l denotes the number of strain factors E, m denotes the number of magnetization factors, and n denotes the number of magnetization gradient factors in the term in question. As stated previously, the stored energy must be invariant under crystal group operations. Since the variables Λ^{μ}_{A}, E_{AB}, $\Gamma^{T\alpha}_{A}$ and $\Gamma^{\alpha}_{A;B}$ are altered by crystal group operations, conditions are imposed on the series coefficients, $^{(k,l,m,n)}H$ by Eq. (3.3.15).

Consider a particular term in the series Eq. (3.3.16) for which Eq. (3.3.15) leads to the requirement

$$\sum_{\mu,\alpha,\beta} H^{\mu\alpha\beta}_{ABC}\, \Lambda^{\mu}_{A}\, \Gamma^{T\alpha}_{B}\, \Gamma^{T\beta}_{C}$$

$$= \sum_{\substack{\mu,\alpha,\beta\\ \gamma,\nu,}} H^{\mu\alpha\beta}_{ABC}\, U^{\mu\gamma} V^{S^{-1}(\gamma)\nu} S_{AA'} \Lambda^{\nu}_{A'} S_{BB'} \Gamma^{S^{-1}(\alpha)}_{B'} S_{CC'} \Gamma^{S^{-1}(\beta)}_{C'} \quad . \tag{3.3.17}$$

If we change the summation variables on the righthand side from α,β to $S(\alpha),S(\beta)$ and compare corresponding terms, we find that crystal symmetry requirements have imposed the condition

$$^{(1020)}H^{\mu\alpha\beta}_{ABC} = \sum_{\nu,\gamma} {}^{(1020)}H^{\nu S(\alpha) S(\beta)}_{A'B'C'}\; U^{\nu S(\gamma)}\; V^{\gamma\mu}\; S_{A'A}\; S_{B'B}\; S_{C'C} \quad . \tag{3.3.18}$$

For the special case in which the group operation S does not interchange the sublattices $[S(\alpha) = \alpha, S(\beta) = \beta, S(\gamma) = \gamma]$, Eq. (3.3.18) reduces to

$$^{(1020)}H^{\mu\alpha\beta}_{ABC} = {}^{(1020)}H^{\mu\alpha\beta}_{A'B'C'}\ S_{A'A}\ S_{B'B}\ S_{C'C} \quad . \qquad (3.3.19)$$

We note that if inversion symmetry is present, the above expansion coefficient vanishes. For the case in which S does interchange the sublattices, the reader is referred to the comparison made by Lax and Nelson[83] between the diamond and NaCl structures. The use of subgroup techniques[84] could greatly facilitate any calculation of the symmetry requirements imposed on the expansion coefficient in Eq. (3.3.18) by the full group.

C. The Stored Energy and the Natural State

The choice of $\Gamma^{T\alpha}_{A}$ as expansion parameters for $\rho^0\Sigma$ has one drawback. In the natural state of the crystal, i.e. when no external influences are applied to the crystal and all space and time derivatives vanish, $E_{AB} = 0$, $\Lambda^{\mu}_{A} = 0$, $\Gamma^{\alpha}_{A;B} = 0$ but $\Gamma^{T\alpha}_{A}$ has a spontaneous value of

$$\Gamma^{S\alpha}_{A} \equiv \delta_{iA}\, m^{S\alpha}_{i} \quad . \qquad (3.3.20)$$

This leads to the spontaneous value of $\rho^0\Sigma$ and $\partial\rho^0\Sigma/\partial\Gamma^{T\alpha}_{A}$ having an infinite series of terms. This can be avoided by introducing a new quantity

$$\Gamma^{\alpha}_{A} \equiv \Gamma^{T\alpha}_{A} - \Gamma^{S\alpha}_{A} \quad (\alpha = 1,\ldots,N) \quad . \qquad (3.3.21)$$

Using Eqs. (3.3.5),(3.3.20) and (3.3.21) we obtain

$$\Gamma_A^{\alpha} = (R_{iA} - \delta_{iA})m_i^{S\alpha} + R_{iA}m_i^{\alpha} \quad . \tag{3.3.22}$$

Equation (3.3.16) can now be reexpressed as

$$\begin{aligned}\rho^0 \sum = {}& {}^{(1000)}K_A^{\mu}\,\Lambda_A^{\mu} + {}^{(0100)}K_{AB}\,E_{AB} + {}^{(2000)}K_{AB}^{\mu\nu}\,\Lambda_A^{\mu}\,\Lambda_B^{\nu} \\ & + {}^{(0200)}K_{ABCD}E_{AB}E_{CD} + {}^{(1100)}K_{ABC}^{\mu}\Lambda_A^{\mu}\,E_{BC} + {}^{(0010)}K_A^{\alpha}\,\Gamma_A^{\alpha} \\ & + {}^{(0001)}K_{AB}^{\alpha}\,\Gamma_{A;B}^{\alpha} + {}^{(0020)}K_{AB}^{\alpha\beta}\,\Gamma_A^{\alpha}\,\Gamma_B^{\beta} + {}^{(0011)}K_{ABC}^{\alpha\beta}\,\Gamma_A^{\alpha}\,\Gamma_{B;C}^{\beta} \\ & + {}^{(0002)}K_{ABCD}^{\alpha\beta}\Gamma_{A;B}^{\alpha}\,\Gamma_{C;D}^{\beta} + {}^{(1010)}K_{AB}^{\mu\alpha}\,\Lambda_A^{\mu}\,\Gamma_B^{\alpha} + {}^{(0110)}K_{ABC}^{\alpha}E_{AB}\Gamma_C^{\alpha} \\ & + {}^{(1001)}K_{ABC}^{\mu\alpha}\,\Lambda_A^{\mu}\,\Gamma_{B;C}^{\alpha} + {}^{(0101)}K_{ABCD}^{\alpha}\,E_{AB}\,\Gamma_{C;D}^{\alpha} + \ldots\end{aligned} \tag{3.3.23}$$

Expressions for the new expansion coefficients ${}^{(k,l,m,n)}K$ in terms of the old expansion coefficients can easily be found from Eqs. (3.3.16),(3.3.21) and (3.3.23) but they will not be presented here. It is noted that odd power terms in the magnetizations and/or magnetization gradients are present in the new expansion. This apparent violation of time reversal is explained by the fact that the new expansion coefficients relating to these terms are themselves odd power functions of the spontaneous sublattice magnetizations. The stored energy is now expanded in rotationally invariant variables which vanish in the natural state. The constant term in $\rho^0\sum$ which cannot contribute to the equations of motion, has been dropped.

3.4 Maxwell-Lorentz Equations

The Lagrange equation (3.2.39) for the scalar potential, regarded as a generalized coordinate, yields

$$\epsilon_0 \nabla \cdot \vec{E} = -\nabla \cdot \vec{P} \equiv q^D \tag{3.4.1}$$

with the aid of Eqs. (3.1.3), (3.1.5), (3.1.6), (3.2.21) and (3.2.31). The righthand side of Eq. (3.4.1) is the dielectric or bound charge in the dipole approximation, and the polarization $\vec{P}$ is defined by

$$\vec{P}(\vec{z},t) \equiv \sum_{\mu}{}' q^{\mu} \left[\vec{y}^{\mu}(\vec{X},t)/J(\vec{X},t) \right]_{\vec{z}=\vec{x}(\vec{X},t)} \quad . \tag{3.4.2}$$

The Lagrange equation (3.2.39) for the vector potential $\vec{A}$, regarded as a generalized coordinate, yields

$$\nabla \times \vec{B}(\vec{z},t)/\mu_0 - \epsilon_0 \frac{\partial \vec{E}}{\partial t} = \vec{j}^D + \vec{j}^M + \vec{j}^N \quad , \tag{3.4.3}$$

with the aid of Eqs. (3.1.3), (3.1.5), (3.1.6), (3.2.21) and (3.2.31). The dielectric or bound charge current in the dipole approximation is given by

$$\vec{j}^D \equiv \frac{\partial \vec{P}}{\partial t} + \nabla \times (\vec{P} \times \dot{\vec{x}}) \quad . \tag{3.4.4}$$

The magnetization current is given by the curl of the spatial frame magnetization

$$\vec{j}^M \equiv \nabla \times \vec{M}(\vec{z},t) \quad , \tag{3.4.5}$$

and the intrinsic spin magnetic quadrupole current is given by

$$\vec{j}^N \equiv -\nabla \times (\nabla \cdot \underline{N}(\vec{z},t)) \quad . \tag{3.4.6}$$

Alternatively, we could have obtained the same results in Eqs. (3.4.1) and (3.4.3) by using the interaction Lagrangian correct to all orders, Eq. (3.2.23), in which case the Lagrange equations become

$$\epsilon_0 \nabla \cdot \vec{E} = q(\vec{z},t) \quad , \tag{3.4.7}$$

$$\nabla \times \vec{B}(\vec{z},t)/\mu_0 - \epsilon_0 \frac{\partial \vec{E}}{\partial t} = \vec{j}(\vec{z},t) + \nabla \times \vec{m}(\vec{z},t) \quad . \tag{3.4.8}$$

To dipole order, the righthand sides of Eqs. (3.4.7) and (3.4.8) reduce to the righthand sides of Eqs. (3.4.1) and (3.4.3) respectively.[85]

By comparison of Eqs. (3.4.1) and (3.4.3) with the conventional form of Maxwell's equations, we must define the electric displacement vector $\vec{D}$ and the magnetic field $\vec{H}$ by

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P} \quad , \tag{3.4.9}$$

$$\vec{H} = \vec{B}/\mu_0 - \vec{P} \times \dot{\vec{x}} - \vec{M} + \nabla \cdot \underline{N} \quad . \tag{3.4.10}$$

The two remaining Maxwell equations,

$$\nabla \times \vec{E} + \partial\vec{B}/\partial t = 0 \quad , \tag{3.4.11}$$

$$\nabla \cdot \vec{B} = 0 \quad , \tag{3.4.12}$$

are direct consequences of the definitions, Eqs. (3.1.5) and (3.1.6), of the $\vec{E}$ and $\vec{B}$ fields in terms of the Lagrangian coordinates $\vec{A}$ and Φ.

It must be emphasized that we have calculated the Maxwell-Lorentz equations in this section with all quantities referred to the spatial frame. To perform a similar calculation in the material frame, the Lagrangian must be transformed to that frame, and new Lagrangian coordinates different from $\vec{A}$ and Φ must be chosen. Moreover, all electromagnetic quantities such as the electromagnetic fields, the polarization, etc., will have different forms when expressed in the material frame. For a complete discussion of this problem the reader is referred to Lax and Nelson,[86] Walker et al,[87] and Thurston.[88]

3.5 Center of Mass Equation

The material frame Lagrangian equation (3.2.40) for the center of mass position $\vec{x}$ yields

$$\rho^0 \ddot{x}_i = (\vec{p} \times \dot{\vec{B}})_i + (\dot{\vec{p}} \times \vec{B})_i + \vec{p} \cdot \vec{E}_{,i} + \vec{p} \cdot (\dot{\vec{x}} \times \vec{B}_{,i})$$

$$+ \{\sum_\alpha \vec{m}^{T\alpha} + \frac{1}{2} \sum_{\mu,\nu}{}' q^{\mu\nu} (\vec{y}^\mu \times \dot{\vec{y}}^\nu)\} \cdot \vec{B}_{,i}$$

$$+ \sum_\alpha \vec{m}^{T\alpha} \cdot (\vec{u}^\alpha \cdot \nabla) \vec{B}_{,i} + T_{iA,A} \quad , \qquad (3.5.1)$$

with the aid of Eqs. (3.2.21), (3.2.30), (3.2.38) and (3.3.23). Here

$$\vec{p} \equiv \sum_\mu{}' q^\mu \vec{y}^\mu = \sum_\alpha q^\alpha \vec{u}^\alpha \quad , \qquad (3.5.2)$$

is the dipole moment per unit volume in the material frame, and the Piola-Kirchoff mixed frame stress tensor is defined by

$$T_{iA} \equiv \rho^0 \, \partial\Sigma/\partial x_{i,A} \quad . \qquad (3.5.3)$$

In Eq. (3.5.3) Σ may be thought of as a function of the original dynamical variables $x_{i,A}$, y_i^μ, $m_i^{T\alpha}$, $m_{i,A}^\alpha$ which is clear from the definitions Eqs. (3.3.1)-(3.3.6) and Eq. (3.3.9). The derivative in Eq. (3.5.3) is to be taken holding all the original dynamical variables fixed except $x_{i,A}$. With the use of vector identities and Eqs. (3.2.19) and (3.4.11), the center of mass equation (3.5.1) can be reexpressed as

$$\rho^0 \ddot{x}_i = T_{iA,A} + (\vec{p} \cdot \nabla) E_i + (\dot{\vec{p}} \times \vec{B})_i + [\dot{\vec{x}} \times (\vec{p} \cdot \nabla) \vec{B}]_i$$

$$+ \{\sum_\alpha \vec{m}^{T\alpha} + \frac{1}{2} \sum_{\mu,\nu}{}' q^{\mu\nu}(\vec{y}^\mu \times \dot{\vec{y}}^\nu)\} \cdot \vec{B}_{,i}$$

$$+ \sum_\alpha \vec{m}^{T\alpha} \vec{u}^\alpha : \nabla\vec{B}_{,i} \quad . \tag{3.5.4}$$

This equation can be transformed to a spatial frame equation by multiplying by J^{-1} defined in Eq. (3.2.37), and rearranging terms. With the use of the Euler-Piola-Jacobi identity,[89]

$$(J^{-1} x_{j,A})_{,j} = 0 \quad , \tag{3.5.5}$$

it is easy to show that

$$J^{-1} T_{iA,A} = t^x_{ij,j} \quad , \tag{3.5.6}$$

where the local stress tensor t^x_{ij} is defined by

$$t^x_{ij} \equiv \rho \; (\partial\Sigma/\partial x_{i,B}) \; x_{j,B} \quad , \tag{3.5.7}$$

the x superscript signifying that all variables except $x_{i,B}$ are to be held fixed while taking the derivative. The mass density referred to the spatial frame is defined by

$$\rho \equiv J^{-1} \rho^0 \quad . \tag{3.5.8}$$

We state for future reference a theorem[90] relating material and spatial time derivatives. If $\Gamma(\vec{z},t)$ is any physical quantity expressed in the spatial frame and $\Upsilon(\vec{X},t)$ the corresponding quantity expressed in the material frame related to one another by

$$\Gamma(\vec{z},t) = \Upsilon(\vec{X},t)/J(\vec{X},t)\Big|_{\vec{z}=\vec{x}(\vec{X},t)}$$

$$\equiv \hat{\Upsilon}(\vec{z},t)/\hat{J}(\vec{z},t) \quad , \qquad (3.5.9)$$

then we have

$$\frac{\partial\Gamma(\vec{z},t)}{\partial t} + \frac{\partial[\Gamma(\vec{z},t)\dot{x}_i]}{\partial z_i} = \hat{J}^{-1}\,\frac{d(\hat{J}\Gamma)}{dt} \quad . \qquad (3.5.10)$$

With the use of the above theorem, we find that

$$J^{-1}\overset{*}{p} = \partial\vec{P}/\partial t + \nabla \times (\vec{P} \times \overset{*}{x}) + (P_i\,\overset{*}{x})_{,i} \quad . \qquad (3.5.11)$$

Equation (3.5.4) can then be put in the spatial frame form

$$\rho\ddot{x}_i = [t^x_{ij} + P_j\,\hat{E}_i + (\vec{M}\cdot\vec{B})\delta_{ij} - B_j\,M_i + B_j(\nabla\cdot\underline{N})_i$$

$$- (\nabla\cdot\underline{N})_k\,B_k\,\delta_{ij} + N_{kj}\,B_{k,i}]_{,j}$$

$$+ q^D\,E_i + [(\vec{J}^D + \vec{J}^M + \vec{J}^N) \times \vec{B}]_i \quad . \qquad (3.5.12)$$

3.6 Effective Field Contributions

The problem of calculating the exact electric field at a point inside a crystal has been investigated previously.[91,92] When we expanded the electric and magnetic fields about the center of mass value, we did so under the assumption that these fields were slowly varying. In reality the electromagnetic fields are only slowly varying if one compares corresponding points in adjacent cells, but within any given cell these fields are rapidly varying. It is clear that in calculating the fields which act on a particle, the self field due to the particle's charge and spin dipole must be omitted. Following the definition of Lax and Nelson,[93] we use the term effective field to refer to that field acting on a particle which includes the local-field contribution but excludes the self field. Such effective field contributions imply that the exact field can be expressed as the sum of a macroscopic field and, in the terminology of Born and Huang,[92] an "inner field".

For the exact field acting on a particle at position $\vec{x}^{\alpha}$, it is sufficient to make the replacements

$$\vec{E}(\vec{x}^{\alpha}) \rightarrow \vec{E}(\vec{x}^{\alpha}) + \sum_{\beta} \underline{L}_1^{\alpha\beta} \cdot \vec{p}^{\beta} \quad , \tag{3.6.1}$$

$$\vec{B}(\vec{x}^{\alpha}) \rightarrow \vec{B}(\vec{x}^{\alpha}) + \sum_{\beta} \underline{L}_2^{\alpha\beta} \cdot \vec{m}^{T\beta} \quad , \tag{3.6.2}$$

where

$$\vec{p}^{\beta}(\vec{X},t) \equiv q^{\beta} \, \vec{u}^{\beta}(\vec{X},t) \quad , \tag{3.6.3}$$

is the dipole moment per unit volume associated with a particle of type β. The matrices $\underline{L}_1^{\alpha\beta}$ and $\underline{L}_2^{\alpha\beta}$ are parameters that describe the strength of the local fields at the site of particle α due to the electric and magnetic dipoles situated at the position of particle β. To a first approximation $\underline{L}_1^{\alpha\beta}$ and $\underline{L}_2^{\alpha\beta}$ are taken to be constant matrices for fixed α and β; that is, their values are assumed to correspond to those of the static undeformed lattice (or a homogeneously deformed lattice). Moreover, it will be assumed that the matter interacts with these local inner fields only through electric dipole, magnetic dipole and higher order multipole interactions.

Subject to the validity of the above assumptions, we may associate with the Lagrangian a local part of the form

$$
\begin{aligned}
\hat{L}_{local} = \frac{1}{2} \sum_{\alpha\neq\beta} q^{\alpha} q^{\beta} \vec{u}^{\alpha} \cdot \underline{L}_1^{\alpha\beta} \cdot \vec{u}^{\beta} \\
+ \frac{1}{2} \sum_{\alpha\neq\beta} \vec{m}^{T\alpha} \cdot \underline{L}_2^{\alpha\beta} \cdot \vec{m}^{T\beta} \quad , \\
+ \sum_{\alpha\neq\beta} \underline{q}^{\alpha} : \nabla \underline{L}'^{\alpha\beta}_1 \cdot q^{\beta} \vec{u}^{\beta} \\
+ \sum_{\alpha\neq\beta} \vec{m}^{T\alpha} \vec{u}^{\alpha} : \nabla \underline{L}'^{\alpha\beta}_2 \cdot \vec{m}^{T\beta} + \ldots
\end{aligned}
\qquad (3.6.4)
$$

where

$$
q^{\alpha}_{ij}(\vec{X},t) \equiv q^{\alpha} u^{\alpha}_i(\vec{X},t)\, u^{\alpha}_j(\vec{X},t) \quad , \qquad (3.6.5)
$$

is the electric quadrupole moment moment per unit volume associated with a particle of type α, and the parameters $\underline{L}'^{\alpha\beta}_{(1,2)}$ are not necessarily identical to $\underline{L}^{\alpha\beta}_{(1,2)}$. The rotationally invariant generalization of the terms in Eq. (3.6.4) will have the same form as terms already present in the stored energy and may be absorbed into them with only a modification of the relevant expansion coefficients. The absorption of the inner fields into the stored energy means that the electromagnetic fields used in this paper are in fact the long wavelength macroscopic Maxwellian fields.

3.7 Internal Motion Equations

The material frame Lagrange equation (3.2.40) for the internal coordinate $\vec{y}^{\mu}(\mu \neq 0)$ yields

$$m^{\mu}\, \ddot{y}^{\mu}_{i} = q^{\mu}\, \hat{E}_{i} + \vec{m}^{T\mu} \cdot \vec{B}_{,i} + \sum_{\nu}{}' q^{\mu\nu}\, (\dot{\vec{y}}^{\nu} \times \vec{B})_{i}$$

$$- \rho^{0}\, \partial\Sigma/\partial y^{\mu}_{i} \qquad (3.7.1)$$

with the use of Eqs. (3.2.21), (3.2.30), (3.2.32), (3.2.33), (3.2.34), (3.2.38) and (3.3.23). In the natural state (NS) of the magnetic crystal $\vec{B} = \vec{B}^{S}$ (the spontaneous magnetic field), $R_{iA} = \delta_{iA}$, and all time and space derivatives are zero. Equation (3.7.1) then becomes with the help of Eqs. (3.3.3) and (3.3.23)

$$0 = -\delta_{iA}(\partial\rho^0\Sigma/\partial\Lambda^\mu_A)^{NS} = -\delta_{iA}\ {}^{(1000)}K^\mu_A \quad , \qquad (3.7.2)$$

which implies that we may choose ${}^{(1000)}K^\mu_A = 0$ in Eq. (3.3.23). However, this result is not true for pyroelectric materials[94] where a non-zero electric field (the spontaneous electric field) is present in the natural state.

3.8 Equations of Motion for Sublattice Magnetizations

Although the general structure of our theory does not require it, we will assume for simplicity here that the sublattice magnetization vectors are perfectly rigid along their lengths, i.e.,

$$|\vec{m}^{T\alpha}|^2 = |\vec{m}^{S\alpha}|^2 \quad . \qquad (3.8.1)$$

As discussed in Chapter II, it is necessary to account for the angular momenta of the spin and electron orbital motion which generate the magnetization. This can be done by supplementing the material frame Lagrangian density of the system [Eq. (3.2.21)] with a gyroscopic term which is the generalization of Eq. (2.1.2) for the case of more than one particle per unit cell. This term is given by

$$\hat{K}^M = \frac{1}{3}\sum_{\alpha,J}\frac{1}{\gamma^\alpha}\frac{(\vec{m}^{T\alpha}\cdot\hat{e}^J)[\dot{\vec{m}}^{T\alpha}\cdot(\vec{m}^{T\alpha}\times\hat{e}^J)]}{|\vec{m}^{T\alpha}|^2-(\vec{m}^{T\alpha}\cdot\hat{e}^J)^2} \quad , \qquad (3.8.2)$$

where γ^α is the gyromagnetic ratio of magnetic sublattice α.

Considering only that part of our Lagrangian density which involves the magnetic coordinates, and putting in the contraints by means of undetermined multipliers, we have in the material frame

$$\hat{L}_{MAG} = \hat{K}^{M} + \sum_{\alpha} \vec{m}^{T\alpha} \cdot \vec{B}(\vec{x}^{\alpha},t) - \rho^{0} \Sigma + \sum_{\alpha} \lambda^{\alpha} \{ | \vec{m}^{T\alpha} |^{2} - | \vec{m}^{S\alpha} |^{2} \} \quad . \tag{3.8.3}$$

λ^{α} is the Lagrange multiplier for sublattice α.

We choose $\{\vec{m}^{T\alpha}(\vec{X},t), \dot{\vec{m}}^{T\alpha}(\vec{X},t)\}$ as magnetic variables for the Lagrange equation of motion (3.2.40) from which we obtain the effective magnetic field for sublattice α:

$$\frac{d}{dt} \frac{\partial \hat{K}}{\partial \dot{\vec{m}}^{T\alpha}} - \frac{\partial \hat{K}}{\partial \vec{m}^{T\alpha}} - 2\lambda^{\alpha} \vec{m}^{T\alpha} = \vec{B}^{\alpha\, eff} \quad , \tag{3.8.4}$$

where

$$\vec{B}^{\alpha\, eff} \equiv \vec{B}(\vec{x}^{\alpha},t) - \rho^{0} \frac{\partial \Sigma}{\partial \vec{m}^{T\alpha}} + \rho^{0} \frac{\partial}{\partial X_{A}} \frac{\partial \Sigma}{\partial \vec{m}^{T\alpha}_{,A}} \quad . \tag{3.8.5}$$

In the natural state of the crystal, $\dot{\vec{m}}^{T\alpha} = 0$ and $\vec{m}^{T\alpha} = \vec{m}^{S\alpha}$ so that the spontaneous effective field must obey

$$(\vec{B}^{\alpha\, eff})^{NS} = - 2(\lambda^{\alpha})^{NS} \vec{m}^{S\alpha} \quad , \tag{3.8.6}$$

i.e., it must be along the spin direction so as to produce no torque. This agrees with the results of Alexander[95] who analyzed time independent "magnetic structures" at the limit of zero temperature. Moreover, in the natural state $\vec{B}(\vec{x}^{\alpha},t) = \vec{B}^S$ and $R_{iA} = \delta_{iA}$. Here $\vec{B}^S$ is the spontaneous magnetic field inside the crystal when no external influences are applied. Then with the aid of Eqs. (3.3.5), (3.3.21), (3.3.23) and (3.8.5), we have the condition

$$(B^{\alpha\ \mathrm{eff}})_i^{NS} = B_i^S - {}^{(0010)}K_A^{\alpha}\ \delta_{iA} \ . \tag{3.8.7}$$

Multiplying Eq. (3.8.4) by γ^{α} and taking the vector cross product with $\vec{m}^{T\alpha}$, we can perform the same manipulations as presented in Section 2.1 to obtain the magnetic torque equation generalized to elastic continua:

$$\dot{\vec{m}}^{T\alpha}(\vec{X},t) = \gamma^{\alpha}\ \vec{m}^{T\alpha}(\vec{X},t) \times \vec{B}^{\alpha\ \mathrm{eff}} \ . \tag{3.8.8}$$

In the natural state, the equation of motion Eq. (3.8.8) combined with Eq. (3.8.7) yields

$$\epsilon_{ijk}\ m_j^{S\alpha}(B_k^S - {}^{(0010)}K_A^{\alpha}\ \delta_{kA}) = 0 \ . \tag{3.8.9}$$

This means that only the component of the quantity in parentheses above which is perpendicular to the spontaneous magnetic moment need vanish.

The calculation of the Dirac brackets between components of the sublattice magnetization proceeds in exactly the same fashion as presented in Section 2.3, the result being

$$\{m_i^{T\alpha}(\vec{X},t),\ m_j^{T\alpha}(\vec{X}',t)\}^* = \gamma^\alpha\ \epsilon_{ijk}\ m_k^{T\alpha}\ \delta^3(\vec{X} - \vec{X}') \quad . \qquad (3.8.10)$$

For future reference we note that to the electric dipole approximation,

$$\sum_\alpha \vec{m}^{T\alpha} \cdot \vec{B}(\vec{x}^\alpha(\vec{X},t),t) = \sum_\alpha \vec{m}^{T\alpha} \cdot \vec{B}(\vec{x}(\vec{X},t),t)$$

$$+ \sum_\mu{}' \ \vec{m}^{T\mu} \cdot (\vec{y}^\mu(\vec{X},t) \cdot \nabla)\ \vec{B}(\vec{x}(\vec{X},t),t) + \ldots \qquad (3.8.11)$$

Thus, the use of the righthand side of Eq. (3.8.5) in conjunction with Eq. (3.8.11) yields in the dipole approximation

$$\vec{B}^{\alpha\,\mathrm{eff}} = \vec{B}(\vec{x},t) + (\vec{u}^\alpha \cdot \nabla)\vec{B} - \rho^0 \frac{\partial\Sigma}{\partial\vec{m}^{T\alpha}} + \frac{\partial}{\partial X_A}\frac{\partial\rho^0\Sigma}{\partial\vec{m}^{T\alpha}_{,A}} \quad . \qquad (3.8.12)$$

3.9 Momentum Conservation

Momentum conservation results from the invariance to spatial displacement of the equations of motion. This is automatically satisfied since we have constructed in our theory a Lagrangian which is not an explicit function of the position $\vec{x}$. Momentum conservation may be expressed as

$$\frac{\partial g_i^c}{\partial t} - \frac{\partial t_{ij}^c}{\partial z_j} = 0 \ , \qquad (3.9.1)$$

where g_i^c is the canonical momentum density given by

$$g_i^c \equiv - \sum_\alpha \frac{\partial \hat{L}^S}{\partial(\frac{\partial \Psi^\alpha}{\partial t})} \ \Psi^\alpha_{,i} \ , \qquad (3.9.2)$$

and t_{ij}^c is the canonical stress tensor given by

$$t_{ij}^c \equiv \sum_\alpha \{ \ \Psi^\alpha_{,i} \frac{\partial \hat{L}^S}{\partial \Psi^\alpha_{,j}} - \Psi^\alpha_{,i} \ (\frac{\partial \hat{L}^S}{\partial \Psi^\alpha_{,jk}})_{,k} + \Psi^\alpha_{,ik} \frac{\partial \hat{L}^S}{\partial \Psi^\alpha_{,kj}} \} - \hat{L}^S \delta_{ij} \ . \qquad (3.9.3)$$

Here $\hat{L}^S$ is the spatial frame Lagrangian density regarded as a function of the N fields $\Psi^\alpha(\alpha=1,2,\ldots,N)$, their spatial frame time derivatives $\partial\Psi^\alpha/\partial t$ ($\vec{x}$ held fixed), and their first and second space derivatives $\Psi^\alpha_{,j} \equiv \partial\Psi^\alpha/\partial z_j$ and $\Psi^\alpha_{,jk} \equiv \partial^2\Psi^\alpha/\partial z_j \partial z_k$. The independent variables used in this spatial description are $\vec{z}$ (equivalently $\vec{x}$ within matter) and t.

In order to transform any portion of the Lagrangian density from the material to spatial frames, we use Eq. (3.2.20) which implies that

$$\hat{L}^S = J^{-1}(\vec{x}/\vec{X}) \ \hat{L}^M \ , \qquad (3.9.4)$$

where $J(\vec{x}/\vec{X})$ is the Jacobian of the transformation from the material to spatial coordinates as defined in Eq. (3.2.37). From Eqs. (3.1.3), (3.2.21), (3.2.31), (3.2.38), (3.5.8), (3.8.2) and (3.9.4) the Lagrangian density to be used is given as

$$\tilde{L}^S = \frac{1}{2}\rho\,\dot{\vec{x}}\cdot\dot{\vec{x}} + \sum_{\mu}{}' \rho^{\mu}/2\,\dot{\vec{y}}^{\mu}\cdot\dot{\vec{y}}^{\mu} - \rho\Sigma + \vec{P}\cdot\vec{E}$$

$$+ \vec{M}\cdot\vec{B} + \underline{N} : \nabla\vec{B} + \frac{1}{2}\epsilon_0\,[\,\vec{E}^2 - c^2\,\vec{B}^2\,]$$

$$+ \frac{1}{3} J^{-1} \sum_{\alpha,\theta} \frac{1}{\gamma^{\alpha}} \frac{(\vec{m}^{T\alpha}\cdot\hat{e}^{\theta})\,[\vec{m}^{T\alpha}\cdot(\dot{\vec{m}}^{T\alpha}\times\hat{e}^{\theta})]}{|\,\vec{m}^{T\alpha}\,|^2 - (\vec{m}^{T\alpha}\cdot\hat{e}^{\theta})^2}$$

$$+ J^{-1}\sum_{\alpha}\lambda^{\alpha}\{|\,\vec{m}^{T\alpha}\,|^2 - |\,\vec{m}^{S\alpha}\,|^2\} \quad , \tag{3.9.5}$$

where the spatial frame mass density associated with the μ-th internal coordinate is related to the corresponding material frame quantity m^{μ} by

$$\rho^{\mu} = J^{-1}\,m^{\mu} \quad . \tag{3.9.6}$$

The material time derivatives appearing in Eq. (3.9.5) may be reexpressed with the aid of Eq. (3.2.19) as

$$\dot{x}_i = -x_{i,K}\,\frac{\partial X_K}{\partial t} \;, \tag{3.9.7a}$$

$$\dot{y}_i^{\mu} = \frac{\partial y_i^{\mu}}{\partial t} - y_{i,j}^{\mu} \, x_{j,K} \frac{\partial X_K}{\partial t} \quad , \tag{3.9.7b}$$

$$\dot{m}_i^{T\alpha} = \frac{\partial m_i^{T\alpha}}{\partial t} - m_{i,j}^{\alpha} \, x_{j,K} \frac{\partial X_K}{\partial t} \quad . \tag{3.9.7c}$$

Since we wish to refer everything to the spatial frame, the deformation gradient $x_{i,K}$ must be regarded as a function of $X_{L,j}$ as must J^{-1}, ρ and ρ^{μ}. The fields $\psi^{\alpha}(\vec{x},t)$ that appear in Eqs. (3.9.2) and (3.9.3) can be identified as X_A, y_i^{μ}, $m_i^{T\alpha}$, A_i, and φ. Substitution of $\hat{\mathcal{L}}^S$ from Eq. (3.9.5) into Eqs. (3.9.2) and (3.9.3) yields with the aid of Eqs. (3.1.5), (3.1.6), (3.4.9), (3.4.10) and (3.5.7)

$$g_i^c = \rho \dot{x}_i - (\vec{P} \times \vec{B})_i + D_j \, A_{j,i} \quad , \tag{3.9.8}$$

$$t_{ij}^c = t_{ij}^x - \rho \dot{x}_i \, \dot{x}_j - \frac{\epsilon_0}{2} E_K E_K \delta_{ij} + \frac{1}{2\mu_0} B_K B_K \delta_{ij}$$

$$+ (\vec{P} \times \vec{B})_i \, \dot{x}_j + (E_i + \frac{\partial A_i}{\partial t}) \, D_j - H_l \, \epsilon_{ljK} \, A_{K,i}$$

$$+ N_{lj} \, B_{l,i} - \sum_{\alpha} \rho \frac{\partial \Sigma}{\partial m_{K,j}^{\alpha}} \, m_{K,i}^{\alpha} \quad . \tag{3.9.9}$$

The above expressions are not gauge invariant and can not represent the physical quantities we are seeking. The question of uniqueness of the momentum density and the stress tensor will now be addressed.

3.10 Total Stress Tensor

In order to remove the non-gauge invariant terms appearing in the canonical forms of the momentum density and the total stress tensor, we first reexpress Eqs. (3.9.8) and (3.9.9) with the aid of Eqs. (3.1.6), (3.2.33), (3.4.9) and (3.4.10), in the forms

$$g_i^c = \rho\dot{x}_i + \epsilon_0(\vec{E} \times \vec{B})_i + A_{i,j} D_j \quad , \qquad (3.10.1)$$

$$\begin{aligned} t_{ij}^c = {} & t_{ij}^x + m_{ij} + \hat{E}_i P_j + M_k B_k \delta_{ij} - M_i B_j \\ & - (\nabla \cdot \underline{N})_k B_k \delta_{ij} + (\nabla \cdot \underline{N})_i B_j + N_{lj} B_{li} \\ & - \rho\dot{x}_i \dot{x}_j - \sum_\alpha \rho \frac{\partial\Sigma}{\partial m_{k,j}^\alpha} m_{k,i}^\alpha \\ & + \frac{\partial A_i}{\partial t} D_j + A_{i,k} \epsilon_{kjl} H_l \end{aligned} \qquad (3.10.2)$$

where

$$m_{ij} \equiv \epsilon_0 E_i E_j + \frac{1}{\mu_0} B_i B_j - \frac{\epsilon_0}{2} E_k E_k \delta_{ij} - \frac{1}{2\mu_0} B_k B_k \delta_{ij} = m_{ji} \qquad (3.10.3)$$

is the Maxwell vacuum-field stress tensor. When the non-gauge invariant parts of g_i^c and t_{ij}^c are substituted into the momentum conservation law Eq. (3.9.1) they yield a result

$$\frac{\partial}{\partial t}(A_{i,j} D_j) - \frac{\partial}{\partial z_j}\left(\frac{\partial A_i}{\partial t} D_j + A_{i,k} \epsilon_{kjl} H_l \right) = 0 \quad , \qquad (3.10.4)$$

whose vanishing is a consequence of the two Maxwell equations for a magnetic dielectric that follow from the discussion in Section 3.4:

$$\nabla \cdot \vec{D} = 0 \quad , \tag{3.10.5}$$

$$\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} \quad . \tag{3.10.6}$$

The cancellation of the non-gauge invariant terms as exhibited in Eq. (3.10.4) removes one of the obstacles to uniqueness and leads us to tentatively identify

$$g_i \equiv \rho \dot{x}_i + \epsilon_0 (\vec{E} \times \vec{B})_i \quad , \tag{3.10.7}$$

as the <u>total momentum density</u> in the spatial or laboratory frame and

$$t^L_{ij} \equiv t^X_{ij} + m_{ij} + \hat{E}_i P_j + M_K B_K \delta_{ij} - M_i B_j - (\nabla \cdot \underline{N})_K B_K \delta_{ij}$$

$$+ (\nabla \cdot \underline{N})_i B_j + N_{lj} B_{l,i} - \rho \dot{x}_i \dot{x}_j$$

$$- \sum_\alpha \rho \frac{\partial \Sigma}{\partial m^\alpha_{K,j}} m^\alpha_{K,i} \quad , \tag{3.10.8}$$

as the <u>total stress tensor</u> in the laboratory frame. To justify this identification, we employ the same line of reasoning put forth by Nelson and Lax[96] in a recent paper for the case of the dielectric. These arguments are briefly summarized below.

The quantitites of Eqs. (3.10.7) and (3.10.8) satisfy the spatial frame momentum conservation law,

$$\frac{\partial g_i}{\partial t} - t^L_{ij,j} = 0 \quad . \qquad (3.10.9)$$

Application of the conventional "pill box" argument[97] to Eq. (3.10.9) yields a jump condition on t^L_{ij} of the form

$$[\, t^L_{ij} \,]\, n_j = 0 \quad , \qquad (3.10.10)$$

where [ ] denotes the jump in the quantity enclosed in the brackets across the surface considered,

$$[\, t^L_{ij} \,] \equiv (\, t^L_{ij} \,)^{out} - (\, t^L_{ij} \,)^{in} \quad , \qquad (3.10.11)$$

and $\vec{n}$ is the outward pointing normal to the surface which is stationary in the spatial frame. The boundary condition Eq. (3.10.11) is much more restrictive in determining the total stress tensor than the conservation law Eq. (3.10.9) which involves the divergence. For example, the addition of a curl-like quantity inside the divergence of the conservation law yields a new stress tensor,

$$(t^L_{ij})' = t^L_{ij} + \epsilon_{jkl} f_{il,k} \quad , \qquad (3.10.12)$$

but the boundary condition is still found to involve only t^L_{ij}. The transformation

$$(t^L_{ij})' = t^L_{ij} + \frac{\partial h_{ij}}{\partial t} \quad , \qquad (3.10.13)$$

$$g'_i = g_i + h_{ij,j} \quad , \qquad (3.10.14)$$

obtains a new stress tensor and a new momentum density in the conservation law, but this transformation does affect the boundary condition Eq. (3.10.10). The transformation

$$g'_i = g_i + c_i \quad , \qquad (3.10.15)$$

where c_i is independent of time, does not change the stress tensor in either the conservation law or the boundary condition.

It is clear from the above that the boundary condition requirement will enable us to properly determine a unique total stress tensor. The scalar product of the stress tensor with the unit normal will not lead to non-uniqueness because t^L_{ij} cannot be a function of the orientation of a body surface and because that orientation (and hence $\vec{n}$) is arbitrary. The possibility remains that an addition to the stress tensor by means of one of the transformations previously discussed could be continuous at every surface, and hence be part of the total stress tensor while not affecting the boundary condition. Such a transformation could be achieved through Eqs. (3.10.13) and (3.10.14) with h_{ij} spatially independent but linear in time. However, such additions are unobservable and are properly excluded from the definition of the total stress tensor. The way to exclude these so called null stresses is to define a reference state for the total stress tensor which we do by choosing the vacuum to have a total stress tensor and momentum density of

$$t^{L}_{ij} \equiv m_{ij} \qquad \text{(vacuum)}\ , \qquad (3.10.16)$$

$$g_i \equiv \epsilon_0(\vec{E} \times \vec{B}) \qquad \text{(vacuum)}\ . \qquad (3.10.17)$$

Since our definitions of Eqs. (3.10.7) and (3.10.8) reduce to the above for a vacuum, we are in a position to claim that the quantities in Eqs. (3.10.7) and (3.10.8) are unique and represent the momentum density and the total stress tensor respectively.

The total spatial frame stress tensor of Eq. (3.10.8) represents the total force per unit area acting in a direction determined by the first tensor index on an imaginary surface, fixed in the laboratory frame, whose normal is determined by the second index. In order to find a spatial measure of the total force per unit area acting on a moving body surface, we simply apply previous findings[98] resulting in

$$t_{ij}^{B} = t_{ij}^{L} + g_i \dot{x}_j \; . \qquad (3.10.18)$$

It is usually assumed that the natural state (NS) of a boundless perfect crystal is a stress free state. If we adopt this viewpoint, we find using Eqs. (3.10.3) and (3.10.8) that

$$(t_{ij}^{L})^{NS} = t_{ij}^{S} + (\vec{M}^{S} \cdot \vec{B}^{S}) \, \delta_{ij} - M_i^{S} B_j^{S} + B_i^{S} B_j^{S}/\mu_0 - \frac{1}{2\mu_0} B_K^{S} B_K^{S} \delta_{ij} = 0 \; , \qquad (3.10.19)$$

where the spontaneous elastic stress is given by

$$t_{ij}^{S} = (t_{ij}^{X})^{NS} = \delta_{iA} \, \delta_{jB} \; {}^{(0100)}K_{AB} \; . \qquad (3.10.20)$$

In the previous equation we have used Eqs. (3.3.23), (3.5.7), (3.8.7) and the relation[99]

$$\left(\frac{\partial R_{jB}}{\partial x_{i,A}} \right)^{NS} = \frac{1}{2} \, (\delta_{ij} \, \delta_{AB} - \delta_{jA} \, \delta_{iB}) \; . \qquad (3.10.21)$$

3.11 Asymmetry of Total Stress Tensor

It has been shown previously[100] that the total stress tensor for a non-magnetic dielectric is asymmetric when there is more than one particle per unit cell, and hence, internal coordinates $\vec{y}^{\mu}$ are needed to describe the material. We will show that for the case of the magnetic dielectric, the total stress tensor is asymmetric even when there is only one particle per unit cell. This is because of the linear momentum associated with the spin variables.

Examination of Eq. (3.10.8) tells us that we need only consider those terms which are not manifestly symmetric, i.e.,

$$t^{x}_{ij} + \hat{E}_i P_j - M_i B_j + (\nabla \cdot \underline{N})_i B_j + N_{lj} B_{l,i}$$

$$- \sum_{\alpha} \rho \frac{\partial \Sigma}{\partial m^{\alpha}_{K,j}} m^{\alpha}_{K,i} \quad . \tag{3.11.1}$$

We begin by considering t^{x}_{ij} defined in Eq. (3.5.7). It is convenient for purposes of calculation to introduce an equivalent set of rotational invariants for the stored energy. It is seen from an examination of Eqs. (3.3.2) and (3.3.4) that since the tensor $\underline{C}$ is invariant under rotations of the spatial frame, $x_{i,A}$ has precisely the same transformation properties as $R_{i,A}$. Therefore, let

$$\Sigma = \Sigma(E_{AB}, \xi^{\mu}_{A}, \Omega^{\alpha}_{A}, \Omega^{\alpha}_{A;B}) \quad , \tag{3.11.2}$$

where E_{AB} is the Green finite strain tensor (3.3.1), and where

$$\xi^{\mu}_{A} \equiv x_{i,A} Y^{\mu}_{i} \quad , \tag{3.11.3}$$

$$\Omega^{\alpha}_{A} \equiv x_{i,A} m^{T\alpha}_{i} - m^{S\alpha}_{A} \quad , \tag{3.11.4}$$

$$\Omega^{\alpha}_{A;B} \equiv x_{i,A}\, m^{\alpha}_{i,B} \quad . \tag{3.11.5}$$

Here, $m^{S\alpha}_{A}$ is the spontaneous magnetization sublattice α, and is included in the definition so that Ω^{α}_{A} vanishes in the natural state. The introduction of these new rotational invariants does not change the physics but merely rearranges the nonlinear terms in the expansion of the stored energy.

Equation (3.5.7) now becomes

$$t^{x}_{ij} = \rho x_{j,B} \left[\frac{\partial E_{AC}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial E_{AC}} + \sum_{\mu}{}' \frac{\partial \xi^{\mu}_{A}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial \xi^{\mu}_{A}} \right.$$

$$\left. + \sum_{\alpha} \left(\frac{\partial \Omega^{\alpha}_{A}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial \Omega^{\alpha}_{A}} + \frac{\partial \Omega^{\alpha}_{A;C}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial \Omega^{\alpha}_{A;C}} \right) \right] . \tag{3.11.6}$$

We now proceed to determine the asymmetric parts contained in Eq. (3.11.6). It is easy to show, using Eqs. (3.3.1) and (3.3.2) that the first term on the righthand side of Eq. (3.11.6)

$$\rho x_{j,B} \frac{\partial E_{AC}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial E_{AC}} = \rho x_{j,B}\, x_{i,C} \frac{\partial \Sigma}{\partial E_{BC}} \quad , \tag{3.11.7}$$

is symmetric. The equation of motion for the internal coordinates, Eq. (3.7.1), in terms of the new rotational invariants becomes

$$m^{\mu} \ddot{y}^{\mu}_{i} = q^{\mu} \hat{E}_i + \vec{m}^{T\mu} \cdot \vec{B}_{,i} - \rho^0 x_{i,A} \frac{\partial \Sigma}{\partial \xi^{\mu}_{A}} \quad . \qquad (3.11.8)$$

This can be used to solve for the stored energy derivative $\partial\Sigma/\partial\xi^{\mu}_{A}$. Then the second term on the righthand side of Eq. (3.11.6) becomes with the aid of Eqs. (3.2.36), (3.4.2), (3.5.8), (3.9.6) and (3.11.3)

$$\rho x_{j,B} \sum_{\mu}' \frac{\partial \xi^{\mu}_{A}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial \xi^{\mu}_{A}}$$

$$= P_i \hat{E}_j + M_{Ki} B_{K,j} - \sum_{\mu}' \rho^{\mu} y^{\mu}_{i} \ddot{y}^{\mu}_{j} \qquad (3.11.9)$$

The remaining two terms on the righthand side of Eq. (3.11.6) can be rewritten using Eqs. (3.11.4) and (3.11.5) in the form

$$\rho x_{j,B} \sum_{\alpha} \left(\frac{\partial \Omega^{\alpha}_{A}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial \Omega^{\alpha}_{A}} + \frac{\partial \Omega^{\alpha}_{A;C}}{\partial x_{i,B}} \frac{\partial \Sigma}{\partial \Omega^{\alpha}_{A;C}} \right)$$

$$= \rho \sum_{\alpha} \left(m^{T\alpha}_{i} \frac{\partial \Sigma}{\partial m^{T\alpha}_{j}} + m^{\alpha}_{i,K} \frac{\partial \Sigma}{\partial m^{\alpha}_{j,K}} \right) \quad . \qquad (3.11.10)$$

With the aid of Eqs. (3.11.1), (3.11.6)-(3.11.10), we may conclude that the total stress tensor has the asymmetric terms

$$- [M_i - (\nabla \cdot \underline{N})_i] B_j - \sum_{\mu}' \rho^{\mu} y^{\mu}_{i} \ddot{y}^{\mu}_{i}$$

$$+ \rho \sum_{\alpha} \left(m_i^{T\alpha} \frac{\partial \Sigma}{\partial m_j^{T\alpha}} + m_{i,k}^{\alpha} \frac{\partial \Sigma}{\partial m_{j,k}^{\alpha}} - m_{k,i}^{\alpha} \frac{\partial \Sigma}{\partial m_{k,j}^{\alpha}} \right) \neq 0 \quad . \qquad (3.11.11)$$

Thus, the asymmetry of the total stress tensor results not only from the presence of internal coordinates, but also from the magnetic coordinates. This asymmetry and the validity of angular momentum conservation are not mutually exclusive as will be shown presently.

3.12 Conservation of Angular Momentum

The definition of the canonical angular momentum density as introduced by Morse and Fesnbach[101] and Landau and Lifshitz[102] is valid for non-magnetic materials with only one particle per unit cell. Their conclusion that the stress tensor must be symmetric for angular momentum conservation to be obeyed, is true only for such materials. The stress tensor is not symmetric for the case we are considering.

The angular momentum conservation law may be found by combining contributions from the center of mass motion, each of the internal motions, the spin rotations and the electromagnetic fields. The center of mass contribution is found by forming the vector product of Eq. (3.5.12) and $\vec{x}$ and by using Eq. (3.5.10) to obtain

$$\partial/\partial t \ [\rho(\vec{x} \times \dot{\vec{x}})_i] + \partial/\partial z_l \ [\rho(\vec{x} \times \dot{\vec{x}})_i \ \dot{x}_l]$$

$$= [\epsilon_{ijk} x_j \{t^x_{kl} + \hat{E}_k P_l + (\vec{M} \cdot \vec{B})\delta_{kl} - B_l M_k + B_l N_{ka,a}$$

$$- (\nabla \cdot \underline{N})_a B_a \delta_{kl} + N_{al} B_{a,k}\}]_{,l}$$

$$- \epsilon_{ilk} \{t^x_{kl} + \hat{E}_k P_l + (\vec{M} \cdot \vec{B})\delta_{kl} - B_l M_k + B_l N_{ka,a}$$

$$- (\nabla \cdot \underline{N})_a B_a \delta_{kl} + N_{al} B_{a,k}\}$$

$$+ q^D(\vec{x} \times \vec{E})_i + \{\vec{x} \times [(\vec{j}^D + \vec{j}^M + \vec{j}^N) \times \vec{B}]\} \quad . \quad (3.12.1)$$

The contribution from the internal motions is found by forming the vector product of the equation of motion Eq. (3.7.1) of the internal coordinates with $\vec{y}^{\mu}$, multiplying by J^{-1}, summing over μ from 1 to $N - 1$, applying Eq. (3.5.10) and using the definition of the polarization Eq. (3.4.2). The result is

$$\partial/\partial t(l_i) + \partial/\partial z_l(l_i \dot{x}_l) = (\vec{P} \times \hat{E})_i - J^{-1} \epsilon_{ijk} \sum_{\mu}{}' y^{\mu}_j \partial\rho^0 \Sigma/\partial y^{\mu}_k$$

$$+ J^{-1} \epsilon_{ijk} \sum_{\mu}{}' y^{\mu}_j m^{T\mu}_l B_{l,k}$$

$$+ J^{-1} \sum_{\mu,\nu}{}' q^{\mu\nu} [\vec{y}^{\mu} \times (\dot{\vec{y}}^{\nu} \times \vec{B})]_i \, , \quad (3.12.2)$$

where the internal angular momentum is defined by

$$\vec{l} \equiv \sum_{\mu}{}' \rho^{\mu} \vec{y}^{\mu} \times \dot{\vec{y}}^{\mu} \quad . \quad (3.12.3)$$

The linear momentum of the electromagnetic field is obtained by forming the cross product of Eq. (3.4.11) with $\epsilon_0 \vec{E}$ and the cross product of Eq. (3.4.8) with $\vec{B}$ and adding the results. After use of Eqs. (3.4.1), (3.4.12) and (3.10.3) we find

$$\frac{\partial}{\partial t} (\epsilon_0 \vec{E} \times \vec{B})_i - m_{ij,j} = -q^D E_i - [(\vec{j}^D + \vec{j}^M + \vec{j}^N) \times \vec{B}]_i \quad . \tag{3.12.4}$$

The angular momentum of the electromagnetic field is easily found now by forming the vector product of the electromagnetic momentum equation (3.12.4) with $\vec{x}$ and rearranging with the result

$$\frac{\partial}{\partial t}[\vec{x} \times (\epsilon_0 \vec{E} \times \vec{B})]_i - \partial/\partial z_l [\epsilon_{ijk} x_j m_{kl}]$$

$$= -q^D(\vec{x} \times \vec{E})_i - \{\vec{x} \times [(\vec{j}^D + \vec{j}^M + \vec{j}^N) \times \vec{B}]\}_i \quad . \tag{3.12.5}$$

The contribution from the intrinsic spin is found directly from Eq. (3.8.8), multiplying by J^{-1} and with the use of Eq. (3.5.10). The result is

$$\frac{\partial s_i}{\partial t} + \frac{\partial}{\partial z_l}(s_i \dot{x}_l) = \frac{1}{J} \epsilon_{ijk} \sum_\alpha m_j^{T\alpha} B_k^{\alpha\, eff} \quad , \tag{3.12.6}$$

where the intrinsic spin angular momentum is defined by

$$\vec{s} \equiv J^{-1} \sum_\alpha \vec{m}^{T\alpha}/\gamma^\alpha \quad . \tag{3.12.7}$$

We now add the contributions represented by Eqs. (3.12.1), (3.12.2), (3.12.5) and (3.12.6) to obtain after rearrangement and manipulation the statement of angular momentum conservation in the spatial frame

$$\partial/\partial t \; [\{\vec{x} \times (\rho\dot{\vec{x}} + \epsilon_0 \; \vec{E} \times \vec{B})\}_i + l_i + s_i \;]$$

$$+ \; \partial/\partial z_l [\{l_i + s_i\} \; \dot{x}_l - \epsilon_{ijk} \; \{x_j \; t^L_{kl} + x_j \sum_\alpha \rho \frac{\partial \Sigma}{\partial m^\alpha_{a,l}} \; m^\alpha_{a,k}$$

$$+ \sum_\alpha \rho \; m^{T\alpha}_j \frac{\partial \Sigma}{\partial m^\alpha_{k,l}} + N_{ak} \; B_a \; \delta_{lj} \;\}\;] = 0 \quad , \qquad (3.12.8)$$

where we have used Eqs. (3.8.12) and (3.10.8).

By arguments similar to those of Section 3.10, the quantity within the spatial frame time derivative is the spatial frame density of angular momentum, and the quantity in the divergence is the total flow of angular momentum across a surface fixed in the spatial frame.

It should be emphasized again that even though the total stress tensor is not symmetric, the total angular momentum of a magnetic crystal interacting with the electromagnetic field is conserved.

3.13 Energy Conservation

Energy conservation may be expressed as

$$\frac{\partial W}{\partial t} + \frac{\partial S_j}{\partial z_j} = 0 \quad , \tag{3.13.1}$$

where W is the energy density and $\vec{S}$ the energy flow vector. The canonical energy density is defined as

$$W^c = \sum_\alpha \frac{\partial \Psi^\alpha}{\partial t} \frac{\partial \hat{L}^S}{\partial\left(\frac{\partial \Psi^\alpha}{\partial t}\right)} - \hat{L}^S \quad . \tag{3.13.2}$$

All quantities in Eq. (3.13.2) have previously been introduced in Section 3.9. Use of Eqs. (3.9.5), (3.9.7), (3.4.9) and (3.1.5) in Eq. (3.13.2) yields

$$W^c = \frac{1}{2} \rho \dot{\vec{x}} \cdot \dot{\vec{x}} + \sum_\mu{}' \rho^\mu/2 \, \dot{\vec{y}}^\mu \cdot \dot{\vec{y}}^\mu + \rho \sum$$

$$+ \epsilon_0 \vec{E}^2/2 + \vec{B}^2/2\mu_0 - \vec{M}' \cdot \vec{B} - \underline{N} : \nabla \vec{B} + D_j \Phi_{,j} \quad , \tag{3.13.3}$$

where $\vec{M}' \equiv \sum_\alpha \vec{m}^{T\alpha}$ is the magnetization due to spin and electronic contributions; i.e. $\vec{M}'$ does not include the ionic contribution.

The canonical energy flow vector is defined as

$$S_j^c = \sum_\alpha \left\{ \frac{\partial \Psi^\alpha}{\partial t} \frac{\partial \hat{L}^S}{\partial \Psi^\alpha_{,j}} - \frac{\partial \Psi^\alpha}{\partial t} \left(\frac{\partial \hat{L}^S}{\partial \Psi^\alpha_{,jk}} \right)_{,k} + \left(\frac{\partial \Psi^\alpha}{\partial t} \right)_{,k} \frac{\partial \hat{L}^S}{\partial \Psi^\alpha_{,kj}} \right\} \quad . \tag{3.13.4}$$

Use of Eqs. (3.9.5) and (3.9.7) in Eq. (3.13.4) yields

$$S^c_j = [\, \tfrac{1}{2}\, \rho \dot{\vec{x}} \cdot \dot{\vec{x}} + \sum_{\mu}{}' \rho^{\mu}/2\; \dot{\vec{y}}^{\mu} \cdot \dot{\vec{y}}^{\mu}$$

$$- (\vec{P} \cdot \vec{E}) - (\vec{M}' \cdot \vec{B}) - (N : \nabla B)\,]\; \dot{x}_j$$

$$- t^x_{kj}\, \dot{x}_k - \sum_{\alpha} \rho \frac{\partial \Sigma}{\partial m^{\alpha}_{k,j}} \frac{\partial m^{\alpha}_k}{\partial t} + N_{lj} \frac{\partial B_l}{\partial t}$$

$$- \epsilon_{ljk}\, H_l \frac{\partial A_k}{\partial t} - D_j \frac{\partial \Phi}{\partial t} \quad . \qquad (3.13.5)$$

The non-gauge invariant parts of W^c and S^c_j combine in the energy conservation law (3.13.1) to yield

$$\frac{\partial}{\partial t} (D_j\, \Phi_{,j}) - \epsilon_{ljk} \frac{\partial}{\partial z_j} \left(H_l \frac{\partial A_k}{\partial t}\right) - \frac{\partial}{\partial z_j} \left(D_j \frac{\partial \Phi}{\partial t}\right) = \nabla \cdot (\vec{E} \times \vec{H}) \qquad (3.13.6)$$

with the aid of Eqs. (3.1.5), (3.1.6), (3.4.11), (3.10.5) and (3.10.6). Moreover, we use the vector identity

$$- \frac{\partial}{\partial t} (N_{kl}\, B_k)_{,l} + \left(N_{lj} \frac{\partial B_l}{\partial t}\right)_{,j} = - \left(\frac{\partial N_{lj}}{\partial t} B_l\right)_{,j} \quad , \qquad (3.13.7)$$

to obtain the final form of the spatial frame measures of the total energy density and the total energy flow vector:

$$W^L = w + \epsilon_0\, \vec{E}^2/2 + \vec{B}^2/2\mu_0 + \rho \Sigma \quad , \qquad (3.13.8)$$

$$S_j^L = [W - \vec{P} \cdot \vec{E} - (N_{Kl} \; B_K)_{,l}] \; \dot{x}_j$$

$$- t^x_{Kj} \; \dot{x}_K + (\vec{E} \times \vec{H})_j - B_K \frac{\partial N_{Kj}}{\partial t} - \sum_\alpha \rho \frac{\partial \Sigma}{\partial m^\alpha_{K,j}} \frac{\partial m^\alpha_K}{\partial t} \quad , \tag{3.13.9}$$

where

$$W = \frac{1}{2} \rho \; \dot{\vec{x}} \cdot \dot{\vec{x}} + \sum_\mu{}' \frac{\rho^\mu}{2} \dot{\vec{y}} \cdot \dot{\vec{y}}^\mu - (\vec{M}' - \nabla \cdot \underline{N}) \cdot \vec{B} \quad . \tag{3.13.10}$$

By arguments similar to Section 3.10, these quantities are the unique and proper ones.

IV. LINEARIZED EQUATIONS OF MOTION

4.1 Kinematic Corrections

It will be useful for applications and for any iterative schemes in solving the nonlinear equations to separate the linear terms from the nonlinear ones in the equations of motion. The matter equations of motion as given by Eqs. (3.5.4), (3.7.1) and (3.8.8), are written with the material coordinate $\vec{X}$ as the independent variable. These equations are coupled to the electromagnetic fields which are evaluated at the spatial frame position $\vec{x}$. Moreover, spatial frame gradients and time derivatives of the electromagnetic fields enter into the equations of motion.

It will therefore be convenient to express all terms in the matter equations as functions of the spatial coordinate $\vec{x}$. To differentiate between the same physical quantity with $\vec{x}$ as independent variable rather than $\vec{X}$ we add a caret as in

$$\vec{u}(\vec{X},t) = \hat{\vec{u}}(\vec{x},t) \quad , \tag{4.1.1a}$$

$$\vec{y}^{\mu}(\vec{X},t) = \hat{\vec{y}}^{\mu}(\vec{x},t) \quad , \tag{4.1.1b}$$

$$\vec{m}^{\alpha}(\vec{X},t) = \hat{\vec{m}}^{\alpha}(\vec{x},t) \quad , \tag{4.1.1c}$$

$$J(\vec{X},t) = \hat{J}(\vec{x},t) \quad . \tag{4.1.1d}$$

In transforming from the material to the spatial coordinate system, kinematic corrections[103] will arise as a result of the distinction between derivatives with respect to the spatial and material coordinates. Other kinematic corrections also occur as a result of the distinction between material time derivatives and spatial time derivatives. However, when a common Cartesian coordinate system for both spatial and material systems is used, there will be no distinction, say, between a vector component m_j^{α} and a corresponding vector component m_J^{α}.

Some examples will be given in order to clarify these points. For a general function F of the material coordinates, we have

$$\frac{\partial F}{\partial X_A} = \frac{\partial \hat{F}}{\partial x_j}\frac{\partial x_j}{\partial X_A} = \frac{\partial \hat{F}}{\partial x_j}\left(\delta_{jA} + u_{j,A}\right)$$

$$\equiv \frac{\partial \hat{F}}{\partial x_a} + \frac{\partial \hat{F}}{\partial x_j} u_{j,A} \quad , \tag{4.1.2}$$

where the displacement vector $\vec{u}(\vec{X},t)$ in our common Cartesian coordinate system, is defined by the relation

$$\vec{x}(\vec{X},t) \equiv \vec{X} + \vec{u}(\vec{X},t) \quad , \tag{4.1.3}$$

and represents the displacement of the center of mass from its equilibrium or undistorted position $\vec{X}$. Unfortunately, the term $u_{j,A}$ on the righthand side of Eq. (4.1.2) is a material derivative so that we have not completely transformed to spatial derivatives yet. However, replace the function F in Eq. (4.1.2) with the variable u_i, obtaining

$$u_{i,A} = \hat{u}_{i,a} + \hat{u}_{i,j} u_{j,A} \quad . \tag{4.1.4}$$

Equation (4.1.4) can be recast into the matrix form

$$(\delta_{ij} - \hat{u}_{i,j})u_{j,A} = \hat{u}_{i,a} \quad . \tag{4.1.5}$$

The unknown $u_{j,A}$ can be determined now by inverting the matrix factor $\delta_{ij} - \hat{u}_{i,j} \equiv (\underline{1} - \underline{\hat{u}})_{ij}$. This obtains the explicit solution

$$u_{i,A} = (\underline{1} - \underline{\hat{u}})^{-1}_{ij} \hat{u}_{j,a}$$

$$= \hat{u}_{i,a} + \hat{u}_{i,j} \hat{u}_{j,a} + \hat{u}_{i,j} \hat{u}_{j,k} \hat{u}_{k,a} + \ldots \tag{4.1.6}$$

Equation (4.1.2) may now be rewritten with the help of Eq. (4.1.6) as an explicit transformation from a material derivative to an expression containing only spatial derivatives:

$$\frac{\partial F}{\partial X_A} = \frac{\partial \hat{F}}{\partial x_a} + \frac{\partial \hat{F}}{\partial x_j}(\hat{u}_{j,a} + \hat{u}_{j,k}\,\hat{u}_{k,a} + \ldots.) \quad . \qquad (4.1.7)$$

Equation (4.1.7) will enable us to write, for example,

$$m^{\alpha}_{i,A} = \hat{m}^{\alpha}_{i,a} + \hat{m}^{\alpha}_{i,j}\;(\hat{u}_{j,a} + \hat{u}_{j,k}\,\hat{u}_{k,a} + \ldots) \quad . \qquad (4.1.8)$$

It will also be necessary to express the Jacobian of Eq. (3.2.37) in a purely spatial form. To accomplish this, the Jacobian is rewritten in the form

$$\begin{aligned} J &= \det\,(\delta_{iA} + u_{i,A}) \\ &= 1 + u_{i,I} + \frac{1}{2}\,(u_{i,I}\,u_{j,J} - u_{i,J}\,u_{j,I}) \\ &\quad + \det u_{i,A} \quad , \end{aligned} \qquad (4.1.9)$$

where for purposes of the summation convention $I = i$, $J = j$, e.g., $u_{i,I} \equiv u_{1,1} + u_{2,2} + u_{3,3}$. With the help of Eq. (4.1.6), we have the purely spatial form

$$\hat{J} = 1 + \hat{u}_{i,i} + \frac{1}{2}\,(\hat{u}_{i,i}\,\hat{u}_{j,j} + \hat{u}_{i,j}\,\hat{u}_{j,i}) + \ldots. \qquad (4.1.10)$$

Finally, kinematic corrections arise in the transformation of material time derivatives to their spatial counterparts. As seen from the general expression Eq. (3.2.19), the material time derivative $\dot{x}_i$ is contained on the right-hand side of that equation, and it is this derivative which must be evaluated before the transformation is complete. To do this, we set the function F equal to the variable X_A and since by definition the material time derivative of a material coordinate vanishes, we have

$$\frac{dX_A}{dt} = 0 = \frac{\partial X_A(\vec{x},t)}{\partial t} + \frac{\partial X_A(\vec{x},t)}{\partial x_i}\dot{x}_i \quad . \qquad (4.1.11)$$

Solving now for $\dot{x}_i$, we obtain

$$\dot{x}_i = -x_{i,A}\frac{\partial X_A(\vec{x},t)}{\partial t} \quad . \qquad (4.1.12)$$

With the use of Eqs. (4.1.3) and (4.1.6), Eq. (4.1.12) may be rewritten in the purely spatial form

$$\dot{x}_i = (\delta_{iA} + u_{i,A})\frac{\partial \hat{u}_a}{\partial t}$$

$$= \frac{\partial \hat{u}_i}{\partial t} + (\hat{u}_{i,a} + \hat{u}_{i,b}\,\hat{u}_{b,a} + \ldots.)\frac{\partial \hat{u}_a}{\partial t} \quad . \qquad (4.1.13)$$

By choosing F in Eq. (3.2.19) to be either $m_i^{T\alpha}$ or y_i^{μ}, we obtain respectively

$$\dot{m}_i^{T\alpha} = \frac{\partial \hat{m}_i^{\alpha}}{\partial t} + \hat{m}_{i,j}^{\alpha}\frac{\partial \hat{u}_j}{\partial t} + \ldots \quad , \qquad (4.1.14)$$

$$\dot{y}_i^{\mu} = \frac{\partial \hat{y}_i}{\partial t} + \hat{y}_{i,j}^{\mu}\frac{\partial \hat{u}_j}{\partial t} + \ldots \quad , \qquad (4.1.15)$$

where use of Eq. (4.1.13) has been made. To calculate accelerations (material second time derivatives), we choose F in Eq. (3.2.19) to be the corresponding velocities, in our case $\dot{x}_i$ and $\dot{y}_i^\mu$. Doing this, we obtain

$$\ddot{x}_i = \frac{\partial}{\partial t}\left(\frac{\partial \hat{u}_i}{\partial t} + \hat{u}_{i,a}\frac{\partial \hat{u}_a}{\partial t}\right)$$

$$+ \dot{x}_j \frac{\partial}{\partial x_j}\left(\frac{\partial \hat{u}_i}{\partial t} + \hat{u}_{i,a}\frac{\partial \hat{u}_a}{\partial t}\right) + \ldots$$

$$= \frac{\partial^2 \hat{u}_i}{\partial t^2} + \hat{u}_{i,a}\frac{\partial^2 \hat{u}_a}{\partial t^2} + 2\,\frac{\partial \hat{u}_{i,a}}{\partial t}\frac{\partial \hat{u}_a}{\partial t} + \ldots \quad , \qquad (4.1.16)$$

$$\ddot{y}_i^\mu = \frac{\partial}{\partial t}\left(\frac{\partial \hat{y}_i^\mu}{\partial t} + \dot{x}_j\,\hat{y}^\mu_{i,j}\right) + \dot{x}_j \frac{\partial}{\partial x_j}\left(\frac{\partial \hat{y}_i^\mu}{\partial t} + \dot{x}_K\,\hat{y}^\mu_{i,K}\right)$$

$$= \frac{\partial^2 \hat{y}_i^\mu}{\partial t^2} + \frac{\partial^2 \hat{u}_j}{\partial t^2}\,\hat{y}^\mu_{i,j} + 2\,\frac{\partial \hat{u}_j}{\partial t}\frac{\partial \hat{y}^\mu_{i,j}}{\partial t} + \ldots \qquad (4.1.17)$$

after making use of Eqs. (4.1.13) and (4.1.15).

It will also be necessary in the linearization procedure to know the spatial form of the finite rotation tensor defined by Eq. (3.3.4). By utilizing this equation and Eq. (3.3.1), the rotation tensor can be expressed in the form

$$R_{iA} = x_{i,B} \left[(\underline{1} + 2\,\underline{E})^{-1/2} \right]_{BA}$$

$$= (\delta_{iB} + u_{i,B})(\delta_{BA} - E_{BA} + \frac{3}{2} E_{BC} E_{CA} - \ldots) \; . \qquad (4.1.18)$$

The Green finite strain tensor E_{AB} may be expressed with the aid of Eqs. (3.3.1), (3.3.2) and (4.1.3) as

$$E_{AB} = \frac{1}{2} (u_{A,B} + u_{B,A}) + \frac{1}{2} u_{i,A} u_{i,B} \quad , \qquad (4.1.19)$$

so that Eq. (4.1.18) may be rewritten as

$$R_{iA} = \delta_{iA} + \frac{1}{2} u_{i,A} - \frac{1}{2}\delta_{iB} u_{A,B} - \frac{1}{2} \delta_{iB} u_{K,B} u_{K,A}$$

$$+ \frac{3}{2} \delta_{iB} S_{BC} S_{CA} - u_{i,B} S_{BA} + \ldots \qquad (4.1.20)$$

Here,

$$S_{AB} = \frac{1}{2} (u_{A,B} + u_{B,A}) \qquad (4.1.21)$$

is defined as the infinitesimal strain. The transformation to the spatial form is accomplished through use of Eq. (4.1.16) to yield

$$R_{ia} = \delta_{ia} + \frac{1}{2} (\hat{u}_{i,a} - \hat{u}_{a,i})$$

$$+ \frac{1}{8} (3\hat{u}_{i,c} \hat{u}_{c,a} - \hat{u}_{i,c} \hat{u}_{a,c} - \hat{u}_{c,i} \hat{u}_{c,a} - \hat{u}_{c,i} \hat{u}_{a,c}) + \ldots \qquad (4.1.22)$$

The techniques presented in this section are sufficiently general to permit the evaluation of the spatial form of any quantity of interest.

4.2 Nonlinear Equations of Motion

The equations of motion for the center of mass and the internal coordinates were derived previously in Sections 3.5 and 3.7 under the restrictions of the dipole approximation. That is to say that the Lagrangian was first linearized in the displacements u^α before the equations of motion were derived. No guarantee exists that all linear terms would be present after linearization of these equations of motion. Neither could the full form of the nonlinear terms be written down from these equations. It is necessary therefore to derive these equations of motion valid to all orders before any linearization procedure be attempted. The magnetic equation of motion, Eq. (3.8.8) is already valid to all orders.

The equation of motion for the particle described by position $\vec{x}^\alpha(\vec{X},t)$ will be given by

$$\rho^\alpha \ddot{\vec{x}}^\alpha(\vec{X},t) = \vec{f}^\alpha(\vec{X},t) + \vec{f}^{\alpha e}(\vec{X},t) \quad , \tag{4.2.1}$$

where use of Eqs. (3.1.5), (3.1.6), (3.2.11), (3.2.17), (3.2.22), (3.2.38) and (3.2.40) has been made. Here the mechanical force $\vec{f}^\alpha$ and the electromagnetic force $\vec{f}^{\alpha e}$ are given respectively by

$$\vec{f}^{\alpha} = -\rho^{0} \frac{\partial \Sigma}{\partial \vec{x}^{\alpha}} + \rho^{0} \frac{\partial}{\partial X_{A}} \left(\frac{\partial \Sigma}{\partial \vec{x}^{\alpha}_{,A}} \right) , \qquad (4.2.2)$$

$$\vec{f}^{\alpha e} = q^{\alpha} [\vec{E}(\vec{x}^{\alpha},t) + \dot{\vec{x}}^{\alpha} \times \vec{B}(\vec{x}^{\alpha},t)]$$

$$+ \frac{\partial}{\partial \vec{x}^{\alpha}} \sum_{\beta} [\vec{m}^{T\beta} \cdot \vec{B}(\vec{x}^{\beta},t)] . \qquad (4.2.3)$$

The electromagnetic force $\vec{f}^{\alpha e}$ is now expanded in powers of the displacements u^{α}, defined by Eq.(3.2.6), so that the results are valid to all multipole orders:

$$\vec{f}^{\alpha e} = q^{\alpha} [\vec{E}(\vec{x} + \vec{u}^{\alpha}) + (\dot{\vec{x}} + \dot{\vec{u}}^{\alpha}) \times \vec{B}(\vec{x} + \vec{u}^{\alpha})]$$

$$+ \frac{\partial}{\partial \vec{x}^{\alpha}} \sum_{\beta} [\vec{m}^{T\beta} \cdot \vec{B}(\vec{x} + \vec{u}^{\beta})]$$

$$= q^{\alpha} \vec{E}(\vec{x}) + q^{\alpha}(\vec{u}^{\alpha} \cdot \nabla) \vec{E}(\vec{x})$$

$$+ \frac{1}{2} q^{\alpha} \vec{u}^{\alpha} \vec{u}^{\alpha} : \nabla\nabla\vec{E}(\vec{x}) + q^{\alpha}(\dot{\vec{x}} + \dot{\vec{u}}^{\alpha}) \times \vec{B}(\vec{x})$$

$$+ \dot{\vec{x}} \times (q^{\alpha} \vec{u}^{\alpha} \cdot \nabla) \vec{B}(\vec{x}) + q^{\alpha} \dot{\vec{u}}^{\alpha} \times (\vec{u}^{\alpha} \cdot \nabla) \vec{B}(\vec{x})$$

$$+ \frac{\partial}{\partial \vec{x}^{\alpha}} \sum_{\beta} \vec{m}^{T\beta} \cdot (\vec{B}(\vec{x}) + (\vec{u}^{\beta} \cdot \nabla) \vec{B}(\vec{x}))$$

$$+ \ldots \qquad (4.2.4)$$

We note that

$$\sum_{\alpha=1}^{N} \frac{\partial}{\partial x_j^{\alpha}} = \frac{\partial}{\partial x_j} \tag{4.2.5}$$

where Eqs. (3.2.7), (3.2.11) and (3.2.12) have been used. Thus, displacement invariance (see Sec. 3.3-A) yields

$$\sum_{\alpha=1}^{N} \frac{\partial \Sigma}{\partial \vec{x}^{\alpha}} = \frac{\partial \Sigma}{\partial \vec{x}} = 0 \quad . \tag{4.2.6}$$

Moreover,

$$\sum_{\alpha=1}^{N} \frac{\partial \Sigma}{\partial \vec{x}^{\alpha}_{,A}} = \frac{\partial \Sigma}{\partial \vec{x}_{,A}} \quad . \tag{4.2.7}$$

Summing Eq. (4.2.1) over the index α, we obtain the center of mass equation of motion:

$$\rho^0 \ddot{\vec{x}}(\vec{X},t) = \vec{f}(\vec{X},t) + \vec{f}^e(\vec{X},t) \quad , \tag{4.2.8}$$

where

$$\vec{f} \equiv \sum_{\alpha} \vec{f}^{\alpha} = \rho^0 \frac{\partial}{\partial X_A} \left(\frac{\partial \Sigma}{\partial \vec{x}_{,A}} \right) \quad , \tag{4.2.9}$$

and

$$f_i^e \equiv \sum_{\alpha} f_i^{\alpha e}$$

$$= (\vec{p} \cdot \nabla)\, E_i + (\dot{\vec{p}} \times \vec{B})_i + [\, \dot{\vec{x}} \times (\vec{p} \cdot \nabla)\vec{B}\,]_i$$

$$+ \sum_{\alpha} \vec{m}^{T\alpha} \cdot \vec{B}_{,i} + \sum_{\mu}' \vec{m}^{T\mu}\, \vec{y}^{\mu} : \nabla\vec{B}_{,i}$$

$$+ \sum_{\alpha} \frac{1}{2} q^{\alpha}\, \vec{u}^{\alpha}\, \vec{u}^{\alpha} : \nabla\nabla\, E_i$$

$$+ \sum_{\alpha} q^{\alpha}\, [\, \dot{\vec{u}}^{\alpha} \times (\vec{u}^{\alpha} \cdot \nabla)\, \vec{B}\,]_i + \ldots \qquad (4.2.10)$$

In the above, Eqs. (3.2.4), (3.2.29), (3.2.34), (3.5.2), (4.2.2), (4.2.4), (4.2.6) and (4.2.7) have been used. Multiplying Eq. (4.2.1) by $V^{\alpha\mu}, \mu \neq 0$, and summing over the index α, we obtain the internal coordinate equations of motion:

$$m^{\mu}\, \ddot{\vec{y}}^{\mu}(\vec{X},t) = \vec{f}^{\mu}(\vec{X},t) + \vec{f}^{\mu e}(\vec{X},t), \qquad (4.2.11)$$

where

$$\vec{f}^{\mu} \equiv \sum_{\alpha} V^{\alpha\mu}\, \vec{f}^{\alpha} = -\rho^0 \frac{\partial\Sigma}{\partial\vec{y}^{\mu}} + \rho^0 \frac{\partial}{\partial X_A} \left(\frac{\partial\Sigma}{\partial\vec{y}^{\mu}_{,A}} \right), \qquad (4.2.12)$$

and

$$f_i^{\mu e} \equiv \sum_{\alpha} V^{\alpha\mu}\, f_i^{\alpha e}$$

$$= \{\, q^{\mu}\, \vec{E} + \dot{\vec{p}}^{\mu} \times \vec{B} + [\, \vec{p}^{\mu} \cdot \nabla\,]\, \vec{E}$$

$$+ \frac{1}{2} [\underline{q}^{\mu} : \nabla\nabla] \vec{E} + \dot{\vec{x}} \times [\vec{p}^{\mu} \cdot \nabla] \vec{B} \}_i$$

$$+ \vec{m}^{T\mu} \cdot \vec{B}_{,i} + \ldots \qquad (4.2.13)$$

In the above, Eqs. (3.2.6)-(3.2.8), (3.2.16),(3.2.32)-(3.2.34), (4.2.2) and (4.2.4) have been used, and we define

$$\vec{p}^{\mu} \equiv \sum_{\alpha} q^{\alpha} \vec{u}^{\alpha} V^{\alpha\mu} = \sum_{\nu}{}' q^{\mu\nu} \vec{y}^{\nu} , \qquad (4.2.14)$$

$$q^{\mu\nu} \equiv \sum_{\alpha} q^{\alpha} V^{\alpha\mu} V^{\alpha\nu} , \qquad (4.2.15)$$

$$\underline{q}^{\mu} \equiv \sum_{\alpha} q^{\alpha} \vec{u}^{\alpha} \vec{u}^{\alpha} V^{\alpha\mu}$$

$$= \sum_{\nu\lambda}{}' q^{\mu\nu\lambda} \vec{y}^{\nu} \vec{y}^{\lambda} , \qquad (4.2.16)$$

$$q^{\mu\nu\lambda} \equiv \sum_{\alpha} q^{\alpha} V^{\alpha\mu} V^{\alpha\nu} V^{\alpha\lambda} . \qquad (4.2.17)$$

In the absence of wave vector dispersion, the second term in Eq. (4.2.12) vanishes. We now proceed to obtain the linear equations of motion.

4.3 Linearized Center of Mass Equation

The force in the center of mass Eq. (4.2.9) can be written more explicitly as

$$\frac{f_i}{\rho^0} = \frac{\partial}{\partial X_A} \{ x_{i,B} \frac{\partial\Sigma}{\partial E_{AB}} + \sum_{\mu=1}^{N-1} y_j^{\mu} \frac{\partial\Sigma}{\partial \Lambda_B^{\mu}} \frac{\partial R_{jB}}{\partial x_{i,A}}$$

$$+ \sum_{\alpha=1}^{N} m_j^{T\alpha} \frac{\partial\Sigma}{\partial\Gamma_B^\alpha} \frac{\partial R_{jB}}{\partial x_{i,A}} + \sum_{\alpha=1}^{N} m_{j,C}^{\alpha} \frac{\partial\Sigma}{\partial\Gamma_{B;C}^\alpha} \frac{\partial R_{jB}}{\partial x_{i,A}} \} \quad , \tag{4.3.1}$$

where Eqs. (3.3.1)-(3.3.3), (3.3.6) and (3.3.22) have been utilized. With the aid of the results of Section 4.1 and Eqs. (3.3.23) and (3.10.21) the linear center of mass force, $\hat{f}_i^L$ can be determined as

$$\hat{f}_i^L = J_{icda} \, \hat{u}_{c,da} + J_{ijk}^{\mu} \, \hat{y}_{j,k}^{\mu}$$

$$+ J_{ijk}^{\alpha} \, \hat{m}_{j,k}^{\alpha} + J_{ijkl}^{\alpha} \, \hat{m}_{j,kl}^{\alpha} \quad , \tag{4.3.2}$$

where

$$J_{icda} \equiv {}^{(0100)}K_{da} \, \delta_{ic} + 2\,{}^{(0200)}K_{aicd} + {}^{(0110)}K_{ail}^{\alpha} \, m_j^{S\alpha} \, \delta_{c[j} \, \delta_{l]d}$$

$$+ 2\,{}^{(0020)}K_{bl}^{\alpha\beta} \, \delta_{i[j} \, \delta_{b]a} \, \delta_{c[k} \, \delta_{l]d} \, m_j^{S\alpha} \, m_k^{S\beta}$$

$$+ {}^{(0110)}K_{cdb}^{\alpha} \, \delta_{i[j} \, \delta_{b]a} \, m_j^{S\alpha}$$

$$+ \frac{1}{8} \, {}^{(0010)}K_b^{\alpha} \, m_j^{S\alpha} \{ \delta_{id} \, (3\delta_{bc} \, \delta_{ja} - \delta_{jc} \, \delta_{ba})$$

$$- 2 \, \delta_{ij} \, \delta_{c(a} \, \delta_{b)d}$$

$$+ \delta_{ib} (3 \, \delta_{ac} \, \delta_{jd} - \delta_{jc} \, \delta_{ad}) \} \quad , \tag{4.3.3}$$

$$J^{\mu}_{ijk} \equiv {}^{(1100)}K^{\mu}_{jki} + {}^{(1010)}K^{\mu\alpha}_{jb}\, m^{S\alpha}_{l}\, \delta_{i[l}\, \delta_{b]k} \quad , \tag{4.3.4}$$

$$J^{\alpha}_{ijk} \equiv {}^{(0110)}K^{\alpha}_{kij} + {}^{(0010)}K^{\alpha}_{b}\, \delta_{i[j}\, \delta_{b]k}$$

$$+ {}^{(0020)}K^{\beta\alpha}_{bj}\, m^{S\beta}_{l}\, \delta_{i[l}\, \delta_{b]k} \quad , \tag{4.3.5}$$

$$J^{\alpha}_{ijkl} \equiv {}^{(0101)}K^{\alpha}_{lijk} + {}^{(0011)}K^{\beta\alpha}_{bjk}\, m^{S\beta}_{a}\, \delta_{i[a}\, \delta_{b]l}$$

$$+ {}^{(0001)}K^{\alpha}_{bk}\, \delta_{i[j}\, \delta_{b]l} \quad . \tag{4.3.6}$$

In the above, we have used brackets to denote an antisymmetrization and parentheses to denote a symmetrization on the enclosed indices:

$$\delta_{c[j}\, \delta_{l]d} \equiv \frac{1}{2}\,(\, \delta_{cj}\, \delta_{ld} - \delta_{cl}\, \delta_{jd}\,) \quad , \tag{4.3.7}$$

$$\delta_{c(a}\, \delta_{b)d} \equiv \frac{1}{2}\,(\, \delta_{ca}\, \delta_{bd} + \delta_{cb}\, \delta_{ad}\,) \quad . \tag{4.3.8}$$

In a similar fashion, the <u>linear</u> part of the electromagnetic force, Eq. (4.2.10), is given by

$$(\, \hat{f}^{e}_{i}\,)^{L} = (\, \frac{\partial \hat{\vec{p}}}{\partial t} \times \vec{B}^{S}\,)_{i} + \sum_{\alpha} m^{S\alpha}_{k}\, B^{V}_{k,i} \quad , \tag{4.3.9}$$

where $\vec{B}^{S}$ is the macroscopic spontaneous magnetic field assumed here to be homogeneous as in a boundless magnetic medium. The varying or induced magnetic field present in Eq. (4.3.9) stands for $\vec{B}^{V} = \vec{B} - \vec{B}^{S}$ but will henceforth be denoted simply by $\vec{B}$. The linearized equation for the center of mass can now be written down:

$$\rho^0 \frac{\partial^2 \hat{u}_i}{\partial t^2} - \left(\frac{\partial \hat{\vec{\phi}}}{\partial t} \times \vec{B}^S \right)_i - \sum_\alpha m_K^{S\alpha} B_{K,i}$$

$$- J_{icda} \hat{u}_{c,aa} - J^{\mu}_{ijK} \hat{y}^{\mu}_{j,K} - J^{\alpha}_{ijK} \hat{m}^{\alpha}_{j,K}$$

$$- J^{\alpha}_{ijKl} \hat{m}^{\alpha}_{j,Kl} = G_i \quad . \qquad (4.3.10)$$

Here the nonlinear force G_i is given by

$$G_i = (f_i - f_i^L) + (f_i^e - (f_i^e)^L) - \rho^0 \left(\ddot{x}_i - \frac{\partial^2 \hat{u}_i}{\partial t^2} \right) , \qquad (4.3.11)$$

and can be determined to any order of nonlinearity as needed for any particular application.

4.4 Linearized Equation for Internal Coordinates

The force in the internal coordinate equation (4.2.12) can be written more explicitly as

$$f_i^\mu = -\rho^0 \frac{\partial \Sigma}{\partial y_i^\mu} = -R_{iA} \, \rho^0 \frac{\partial \Sigma}{\partial \Lambda_A^\mu}$$

$$= -R_{iA} \left[2 \, {}^{(2000)}K^{\mu\nu}_{AB} \Lambda^\nu_B + {}^{(1100)}K^{\mu}_{ABC} E_{BC} \right.$$

$$+ {}^{(1010)}K^{\mu\alpha}_{AB}\, \Gamma^{\alpha}_{B} + {}^{(1001)}K^{\mu\alpha}_{ABC}\, \Gamma^{\alpha}_{B;C}$$

$$+ \ldots] \quad , \tag{4.4.1}$$

where use of Eqs. (3.3.3), (3.3.23) and (3.7.2) has been made. With the aid of the results of Section 4.1 and Eq. (3.3.22), the <u>linear</u> internal coordinate force $[\hat{f}^{\mu}_{i}]^{L}, \mu \neq 0$, can be determined as

$$[\, \hat{f}^{\mu}_{i}\,]^{L} = -\, 2\, {}^{(2000)}K^{\mu\nu}_{ij}\, \hat{y}^{\nu}_{j} + \bar{K}^{\mu}_{ijk}\, \hat{u}_{j,k}$$

$$+ {}^{(1010)}K^{\mu\alpha}_{ij}\, \hat{m}^{\alpha}_{j} + {}^{(1001)}K^{\mu\alpha}_{ijk}\, \hat{m}^{\alpha}_{j,k} \quad , \tag{4.4.2}$$

where

$$\bar{K}^{\mu}_{ijk} \equiv {}^{(1100)}K^{\mu}_{ijk} + {}^{(1010)}K^{\mu\alpha}_{il}\, m^{S\alpha}_{a}\, \delta_{j[a}\, \delta_{l]k} \quad . \tag{4.4.3}$$

In a similar fashion, the <u>linear</u> part of the electromagnetic force on the μ-th degree of freedom [Eq. (4.2.13)] is given by

$$[\, \hat{f}^{e\mu}_{i}\,]^{L} = q^{\mu}\, E_{i} + q^{\mu} \left(\frac{\partial \hat{\vec{u}}}{\partial t} \times \vec{B}^{S} \right)_{i}$$

$$+ \left(\frac{\partial \hat{\vec{p}}^{\mu}}{\partial t} \times \vec{B}^{S} \right)_{i} + m^{\mu S}_{k}\, B_{k,i} \quad . \tag{4.4.4}$$

The linearized equation for the internal coordinate degrees of freedom can now be written down:

$$m^{\mu} \frac{\partial^2 \hat{y}_i^{\mu}}{\partial t^2} - q^{\mu} E_i - q^{\mu} \left(\frac{\partial \hat{\vec{u}}}{\partial t} \times \vec{B}^S \right)_i - \left(\frac{\partial \hat{\vec{p}}^{\mu}}{\partial t} \times \vec{B}^S \right)_i$$

$$- m_K^{\mu S} B_{K,i} + 2^{(2000)} K_{ij}^{\mu\nu} \hat{y}_j^{\nu} + \bar{K}_{ijK}^{\mu} \hat{u}_{j,K}$$

$$+ {}^{(1010)}K_{ij}^{\mu\alpha} \hat{m}_j^{\alpha} + {}^{(1001)}K_{ijK}^{\mu\alpha} \hat{m}_{j,K}^{\alpha} = G_i^{\mu} \quad . \qquad (4.4.5)$$

Here the nonlinear force G_i^{μ} is given by

$$G_i^{\mu} = \left(f_i^{\mu} - (f_i^{\mu})^L \right) + \left(f_i^{\mu e} - (f_i^{\mu e})^L \right) - m^{\mu} \left(\ddot{y}_i^{\mu} - \frac{\partial^2 \hat{y}_i^{\mu}}{\partial t^2} \right) \quad . \qquad (4.4.6)$$

This nonlinear force can be easily determined to any order of nonlinearity to fit a particular application. An example will be given in the next chapter in relation to light scattering.

It is interesting to note that the linear terms present in the center of mass Eq. (4.3.10), could have been obtained from the corresponding equation in the dipole approximation Eq. (3.5.4). Moreover, all the linear terms present in the internal coordinate equation, Eq. (4.4.5), could also have been obtained from the corresponding dipole approximation Eq. (3.7.1).

4.5 Linearized Magnetic Equation of Motion

The effective magnetic field in the magnetic equation of motion, as given through Eqs. (3.8.5) and (3.8.8), may be written more explicitly as

$$B_i^{\alpha\,\mathrm{eff}} = B_i + (\vec{u}^{\alpha} \cdot \nabla)\, B_i + \ldots - \rho^0 R_{iA} \frac{\partial \Sigma}{\partial \Gamma_A^{\alpha}}$$

$$+ \frac{\partial}{\partial X_A} \left[\rho^0 R_{iB} \frac{\partial \Sigma}{\partial \Gamma_{B;A}^{\alpha}} \right] \quad . \qquad (4.5.1)$$

In the above, the magnetic field $\vec{B}(\vec{x}^{\alpha},t)$ has been expanded about the center of mass position $\vec{x}$, and Eqs. (3.3.6) and (3.3.22) have been used. Employing Eqs. (3.3.23) and (3.8.9) and the results of Section 4.1, the linearized magnetic equation of motion may be written:

$$\frac{1}{\gamma^{\alpha}} \frac{\partial \hat{m}_i^{\alpha}}{\partial t} - (\hat{m}^{S\alpha} \times \vec{B})_i - \epsilon_{irt}\, \hat{m}_r^{\alpha} \left(B_t^S - {}^{(0010)}K_t^{\alpha}\right)$$

$$+ 2\epsilon_{irt}\, {}^{(0020)}K_{tj}^{\alpha\beta}\, \hat{m}_r^{S\alpha}\, \hat{m}_j^{\beta} + \epsilon_{irt}\, \hat{m}_r^{S\alpha}\, {}^{(1010)}K_{jt}^{\mu\alpha}\, \hat{y}_j^{\mu}$$

$$- \epsilon_{irt}\, \hat{m}_r^{S\alpha} \left({}^{(0011)}K_{jta}^{\beta\alpha} - {}^{(0011)}K_{tja}^{\alpha\beta} \right) \hat{m}_{j,a}^{\beta}$$

$$- \epsilon_{irt}\, \hat{m}_r^{S\alpha}\, {}^{(1001)}K_{jta}^{\mu\alpha}\, \hat{y}_{j,a}^{\mu}$$

$$- 2\ \epsilon_{irt}\ \hat{m}_r^{S\alpha}\ {}^{(0002)}K^{\alpha\beta}_{tajd}\ \hat{m}^{\beta}_{j,da}$$

$$+ \bar{K}^{\alpha}_{ijk}\ \hat{u}_{j,k} - \bar{K}^{\alpha}_{ijkl}\ \hat{u}_{j,kl}$$

$$= \zeta^{\alpha}_{i}\ , \qquad (4.5.2)$$

where

$$\bar{K}^{\alpha}_{ijk} \equiv \epsilon_{irt}\ \hat{m}_r^{S\alpha}\ [\ {}^{(0010)}K^{\alpha}_{a}\ \delta_{j[t}\ \delta_{a]k}$$

$$+ 2\ {}^{(0020)}K^{\alpha\beta}_{tb}\ \delta_{j[l}\ \delta_{b]k}\ \hat{m}^{S\beta}_{l}$$

$$+ {}^{(0110)}K^{\alpha}_{bct}\ \delta_{j(b}\ \delta_{c)k}\ ]\ ,$$

$$\bar{K}^{\alpha}_{ijkl} = \epsilon_{irt}\ \hat{m}_r^{S\alpha}\ [\ {}^{(0001)}K^{\alpha}_{bl}\ \delta_{j[t}\ \delta_{b]k} + {}^{(0101)}K^{\alpha}_{catl}\ \delta_{j[c}\ \delta_{a]k}$$

$$+ {}^{(0001)}K^{\beta\alpha}_{ctl}\ \hat{m}^{S\beta}_{a}\ \delta_{j[a}\ \delta_{c]k}\ ]\ . \qquad (4.5.3)$$

$\vec{\zeta}^{\alpha}$ represents the nonlinear torque on the sublattice magnetization $\vec{m}^{T\alpha}$. This quantity is not written out explicitly, but can be found readily to any order of nonlinearity.

We postpone until the next chapter the determination of the electromagnetic wave equation and its linearization.

V. APPLICATION TO LIGHT SCATTERING

5.1 Simplifications to the Problem

The specific application we have in mind is the nonlinear interaction of light at optical frequencies with magnon-phonon-photon mixed modes hereafter called magneto-polaritons. We restrict ourselves to materials which are magnetic insulators and which (if all other mechanical degrees of freedom are suppressed) possess magnon dispersion curves exhibiting an energy gap in the spectrum of the order of radio frequencies at the zone center. We will be concerned with scattered radiation having Stokes and anti-Stokes lines shifted in frequency from the incoming laser frequency by at least an amount equal to these radio frequencies. Therefore, the elastic degree of freedom (acoustic phonons) which is important only at much lower frequency shifts (Brillouin scattering) will be ignored in this analysis.

As a further simplification, all spatial derivatives in the mechanical equations of motion will be set equal to zero. This places no restriction on our method since we could handle these terms simply by considering Fourier components in $\vec{k}$ space. It is just a convenience which will ease the calculational effort. It should be noted that inclusion of spatial derivatives of the field variables will manifest itself in nonlocal integral expressions relating polarization and magnetization to the electromagnetic fields.

Force terms arising from electric quadrupole, and higher multipole moments will be assumed negligible in comparison to the other interactions. Finally, correction terms which arose because of the transformation of time derivatives from the material to the spatial frame will be assumed negligible and discarded here. This last simplification has been justified for the dielectric case by Nelson and Lax.[31]

5.2 Linear Constitutive Relations

In Eqs. (4.4.6) and (4.5.2), all mechanical and field variables are evaluated at the position $\vec{x}$ whereas the fields in the Maxwell-Lorentz equations (3.4.3) and (3.4.11) are evaluated at the point $\vec{z}$. Since the electric and magnetic fields which enter the mechanical equations are $\vec{E}(\vec{z})$ and $\vec{B}(\vec{z})$ evaluated at $\vec{z} = \vec{x}$ and the polarization and magnetization which enter the Maxwell-Lorentz equations involve $\vec{y}^{\mu}(\vec{x})$ and $\vec{m}^{\alpha}(\vec{x})$ evaluated at $\vec{x} = \vec{z}$, the two variables $\vec{x}$ and $\vec{z}$ can be identified for purposes of solving the coupled equations. They are both spatial variables and shall be denoted by $\vec{z}$

from here on.

Since we will be considering individual frequency components of the fields, we will take Fourier components of each field that appears in our equations:

$$\hat{y}_i^{\mu}(\vec{z},t) = y_i^{\mu}(\vec{z},\omega)\, e^{-i\omega t} \quad , \tag{5.2.1}$$

$$\hat{m}_i^{\alpha}(\vec{z},t) = m_i^{\alpha}(\vec{z},\omega)\, e^{-i\omega t} \quad . \tag{5.2.2}$$

After discarding terms containing spatial derivatives of the elastic degree of freedom u, the internal coordinate equation (4.4.45) may be rewritten with the help of Eqs. (5.2.1) and (5.2.2) as

$$[2\,{}^{(2000)}K_{ij}^{\mu\nu} - m^{\mu}\,\omega^2\,\delta^{\mu\nu}\delta_{ij} + i\omega\, q^{\mu\nu}\,\epsilon_{ijk}\,B_k^S]\, y_j^{\nu}$$

$$= q^{\mu}\,E_i - {}^{(1010)}K_{ij}^{\mu\alpha}\, m_j^{\alpha} + G_i^{\mu} \quad . \tag{5.2.3}$$

Let us now introduce a mechanical admittance matrix $\underline{Y}$ defined through the relation

$$\epsilon_0\, Y_{ij}^{\mu\nu}(\omega)\, [2\,{}^{(2000)}K_{jk}^{\nu\lambda} - m^{\mu}\,\omega^2\,\delta^{\nu\lambda}\delta_{jk} + i\omega\, q^{\nu\lambda}\epsilon_{jkl}B_l^S] = \delta^{\mu\lambda}\delta_{ik} \quad . \tag{5.2.4}$$

Combination of Eqs. (5.2.3) and (5.2.4) yields an expression for $\vec{y}^{\mu}$:

$$y_i^{\mu} = \epsilon_0 \Upsilon_{ij}^{\mu\nu} [q^{\nu} E_j - {}^{(1010)}K_{jl}^{\nu\beta} m_l^{\beta} + G_j^{\nu}] \quad . \qquad (5.2.5)$$

The last term in Eq. (5.2.5) will lead to a nonlinear polarization, a topic we will return to in Section 5.8. For now we set the last term equal to zero since we are only interested in linear relations.

In similar fashion the magnetic equation of motion, Eq. (4.5.2) may be rewritten as

$$\sum_{\beta} [- \frac{i\omega}{\gamma^{\alpha}} \delta_{ij} \delta^{\alpha\beta} - \epsilon_{ijt}(B_t^S - {}^{(0010)}K_t^{\beta}) \delta^{\alpha\beta}$$

$$+ 2 \epsilon_{ikl} m_k^{S\alpha} \, {}^{(0020)}K_{lj}^{\alpha\beta}] m_j^{\beta}$$

$$= (\vec{m}^{S\alpha} \times \vec{B})_i - \epsilon_{irt} m_r^{S\alpha} \, {}^{(1010)}K_{jt}^{\mu\alpha} y_j^{\mu} , \qquad (5.2.6)$$

where the nonlinear term ξ^{α} as in Eq. (4.5.2) has been set to zero. Eliminating $\vec{y}^{\mu}$ from Eq. (5.2.6) with the aid of Eq. (5.2.5), we obtain

$$\sum_{\beta} [- \frac{i\omega}{\gamma^{\alpha}} \delta_{ij} \delta^{\alpha\beta} - \epsilon_{ijt}(B_t^S - {}^{(1010)}K_t^{\beta}) \delta^{\alpha\beta} + 2 \epsilon_{ikl} m_k^{S\alpha} \, {}^{(0020)}K_{lj}^{\alpha\beta}$$

$$- \sum_{\mu,\lambda} \epsilon_{irt} m_r^{S\alpha} \, {}^{(1010)}K_{lt}^{\mu\alpha} \epsilon_0 \Upsilon_{lc}^{\mu\lambda} \, {}^{(1010)}K_{cj}^{\lambda\beta}] m_j^{\beta} \qquad (5.2.7)$$

$$= (\vec{m}^{S\alpha} \times \vec{B})_i - \sum_{\mu,\lambda} \epsilon_{irt} m_r^{S\alpha} \, {}^{(1010)}K_{lt}^{\mu\alpha} \epsilon_0 \Upsilon_{lc}^{\mu\lambda} q^{\lambda} E_c \; .$$

Let us introduce a magnetic admittance matrix $\underline{I}$ with the property

$$\frac{1}{\mu_0} I_{ij}^{\alpha\beta}(\omega) \, [- \frac{i\omega}{\gamma^\beta} \delta_{jk} \, \delta^{\beta\xi} - \epsilon_{jkl}(B_l^S - {}^{(0010)}K_l^\beta)\,\delta^{\beta\xi}$$

$$+ 2 \, \epsilon_{jal} \, m_a^{S\beta} \, {}^{(0020)}K_{lk}^{\beta\xi} - \epsilon_{jrt} m_r^{S\beta} \, {}^{(1010)}K_{lt}^{\mu\beta} \, \epsilon_0 \gamma_{lc}^{\mu\lambda} \, {}^{(1010)}K_{ck}^{\lambda\xi}]$$

$$= \delta^{\alpha\xi} \, \delta_{ik} \quad . \tag{5.2.8}$$

Combining Eqs. (5.2.7) and (5.2.8) will yield an expression for the magnetization variation $\vec{m}^\alpha$:

$$m_i^\alpha = \frac{1}{\mu_0} I_{ij}^{\alpha\beta} \, \epsilon_{jlk} \, m_l^{S\beta} \, B_k$$

$$+ [- \frac{\epsilon_0}{\mu_0} I_{ij}^{\alpha\beta} \epsilon_{jrt} \, m_r^{S\beta} \, {}^{(1010)}K_{lt}^{\mu\beta} \, \gamma_{lk}^{\mu\lambda} \, q^\lambda] \, E_k \quad . \tag{5.2.9}$$

Using the definition of the magnetization, Eq. (3.2.35), and remembering that the Jacobian $J = 1$ after ignoring the elastic degree of freedom [see Eq. (4.1.10)], we obtain the linear constitutive relation for the magnetization:

$$\vec{M} - \vec{M}^S = (\underline{1} - \underline{K}_m^{-1}) \cdot \frac{\vec{B}}{\mu_0} + \underline{x}_{me} \cdot \epsilon_0 \vec{E} \quad , \tag{5.2.10}$$

where

$$(\underline{1} - \underline{K}_m^{-1})_{i\kappa} \equiv \sum_{\alpha,\beta} I_{ij}^{\alpha\beta} \, \epsilon_{jl\kappa} \, m_l^{S\beta} \quad , \tag{5.2.11}$$

$$(\underline{x}_{me})_{i\kappa} \equiv -\frac{1}{\mu_0} \sum_{\substack{\alpha,\beta,\\ \mu,\lambda}} I_{ij}^{\alpha\beta} \, \epsilon_{jrt} \, m_r^{S\beta} \, q^{\lambda} \, {}^{(1010)}K_{lt}^{\mu\beta} \, \Upsilon_{l\kappa}^{\mu\lambda} \quad . \tag{5.2.12}$$

Here $\underline{K}_m$ is the magnetic permeability tensor and $\underline{x}_{me}$ is one of the two magneto-electric tensors which depends very strongly on the material coefficient, ${}^{(1010)}K^{\mu\beta}$, measuring the strength of the spin-phonon coupling.

Eliminating $\vec{m}^{\beta}$ from Eq. (5.2.5) by use of Eq. (5.2.9) and after setting the nonlinear term to zero, we obtain the expression

$$y_i^{\mu} = [\epsilon_0 \, q^{\lambda} \, \Upsilon_{i\kappa}^{\mu\lambda} + \frac{\epsilon_0^2}{\mu_0} \Upsilon_{ic}^{\mu\lambda} \, {}^{(1010)}K_{cl}^{\lambda\alpha} I_{lj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta} \, {}^{(1010)}K_{at}^{\nu\beta} q^{\xi} \Upsilon_{a\kappa}^{\nu\xi}] \, E_{\kappa}$$

$$+ [-\frac{\epsilon_0}{\mu_0} \Upsilon_{ic}^{\mu\lambda} \, {}^{(1010)}K_{cl}^{\lambda\alpha} \, I_{lj}^{\alpha\beta} \, m_a^{S\beta} \, \epsilon_{ja\kappa}] \, B_{\kappa} \quad . \tag{5.2.13}$$

Using the definition of the polarization, Eq. (3.4.2), the linear constitutive relation for the polarization is found to be

$$\vec{P} = \epsilon_0 \, \underline{x}_e \cdot \vec{E} + \underline{x}_{em} \cdot \frac{\vec{B}}{\mu_0}$$

$$= \epsilon_0(\underline{\kappa}_e - \underline{1}) \cdot \vec{E} + \underline{x}_{em} \cdot \vec{B}/\mu_0 \quad , \tag{5.2.14}$$

where

$$(\underline{x}_e)_{i\kappa} \equiv \sum_{\mu,\nu} q^\mu \, \gamma^{\mu\nu} \, q^\nu$$

$$+ \sum_{\substack{\mu,\nu,\xi,\lambda \\ \alpha,\beta}} q^\mu \frac{\epsilon_0}{\mu_0} \gamma_{ic}^{\mu\xi(1010)} K_{cl}^{\xi\alpha} I_{lj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta(1010)} K_{at}^{\nu\beta} \gamma_{a\kappa}^{\nu\lambda} q^\lambda \quad , \tag{5.2.15}$$

$$(\underline{x}_{em})_{i\kappa} \equiv - \epsilon_0 \sum_{\substack{\mu,\lambda \\ \alpha,\beta}} q^\mu \gamma_{ic}^{\mu\lambda(1010)} K_{cl}^{\lambda\alpha} \, I_{lj}^{\alpha\beta} \, m_r^{S\beta} \, \epsilon_{jr\kappa} \quad . \tag{5.2.16}$$

Here $\underline{\kappa}_e$ is the dielectric tensor related to the electric susceptibility $\underline{x}_e$ in the usual way:

$$\underline{\kappa}_e \equiv \underline{1} + \underline{x}_e \ . \tag{5.2.17}$$

Also, $\underline{x}_{em}$ is the other magneto-electric tensor which again is seen to depend on the spin-phonon coupling.

The second term on the righthand side of Eq. (5.2.15) is a magnetic correction to the electric susceptibility. It is interesting to note that because of the presence of the spontaneous sublattice magnetizations $m_r^{S\alpha}$ in this term, the electric susceptibility should show a strong temperature dependence for magnetic dielectrics at low temperatures.

5.3 The Driven Wave Equation

By the elimination of the magnetic field between Eqs. (3.4.3) and (3.4.11), an equation for the electric field is obtained in the form

$$\nabla \times (\nabla \times \vec{E}) + \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = - \mu_0 \frac{\partial \vec{j}}{\partial t} \quad . \qquad (5.3.1)$$

Under the approximations made in Section 5.1, the current density $\vec{j}$ is given by

$$\vec{j} = \frac{\partial \vec{P}}{\partial t} + \nabla \times \vec{M} \quad , \qquad (5.3.2)$$

where Eqs. (3.4.4) and (3.4.5) have been used. Equation (5.3.1) is now separated into linear and nonlinear components by writing

$$\vec{j} = \vec{j}^L + \vec{j}^{NL} \quad , \qquad (5.3.3)$$

$$\vec{j}^L = \frac{\partial \vec{P}^L}{\partial t} + \nabla \times \vec{M}^L \quad . \qquad (5.3.4)$$

Substituting in the linear constitutive relations, Eqs. (5.2.10) and (5.2.14), into Eqs. (5.3.1), (5.3.3) and (5.3.4), the driven electric field wave equation is obtained in which all linear properties of the electromagnetic medium are contained on the left-hand side and all nonlinear properties are displayed on the right-hand side:

$$\nabla \times [\underline{k}_m^{-1} \cdot (\nabla \times \vec{E})] + \frac{1}{c^2} \underline{\kappa}_e \cdot \frac{\partial^2 \vec{E}}{\partial t^2}$$

$$+ \frac{1}{c^2} \nabla \times [\underline{x}_{me} \cdot \frac{\partial \vec{E}}{\partial t}] - \underline{x}_{em} \cdot (\nabla \times \frac{\partial \vec{E}}{\partial t})$$

$$= - \mu_0 \frac{\partial \vec{j}^{NL}}{\partial t} \quad . \qquad (5.3.5)$$

In the above, we understand that $\frac{\partial}{\partial t} = -i\,\omega$. If we use the abbreviated notation

$$\vec{\kappa} = -i\,\nabla \quad , \qquad (5.3.6)$$

the contribution of each single frequency ω in $\vec{j}^{NL}$ to the output field $\vec{E}$ can be obtained by solving

$$\underline{\alpha}(\vec{\kappa},\omega) \cdot \vec{E} = \vec{P}/\epsilon_0 \quad , \qquad (5.3.7)$$

where we define

$$\underline{\alpha}(\vec{\kappa},\omega) \cdot \vec{E} \equiv - n^2\, \hat{s} \times [\underline{\kappa}_m^{-1} \cdot (\hat{s} \times \vec{E})] - \underline{\kappa}_e \cdot \vec{E}$$

$$+ \frac{n}{c}\, \hat{s} \times [\underline{x}_{me} \cdot \vec{E}] - cn\, \underline{x}_{em} \cdot (\hat{s} \times \vec{E}) \quad , \qquad (5.3.8)$$

$$\hat{s} \equiv \frac{\vec{\kappa}}{\kappa} \; , \; \kappa \equiv |\vec{\kappa}| \; , \; n = \frac{c\kappa}{\omega} \quad , \qquad (5.3.9)$$

$$\vec{P} \equiv \vec{P}^{NL} - \frac{n}{c}\, \hat{s} \times \vec{M}^{NL} \quad . \qquad (5.3.10)$$

The form of the wave equation given by Eq. (5.3.7) is particularly suitable when the nonlinear current density is decomposed into Fourier components in time and plane waves in space.

In order to solve the wave equation (5.3.5), three simplifications will be made:

1 The materials we are dealing with have weak magneto-electric effects. Thus, the last two terms on the left-hand side of Eq. (5.3.5) will be considered to be much smaller than the other linear terms and, hence, will be discarded.

2 The dielectric tensor $\underline{\kappa}_e$, and the magnetic permeability tensor $\underline{\kappa}_m$ are real and symmetric.

3 The component $\nabla \times \vec{M}^{NL}$ of the nonlinear current density will be discarded in keeping with the simplications of Section 5.1 ($\vec{M}^{NL}$ can be shown to be a function of $\vec{y}^{\mu}$ and $\vec{m}^{\alpha}$; hence, $\nabla \times \vec{M}^{NL}$ is expressible in terms of spatial derivatives of $\vec{y}^{\mu}$ and $\vec{m}^{\alpha}$ which we have been discarding).

As a result of (1) and (3), Eq. (5.3.5) can be rewritten for the case when the electric field is being driven by a single frequency component of the nonlinear polarization as

$$\underline{\alpha}(-i\nabla,\omega) \cdot \vec{E}(\vec{z},\omega) = \vec{P}^{NL}(\vec{z})\, e^{-i\omega t}/\epsilon_0 \quad , \qquad (5.3.11)$$

where

$$\underline{\alpha}(-i\nabla,\omega) \cdot \vec{E}(\vec{z},\omega)$$

$$= \frac{c^2}{\omega^2} \nabla \times [\underline{K}_m \cdot (\nabla \times \vec{E})] - \underline{K}_e \cdot \vec{E} \; . \qquad (5.3.12)$$

For future convenience, we make a change of variables by using the following transformations:

$$\vec{E} \equiv \underline{k}_m^{-1/2}\, \vec{F} \quad , \qquad (5.3.13)$$

$$\nabla_{\vec{z}} \equiv \underline{k}_m^{-1/2}\, \nabla_{\vec{y}} \quad , \quad \vec{z} \equiv \underline{k}_m^{1/2}\, \vec{y} \quad , \qquad (5.3.14)$$

where $\underline{k}_m^{1/2}$ is the symmetric square root of $\underline{K}_m$. Substitution of Eqs. (5.3.13) and (5.3.14) and use of the mathematical theorem

$$\underline{D} \cdot [\, (\underline{D} \cdot \vec{A}) \times (\underline{D} \cdot \vec{B})\,] = (\det \underline{D})\, \vec{A} \times \vec{B} \quad , \qquad (5.3.15)$$

where $\underline{D}$ is a dyadic and $\vec{A}$ and $\vec{B}$ are vectors, enable us to express Eq. (5.3.11) in the form

$$\underline{\beta}(-i\nabla_{\vec{y}},\omega) \cdot \vec{F}(\vec{y},\omega) = \vec{P}^{eff}(\vec{y})\, e^{-i\omega t}/\epsilon_0 \; . \qquad (5.3.16)$$

Here we define

$$\underline{B}(-i\nabla_{\vec{y}},\omega) \cdot \vec{F}(\vec{y},\omega)$$

$$\equiv \frac{c^2}{\omega^2 \det(\underline{\kappa}_m)} \nabla_{\vec{y}} \times (\nabla_{\vec{y}} \times \vec{F}) - \underline{\kappa}_e^{eff} \cdot \vec{F} \quad , \qquad (5.3.17)$$

where

$$\underline{\kappa}_e^{eff} \equiv \underline{\kappa}_m^{-1/2} \cdot \underline{\kappa}_e \cdot \underline{\kappa}_m^{-1/2} \quad , \qquad (5.3.18)$$

and

$$\vec{P}^{eff}(\vec{y}) \equiv \underline{\kappa}_m^{-1/2} \cdot \vec{P}^{NL}(\vec{y}) \quad . \qquad (5.3.19)$$

Since a point $\vec{z}$ is mapped in real space into another point $\vec{y}$ through the transformation Eq. (5.3.14), we expect a point $\vec{\kappa}$ in reciprocal space to be transformed to a point $\vec{l}$ in a transformed reciprocal space. The connection is given by the relation

$$\vec{\kappa} \cdot \vec{z} = \vec{l} \cdot \vec{y} \quad , \qquad (5.3.20)$$

which implies with the help of Eq. (5.3.14) that

$$\vec{l} = \vec{\kappa} \cdot \underline{\kappa}_m^{1/2} = \underline{\kappa}_m^{1/2} \cdot \vec{\kappa} \quad , \qquad (5.3.21)$$

where the symmetric property of $\underline{\kappa}_m^{1/2}$ has been utilized. If a plane wave component $\vec{F}(\vec{l},\omega)\, e^{i\vec{l} \cdot \vec{y}}$ of the transformed electric field $\vec{F}(\vec{y},\omega)$ is considered, then Eq. (5.3.16) may be rewritten with the help of Eqs. (5.3.17) and (5.3.21) as

$$\underline{\beta}(\vec{l},\omega)\cdot\vec{F}(\vec{l},\omega)\, e^{i\vec{l}\cdot\vec{y}} = \vec{P}^{eff}(\vec{y})\, e^{-i\omega t}/\epsilon_0 \quad , \qquad (5.3.22)$$

where

$$\underline{\beta}(\vec{l},\omega) \equiv (n^{eff})^2(\underline{1} - \hat{l}\hat{l}) - \underline{\kappa}^{eff} \quad , \qquad (5.3.23)$$

$$n^{eff} \equiv \frac{cl}{\omega\sqrt{\det\underline{\kappa}_m}} = \frac{cl}{\omega \det \underline{\kappa}_m^{1/2}} = \frac{cl}{\omega} \det \underline{\kappa}_m^{-1/2} \quad , \qquad (5.3.24)$$

$$\hat{l} \equiv \frac{\vec{l}}{|\vec{l}|} = \frac{\vec{l}}{\sqrt{\vec{l}\cdot\vec{l}}} = \frac{\underline{\kappa}_m^{1/2}\cdot\hat{s}}{\sqrt{\hat{s}\cdot\underline{\kappa}_m\cdot\hat{s}}} \quad . \qquad (5.3.25)$$

5.4 The Inside Green's Function

A. Formulation

When plane waves are not suitable or convenient for the electromagnetic field because of experimental geometry or otherwise, it is best to use a Green's function method to obtain the field created in response to the driving non-linear polarization. The reason the wave equation was transformed to the form given by Eq. (5.3.16), was so that the Green's function method of Lax and Nelson[104] for an anisotropic dielectric could be applied to the case of a magnetic dielectric after appropriate generalization. The solution to Eq. (5.3.16) is given by

$$\vec{F}(\vec{y},\omega) = \int \underline{G}(\vec{y},\vec{y}') \cdot \vec{P}^{eff}(\vec{y}')\, d\vec{y}'\, e^{-i\omega t}/\epsilon_0 \quad , \tag{5.4.1}$$

where the dyadic Green's function is defined by

$$\underline{G}(\vec{y},\vec{y}') = \int_{-\infty}^{+\infty} \frac{e^{i\vec{l}\cdot(\vec{y}-\vec{y}')}}{\underline{B}(\vec{l},\omega)} \frac{d\vec{l}}{(2\pi)^3} \quad . \tag{5.4.2}$$

Here $\underline{B}(\vec{l},\omega)$ is defined in Eq. (5.3.23).

The reciprocal of the matrix $\underline{B}$ has been previously shown by Lax and Nelson to have the eigenvector expansion

$$\underline{B}^{-1}(\vec{l},\omega) = \sum_{\varphi=1,2} \frac{\vec{F}^{\varphi}(\vec{l},\omega)\ \vec{F}^{\varphi}(\vec{l},\omega)}{\left[\left(\frac{n^{eff}}{n_{\varphi}^{eff}}\right)^2 - 1\right]} - \frac{\hat{l}\hat{l}}{\hat{l}\cdot\underline{K}_e^{eff}\cdot\hat{l}} \quad , \tag{5.4.3}$$

where $\vec{F}^{\varphi}$ is an eigenvector and $\left(\frac{1}{n_{\varphi}^{eff}}\right)^2$ the corresponding eigenvalue of the homogeneous wave equation for frequency ω and direction of propagation $\hat{l}$ [see Eqs. (5.3.22) and (5.3.23) with $\vec{P}^{eff} = 0$],

$$(\underline{1} - \hat{l}\hat{l}) \cdot \vec{F}^{\varphi} = \left(\frac{1}{n_{\varphi}^{eff}}\right)^2 \underline{K}_e^{eff} \cdot \vec{F}^{\varphi} \quad , \tag{5.4.4}$$

and the eigenvectors have been chosen to obey the weighted orthogonality condition

$$\vec{F}^{\varphi} \cdot \underline{K}_{e}^{eff} \cdot \vec{F}^{\theta} = \delta^{\varphi\theta} \quad ,(\varphi,\theta = 1,2,3) \quad . \tag{5.4.5}$$

In Eq. (5.4.3) the $\varphi = 3$ or longitudinal eigenvector, $\vec{F}^{3}$ parallel to $\hat{I}$, has been separated because $n_{3}^{eff} = \infty$, and the last term in Eq. (5.4.3) will make no contribution in the asymptotic limit $|\vec{R}_{y}| = |\vec{y} - \vec{y}'| \to \infty$ (although in certain nonlinear driven processes, the longitudinal contribution can be substantial[105]).

If an electric field eigenvector $\vec{E}^{\varphi}$ is defined through Eq. (5.3.13) by the relation

$$\vec{F}^{\varphi} = \underline{K}_{m}^{1/2} \cdot \vec{E}^{\varphi} \quad , \tag{5.4.6}$$

the orthogonality condition Eq. (5.4.5) in conjunction with Eq. (5.3.18) yields the more familiar biorthogonality condition

$$\vec{E}^{\varphi} \cdot \vec{D}^{\theta} = \delta^{\varphi\theta} \quad , \tag{5.4.7}$$

where

$$\vec{D}^{\theta} \equiv \underline{K}_{e} \cdot \vec{E}^{\theta} \tag{5.4.8}$$

is an electric displacement eigenvector. These conditions apply for $\varphi,\theta = 1,2,3$. Since $\vec{F}^{3}$ is parallel to $\hat{I}$, Eqs.

(5.3.25) and (5.4.6) imply that $\vec{E}^3$ is parallel to $\hat{s}$, the direction of propagation. Equation (5.4.7) tells us then that $\vec{D}^1$ and $\vec{D}^2$ are perpendicular to $\hat{s}$. Moreover, if we take the scalar product of the vector $\underline{K}_e^{eff} \cdot \vec{F}^{\theta}$ with the homogeneous wave equation (5.4.4), we obtain, after making use of Eqs. (5.3.18), (5.4.5) and (5.4.8), the relation

$$\vec{D}^{\theta} \cdot \underline{K}_m^{-1} \cdot \vec{D}^{\varphi} = 0 \ , \ \varphi \neq \theta = 1,2 \quad . \tag{5.4.9}$$

Thus, $\vec{D}^1$ is not perpendicular to $\vec{D}^2$. Since the magnetic field intensity $\vec{H}^1$ associated with eigenvector $\vec{E}^1$ is parallel to $\underline{K}_m^{-1} \cdot (\nabla \times \vec{E}^1)$ or $\underline{K}_m^{-1} \cdot (\hat{s} \times \vec{E}^1)$, and since $\hat{s}$ and $\vec{E}^1$ are both perpendicular to $\vec{D}^2$, it is necessary that $\vec{H}^1$ is parallel to $\underline{K}_m^{-1} \cdot \vec{D}^2$, which by means of Eq. (5.4.9), implies that

$$\vec{H}^1 \cdot \vec{D}^1 = 0 \quad . \tag{5.4.10}$$

Thus, $\vec{H}^1$ is perpendicular to $\vec{D}^1$. By similar arguments, it can easily be shown that $\vec{H}^2$ is perpendicular to $\vec{D}^2$.

Let us now transform our Green's function solution for the electric field back into terms of wave vector space $\vec{K}$ and real space $\vec{z}$. Using Eqs. (5.3.13), (5.3.14), (5.3.18), (5.3.19), (5.3.21), (5.3.24), (5.3.25), (5.4.1)-(5.4.3) and (5.4.6), we obtain

$$\vec{E}(\vec{z},\omega) = \int \underline{G}(\vec{z},\vec{z}') \cdot \vec{P}^{NL}(\vec{z}') \, d\vec{z}' \, e^{-i\omega t}/\epsilon_0 \quad , \tag{5.4.11}$$

where

$$\underline{G}(\vec{z},\vec{z}') \equiv \int_{-\infty}^{+\infty} \frac{e^{i\vec{\kappa}\cdot(\vec{z}-\vec{z}')}}{\underline{\alpha}(\vec{\kappa},\omega)} \frac{d\vec{\kappa}}{(2\pi)^3} , \tag{5.4.12}$$

and

$$\underline{\alpha}^{-1}(\vec{\kappa},\omega) \equiv \left(\frac{\omega}{c}\right)^2 \sum_{\varphi=1,2} \frac{\vec{E}^{\varphi}\,\vec{E}^{\varphi}}{\left(\frac{\kappa}{n^{\varphi}(\hat{s})}\right)^2 - \left(\frac{\omega}{c}\right)^2} - \frac{\hat{s}\hat{s}}{\hat{s}\cdot\underline{\kappa}_e\cdot\hat{s}} . \tag{5.4.13}$$

Here, the index of refraction is defined as usual by

$$n^{\varphi}(\hat{s}) \equiv \frac{c\,k^{\varphi}}{\omega} . \tag{5.4.14a}$$

These results appear to have exactly the same form as those of Lax and Nelson for the dielectric case. However, what must be remembered is that $\vec{E}^{\varphi}$ is really not an eigenvector but is related to the true eigenvector $\vec{F}^{\varphi}$ by Eq. (5.4.6); and $n^{\varphi}(\hat{s})$ is not a true eigenvalue here, but is related to the true eigenvalue n^{φ}_{eff} by

$$n^{\varphi}_{eff} = n^{\varphi}(\hat{s}) \sqrt{\hat{s}\cdot\underline{\kappa}_m\cdot\hat{s}} \Big/ \sqrt{\det \underline{\kappa}_m} . \tag{5.4.14b}$$

B. Stationary Phase Method

Since we are interested in radiation at a large distance from the source ($\kappa R \equiv \kappa|\vec{z} - \vec{z}'| \gg 1$), the asymptotic form of $\underline{G}$ given by the stationary phase method is appropriate. The presence of a pole requires the residue integration around the pole to be performed first, after which the stationary phase method can be consistently applied.

We are concerned with evaluating an integral of the form

$$g(\vec{R}) = \int \frac{N(\vec{K})}{D(\vec{K})} e^{i\vec{K} \cdot \vec{R}} d\vec{K} , \qquad (5.4.15)$$

in which $N(\vec{K})$ has no singularities for finite $\vec{K}$, and $D(\vec{K})$ possesses a finite number of zeroes.

A residue integration of Eq. (5.4.15) over a component of $\vec{K}$ parallel to $\vec{R}$ yields

$$g(\vec{R}) = 2\pi i \int_{D=0} \frac{N(\vec{K}(u,v))}{|\nabla_K D(\vec{K}(u,v))|} e^{i\vec{K}(u,v) \cdot \vec{R}} dS , \qquad (5.4.16)$$

where dS is the element of surface area of the surface $D(\vec{K},\omega) = 0$. This surface, which relates ω to $\vec{K}$, is parametrized by the surface parameters u and v; $\vec{K} = \vec{K}(u,v,\omega)$ is automatically on the surface. The stationary phase condition

$$\frac{\partial\vec{K}}{\partial u} \cdot \vec{R} = \frac{\partial\vec{K}}{\partial v} \cdot \vec{R} = 0 , \qquad (5.4.17)$$

selects a point $\vec{K}_0 = \vec{K}(u_0,v_0)$ on the surface whose tangent vectors are perpendicular to $\vec{R}$ - i.e., a point at which the

STATIONARY PHASE POINT k_0

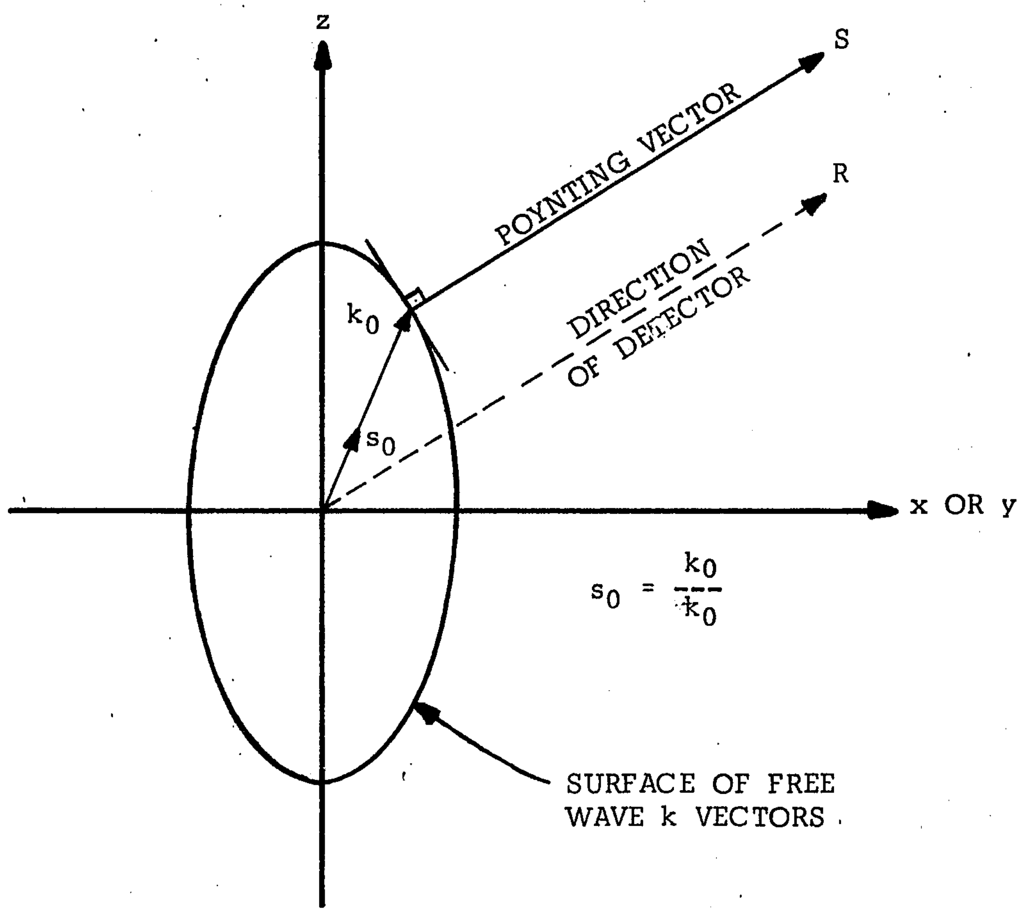


Fig. 1 The energy surface (k) = of a free wave is shown in k space. The stationary phase condition indicates that rariation in the directio R of the detector is produced primarily by a bundle of rays whose] vector, k_0 , is so chosen that the normal to the surface $_k$ (k) at k_0 is parallel to R, i.e., so that the Poynting vector S (which is necessarily parallel to the group velocity $_k$ (k)) is in the directio of observation.

surface normal is parallel to the direction of observation $\vec{R}$, as shown in Fig. 1. Since the energy surface is also the surface $\omega(\vec{\kappa})$ = constant, the unit surface normal $\vec{t}$ is necessarily in the direction of the group velocity

$$v_g \vec{t} = \nabla_\kappa \; \omega(\vec{\kappa}) \quad , \tag{5.4.18}$$

which can be shown to be in the direction of the Poynting vector.[106-108] Thus, the wave vectors that determine the intensity in the direction $\vec{R}$ come from the vicinity of that $\vec{\kappa}_0$ on the energy surface whose Poynting vector is parallel to $\vec{R}$.

The conventional stationary phase evaluation that expands the exponent to second order about $\vec{\kappa}_0$ reduces the integral Eq. (5.4.16) to

$$g(\vec{R}) = 4\pi^2 (i\vec{\kappa}_0 \cdot \vec{R}) \frac{c}{\sqrt{K}} \frac{N(\vec{\kappa}_0)}{| \; \nabla_\kappa D(\vec{\kappa}_0) \; |} \quad , \tag{5.4.19}$$

where K is the Gaussian curvature of the energy surface at $\vec{\kappa}_0$ defined by

$$K \equiv K_u \; k_v \quad , \tag{5.4.20}$$

when u and v are chosen in the directions of principal curvature and

$$K_u \equiv \frac{\partial^2 \vec{k}(u_0,v_0)}{\partial u^2} \cdot \vec{R}/R \quad , \tag{5.4.21}$$

$$K_v \equiv \frac{\partial^2 \vec{k}(u_0, v_0)}{\partial v^2} \cdot \vec{R}/R \quad , \tag{5.4.22}$$

and C = ± 1 or ± 1 according to the signs of the curvatures and the direction of the group velocity. Since only $|C|^2$ enters the power, we shall ignore C.

In view of Eqs. (5.4.12) and (5.4.13), we can rewrite the dyadic Green's function at large $\vec{R}$ in the form

$$\underline{G}(\vec{R}) = \left(\frac{\omega}{c}\right)^2 \sum_{\varphi=1,2} [n^\varphi(\hat{s}_0)]^2 \, \vec{E}^\varphi(\hat{s}_0)\vec{E}^\varphi(\hat{s}_0) g^\varphi(\vec{R}) \quad , \tag{5.4.23}$$

where the dependence of n^φ and $\vec{E}^\varphi$ on ω is not explicitly indicated, since ω remains constant in our discussion. What remains to be evaluated is the scalar Green's function

$$g^\varphi(\vec{R}) = \left(\frac{1}{n^\varphi(\hat{s}_0)}\right)^2 \int \frac{\exp(i\vec{k}\cdot\vec{R})d\vec{k}/(2\pi)^3}{[k/n^\varphi(\vec{s})]^2 - (\omega/c)^2 - i0} \quad . \tag{5.4.24}$$

The gradient of the denominator is given by

$$\nabla_k \left[\frac{k}{n(\hat{s})}\right]^2 = \frac{2k}{n(\hat{s})} \frac{1}{c} \nabla_k \omega \quad . \tag{5.4.25}$$

Using Eq. (5.4.18) and the relation

$$\frac{\partial s_i}{\partial \kappa_i} = \frac{\partial}{\partial \kappa_i} \left(\frac{\kappa_i}{\kappa} \right) = \frac{1}{\kappa} \left(\delta_{ij} - s_i \, s_j \right) \quad , \tag{5.4.26}$$

we obtain

$$v_g \, \vec{t} = \nabla_\kappa \frac{c\kappa}{n(\hat{s})} = \frac{c}{n} \, \hat{s} - \frac{c\kappa}{n^2} \left(\underline{1} - \hat{s}\hat{s} \right) \cdot \nabla_{\hat{s}} \, n(\hat{s}) \quad . \tag{5.4.27}$$

Thus,

$$v_g = \frac{c}{n} \frac{1}{\cos\delta} \quad , \tag{5.4.28}$$

where δ is the angle between $\vec{\kappa}$ and the Poynting vector. Equation (5.4.25) can now be rewritten as

$$\nabla_\kappa \left[\frac{\kappa}{n(\hat{s})} \right]^2 = \frac{2}{n^2} \frac{\kappa}{\cos\delta} \, \vec{t} \quad . \tag{5.4.29}$$

Equation (5.4.19) applied to Eq. (5.4.24) then yields

$$g^\varphi(\vec{R}) = f^\varphi \, \frac{\exp(i \; k_0^\varphi \cdot R)}{4\pi R} \quad , \tag{5.4.30}$$

where

$$f^\varphi \equiv \frac{\cos\delta^\varphi}{\kappa_0^\varphi \, \sqrt{K}} \quad . \tag{5.4.31}$$

The calculations of the electric field and the scattered power will follow after a brief digression on solid angle expansion and source volume demagnification corrections.

5.5 Solid Angle Expansion and Source Volume Demagnification

The ratio of the solid angle of ray (Poynting vector) directions inside an anisotropic medium to that outside the crystal can be simplified by writing it as a product of two factors:

$$\frac{d\Omega_{in}^{r}}{d\Omega_{out}} = \frac{d\Omega_{in}^{r}}{d\Omega_{in}^{k}} \frac{d\Omega_{in}^{k}}{d\Omega_{out}} \quad . \tag{5.5.1}$$

The first factor describes the ratio of solid angle in ray space to that in $\vec{k}$ vector space and is independent of the existence of a boundary. The second factor is a pure boundary effect on $\vec{k}$ vector solid angles. Lax and Nelson[109] have shown that

$$\frac{d\Omega_{in}^{r}}{d\Omega_{in}^{k}} = K\kappa^2/\cos\delta \quad , \tag{5.5.2}$$

$$\frac{d\Omega_{in}^{k}}{d\Omega_{out}} = (\cos\delta)(\cos\alpha)/(n^2\cos\beta) \quad , \tag{5.5.3}$$

where β is the inside angle of incidence by the ray and α is the emergence angle by the ray into free space. Combining Eqs. (5.5.1)-(5.5.3) yields

$$\frac{d\Omega_{in}^{r}}{d\Omega_{out}} = \left(\frac{\omega}{c} \right)^2 K \cos\alpha/\cos\beta \quad . \tag{5.5.4}$$

The above authors have also derived an expression for the linear dimension l_S along the laser beam within the crystal from which radiation will be admitted by a detector with field stop of length l_D:

$$l_S = \frac{l_D}{\sin\theta_S} \frac{\cos\beta}{\cos\alpha} \quad , \tag{5.5.5}$$

where θ_S is the scattering angle. A representation of the geometrical quantities in this section are shown in Fig. 2.

5.6 The Scattered Power

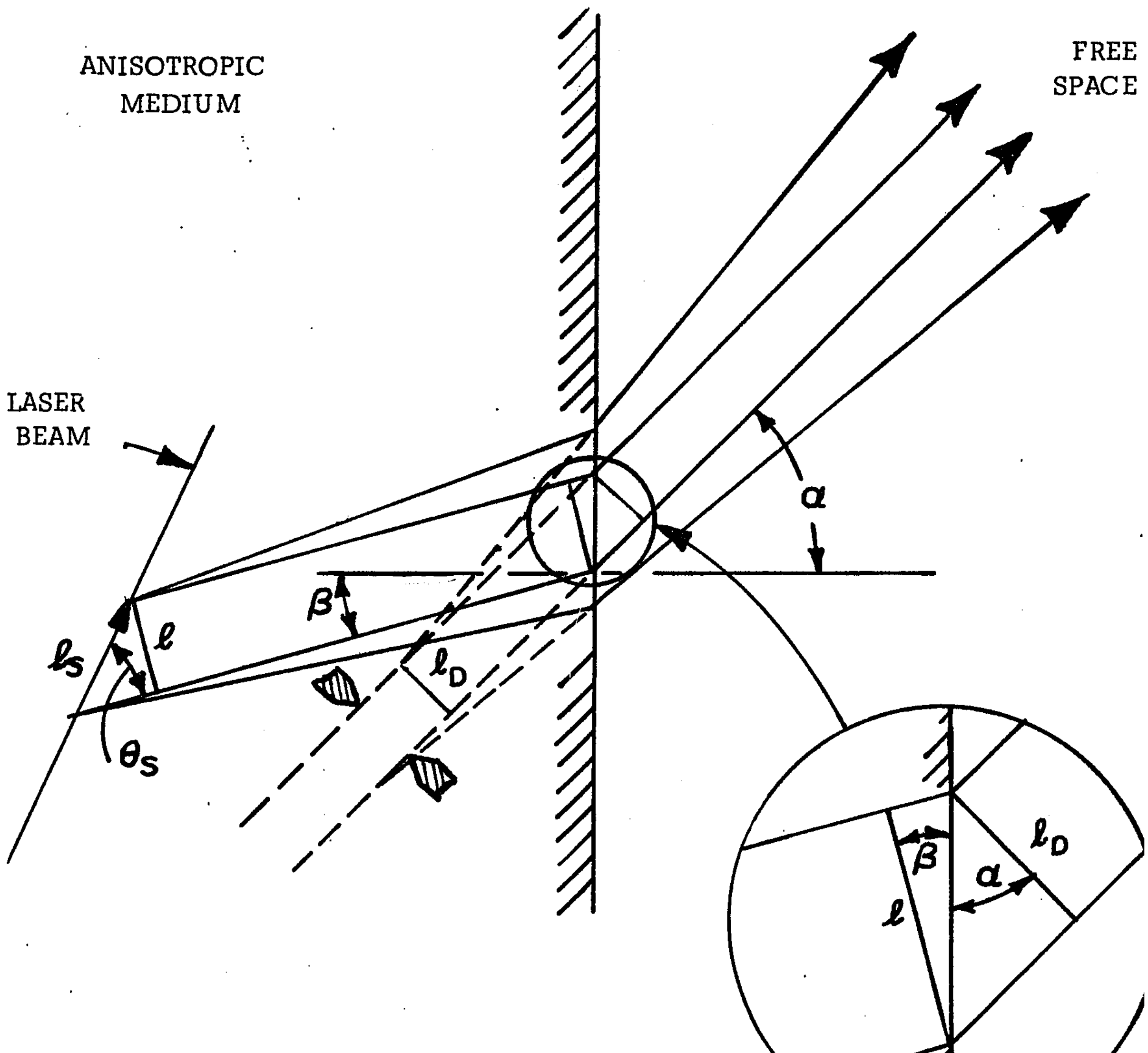
The electric field can be computed using the Green's function equation (5.4.11) with the help of Eqs. (5.4.23) and (5.4.30):

$$\vec{E}(\vec{z},\omega) = \sum_{\varphi=1,2} \frac{(k^{\varphi})^2 \, f^{\varphi} \, e^{i(\vec{k}^{\varphi} \cdot \vec{z} - \omega t)}}{4\pi \, \epsilon_0 \, | \vec{z} |} \vec{\mathcal{E}}^{\varphi} \, C^{\varphi} \quad , \tag{5.6.1}$$

where

$$C^{\varphi} \equiv \vec{\mathcal{E}}^{\varphi} \cdot \int e^{-i\vec{k}^{\varphi} \cdot \vec{z}'} \, \vec{P}^{NL} (\vec{z}') \, d\vec{z}' \quad . \tag{5.6.2}$$

Here we have used the far field approximation to replace R in the denominator of Eq. (5.4.30) with $| \vec{z} |$; i.e., the



$$\therefore\ \ell_S \sin\theta_S/\cos\beta = \ell/\cos\beta = \ell_D/\cos\alpha$$

Fig. 2 Demagnification corrections when the incident ray, refracted ray and surface normal are all in one plane. The detector field stop is represented in image space by knife edges which accept a dimension perpendicular to the beam. The portion of the laser beam accepted is determined by the geometrical conditions shown.

source dimension z' is much less than the distance to the point of observation.

The magnetic field intensity inside the crystal but in the far field region is known from Maxwell's equations to be

$$\vec{H}(\vec{z},\omega) = \frac{1}{i\omega\mu_0} \underline{k}_m^{-1} \cdot [\nabla \times \vec{E}(\vec{z},\omega)] \quad . \tag{5.6.3}$$

In evaluating $\nabla \times \vec{E}$ from Eq. (5.6.1), two terms arise, but one is smaller than the other by a factor $\simeq (\kappa z)^{-1}$. The only non-negligible contribution is that due to the rapidly fluctuating phase. Thus,

$$\vec{H}(\vec{z},\omega) = \sum_{\theta} \frac{e^{i(\vec{\kappa}^{\theta} \cdot \vec{z}-\omega t)}}{4\pi \epsilon_0 |\vec{z}|} \frac{f^{\theta}(\kappa^{\theta})^3}{\omega \mu_0} \underline{k}_m^{-1} \cdot (\hat{s}^{\theta} \times \vec{\mathcal{E}}^{\theta}) C^{\theta} \quad . \tag{5.6.4}$$

The time averaged Poynting vector $\frac{1}{2}$ Re $[\vec{E}^* \times \vec{H}]$ takes the form

$$\vec{S} = \frac{1}{2} \sum_{\varphi,\theta=1,2} \frac{(\kappa^{\varphi})^2(\kappa^{\theta})^3 f^{\varphi} f^{\theta}}{(4\pi|\vec{z}|)^2 \omega \epsilon_0^2 \mu_0} \vec{\mathcal{E}}^{\varphi} \times [\underline{k}_m^{-1} \cdot (\hat{s}^{\theta} \times \vec{\mathcal{E}}^{\theta})]$$

$$\text{Re}\{(C^{\varphi})^* C^{\theta} e^{i(\vec{\kappa}^{\theta}-\vec{\kappa}^{\varphi})\cdot\vec{z}}\} . \tag{5.6.5}$$

Ignoring the rapidly spatially varying cross terms and using Eq. (5.3.15), Eq. (5.6.5) may be rewritten as

$$\vec{S} = \sum_{\varphi=1,2} S^{\varphi}$$

$$= \sum_{\varphi=1,2} \frac{\frac{1}{2}\, c\left(\frac{\omega}{c}\right)^4 (n^{\varphi})^5 (f^{\varphi})^2}{(4\pi|\vec{z}|^2)\,\epsilon_0 \det \underline{\underline{K}}_m}\, \underline{\underline{K}}_m^2$$

$$\cdot\ [|\mathfrak{E}^{\varphi}|^2 \hat{s}^{\varphi} - (\mathfrak{E}^{\varphi}\cdot\hat{s}^{\varphi})\mathfrak{E}^{\varphi}]\,|C^{\varphi}|^2 \ . \qquad (5.6.6)$$

The scattered power from mode φ outside the crystal is given by the relation

$$P_{out}^{\varphi} = |\ \vec{S}^{\varphi}\ |\ |\ \vec{z}\ |^2\, d\Omega^r\ T^{\varphi} \ , \qquad (5.6.7)$$

where T^{φ} is the transmission coefficient.

We will now derive an expression for $|\mathfrak{E}^{\varphi}|$ so as to eliminate it from our results. Consideration of Eqs. (5.4.4) and (5.4.5) yield

$$(n^{\varphi})^2\, \frac{\hat{s}^{\varphi}\cdot\underline{\underline{K}}_m\cdot\hat{s}^{\varphi}}{\det \underline{\underline{K}}_m}\, \{\ |\ \vec{\mathfrak{F}}^{\varphi}\ |^2 - (\vec{\mathfrak{F}}^{\varphi}\cdot\hat{1})^2\ \} = 1 \ . \qquad (5.6.8)$$

Using Eqs. (5.3.25) and (5.4.6), it is easy to show that

$$|\ \vec{\mathfrak{F}}^{\varphi}\ |^2 = |\ \mathfrak{E}^{\varphi}\ |^2\ (\hat{e}^{\varphi}\cdot\underline{\underline{K}}_m\cdot\hat{e}^{\varphi}) \ , \qquad (5.6.9)$$

$$(\vec{\mathfrak{F}}^{\varphi}\cdot\hat{1})^2 = |\ \mathfrak{E}^{\varphi}\ |^2\ \frac{(\hat{e}^{\varphi}\cdot\underline{\underline{K}}_m\cdot\hat{s}^{\varphi})^2}{\hat{s}^{\varphi}\cdot\underline{\underline{K}}_m\cdot\hat{s}^{\varphi}} \ , \qquad (5.6.10)$$

where $\hat{e}^{\varphi}$ is a unit vector in the direction of $\vec{E}^{\varphi}$. Combining Eqs. (5.6.8)-(5.6.10), we obtain

$$|\vec{E}^{\varphi}|^2 = \frac{\det \underline{\kappa}_m}{(n^{\varphi})^2} [(\hat{e}^{\varphi} \cdot \underline{\kappa}_m \cdot \hat{e}^{\varphi})(\hat{s}^{\varphi} \cdot \underline{\kappa}_m \cdot \hat{s}^{\varphi}) - (\hat{e}^{\varphi} \cdot \underline{\kappa}_m \cdot \hat{s}^{\varphi})^2]^{-1} \quad . \tag{5.6.11}$$

The time averaged Poynting vector of an incident mode $\frac{1}{2}$ Re $[(\vec{E}^{\theta})^* \times \vec{H}^{\theta}]$ takes the form

$$\vec{S}^{\theta} = \frac{1}{2} \frac{c \ \epsilon_0 \ n^{\theta}}{\det \underline{\kappa}_m} |\vec{E}^{\theta}|^2 \ \underline{\kappa}_m^2 \cdot [\hat{s}^{\theta} - \hat{e}^{\theta}(\hat{s}^{\theta} \cdot \hat{e}^{\theta})] \quad . \tag{5.6.12}$$

Thus,

$$|\vec{S}^{\theta}| = \frac{1}{2} \frac{c \ n^{\theta} \ \epsilon_0}{\det \underline{\kappa}_m} |\vec{E}^{\theta}|^2 \{ [\hat{s}^{\theta} - \hat{e}^{\theta}(\hat{s}^{\theta} \cdot \hat{e}^{\theta})] \cdot \underline{\kappa}_m^4 \cdot [\hat{s}^{\theta} - \hat{e}^{\theta}(\hat{s}^{\theta} \cdot \hat{e}^{\theta})] \}^{\frac{1}{2}} \quad . \tag{5.6.13}$$

If the Poynting vector for mode θ is in the direction $\vec{t}^{\theta}$, then from Eq. (5.6.12)

$$\vec{t}^{\theta} = \frac{\underline{\kappa}_m^2 \cdot [\hat{s}^{\theta} - \hat{e}^{\theta}(\hat{s}^{\theta} \cdot \hat{e}^{\theta})]}{\{ [\hat{s}^{\theta} - \hat{e}^{\theta}(\hat{s}^{\theta} \cdot \hat{e}^{\theta})] \cdot \underline{\kappa}_m^4 \cdot [\hat{s}^{\theta} - \hat{e}^{\theta}(\hat{s}^{\theta} \cdot \hat{e}^{\theta})] \}^{\frac{1}{2}}} \quad . \tag{5.6.14}$$

The scalar product between the direction of propagation and the ray vector is therefore

$$\cos\delta^{\theta} = \hat{s}^{\theta} \cdot \vec{t}^{\theta}$$

$$= \frac{\hat{s}^{\theta}\cdot\underline{K}_m^2\cdot\hat{s}^{\theta}-(\hat{s}^{\theta}\cdot\underline{K}_m^2\cdot\hat{e}^{\theta})(\hat{s}^{\theta}\cdot\hat{e}^{\theta})}{\{[\hat{s}^{\theta}-\hat{e}^{\theta}(\hat{s}^{\theta}\cdot\hat{e}^{\theta})]\cdot\underline{K}_m^4\cdot[\hat{s}^{\theta}-\hat{e}^{\theta}(\hat{s}^{\theta}\cdot\hat{e}^{\theta})]\}^{\frac{1}{2}}} \quad . \tag{5.6.15}$$

Equation (5.6.13) can then be rewritten

$$|\vec{S}^{\theta}| = \frac{1}{2}\,\frac{c\, n^{\theta}\, \epsilon_0 |\vec{E}^{\theta}|^2}{(\det \underline{K}_m)\cos\delta^{\theta}}\,[\hat{s}^{\theta}\cdot\underline{K}_m^2\cdot\hat{s}^{\theta}-(\hat{s}^{\theta}\cdot\underline{K}_m^2\cdot\hat{e}^{\theta})(\hat{s}^{\theta}\cdot\hat{e}^{\theta})] \quad . \tag{5.6.16}$$

For beam area A, the incident power in mode θ is

$$P^{\theta} = A|\vec{S}^{\theta}| = P_{out}^{\theta}\, T^{\theta} \quad . \tag{5.6.17}$$

The second form is valid since we have made the implicit assumption that the laser beam is not appreciably depleted in travelling through the crystal; i.e., we are considering non-stimulated light scattering.

The ratio of scattered to incident powers outside the crystal is obtained by combining Eqs. (5.6.6), (5.6.7), (5.6.11), (5.6.15)-(5.6.17):

$$\frac{P_{out}^{\varphi}}{P_{out}^{\theta}} = \left(\frac{\omega}{c}\right)^4 BJ\, \frac{\cos\delta^{\varphi}\cos\delta^{\theta}(\det \underline{K}_m)^2}{8\pi^2 n^{\varphi} n^{\theta}}$$

$$\frac{[\hat{s}^{\varphi}\cdot \underline{\kappa}_m^2\cdot\hat{s}^{\varphi}-(\hat{s}^{\varphi}\cdot\underline{\kappa}_m^2\cdot\hat{e}^{\varphi})(\hat{s}^{\varphi}\cdot\hat{e}^{\varphi})]}{[\hat{s}^{\theta}\cdot\underline{\kappa}_m^2\cdot\hat{s}^{\theta}-(\hat{s}^{\theta}\cdot\underline{\kappa}_m^2\cdot\hat{e}^{\theta})(\hat{s}^{\theta}\cdot\hat{e}^{\theta})]\,[\hat{e}^{\varphi}\cdot\underline{\kappa}_m\cdot\hat{e}^{\varphi}(\hat{s}^{\varphi}\cdot\underline{\kappa}_m\cdot\hat{s}^{\varphi})-(\hat{e}^{\varphi}\cdot\underline{\kappa}_m\cdot\hat{s}^{\varphi})^2]^2}. \tag{5.6.18}$$

The geometrical factors (including the source length $l_S = V_S/A$ where V_S is the source volume) are all contained in

$$B = \frac{T^{\varphi}\ T^{\theta}(n^{\varphi}\ f^{\varphi})^2\ d\Omega^r\ l_S}{\cos^2\ \delta^{\varphi}} = \frac{T^{\varphi}\ T^{\theta}\ d\Omega_{out}\ l_D}{\sin\theta_S}\ , \tag{5.6.19}$$

which has been simplified with the help of Eqs. (5.4.14), (5.4.31), (5.5.4) and (5.5.5). The nonlinear optics are all contained in

$$J = \frac{1}{2\epsilon_0^2\ |\vec{E}^{\theta}|^2}\frac{1}{V_S}\ |\ \hat{e}^{\varphi}\cdot\int_{V_S} e^{-i\vec{K}^{\varphi}\cdot\vec{z}}\ \vec{P}^{NL}(\vec{z},\omega)\ d\vec{z}\ |^2\ , \tag{5.6.20}$$

where $\vec{K}^{\varphi}$ is the propagation vector of the scattered light inside the crystal. A power ratio is more appropriate in our case than a cross section since we are assuming a beam area smaller than the crystal. These results reduce to those of Lax and Nelson[109] when we set $(\underline{\kappa}_m)_{ij} = \delta_{ij}$ and $\hat{s}^{\varphi}\cdot\hat{e}^{\varphi} = \sin\delta^{\varphi}$.

The finite source region V_S gives rise to a phase matching factor whose width $\Delta\kappa$ is of order $(V_S)^{-1/3}$, and a line breadth $\Delta\omega \simeq v_g\ \Delta\kappa$, where v_g is the magneto-polariton group velocity. Since this is negligible compared to broadening by damping, we can let $V_s \to \infty$ and apply the Weiner-Khinchin theorem to rewrite Eq. (5.6.20) as

$$J = \frac{e_i^{\varphi} e_a^{\varphi} \int_{-\infty}^{\infty} e^{-i\vec{K}^{\varphi} \cdot \vec{z}} \langle \vec{P}_i^{NL}(0,\omega)^* \, \vec{P}_a^{NL}(\vec{z},\omega) \rangle \, d\vec{z}}{2\epsilon_0^2 |\vec{E}^{\theta}|^2} \quad . \qquad (5.6.21)$$

We define the spatial Fourier components of the polarization by

$$\vec{P}_i^{NL}(\vec{z},\omega) = \Omega^{-\frac{1}{2}} \sum_{\vec{K}} \vec{P}_i(\vec{K},\omega) \, e^{i\vec{K} \cdot \vec{z}} \quad , \qquad (5.6.22)$$

where the sum is over the discrete set of $\vec{K}$ allowed when periodic boundary conditions are introduced over a normalization volume Ω. Therefore, Eq. (5.6.21) can be written as

$$J = \frac{e_i^{\varphi} \, e_a^{\varphi} \, \langle P_i(\vec{K}^{\varphi},\omega)^* \; P_a(\vec{K}^{\varphi},\omega) \rangle}{2\epsilon_0^2 \, |\vec{E}^{\theta}|^2} \quad . \qquad (5.6.23)$$

Equations (5.6.18)-(5.6.23) were derived assuming that nonlinear polarization possessed a single frequency $\omega = \omega^{\varphi}$ (the scattered frequency)

$$\vec{P}^{NL}(\vec{z},t) = \frac{1}{2} \left[\vec{P}^{NL}(\vec{z},\omega) e^{-i\omega t} + \vec{P}^{NL}(\vec{z},\omega)^* \, e^{i\omega t} \right]_{\omega=\omega^{\varphi}} \quad . \qquad (5.6.24)$$

When, because of thermal motion, the nonlinear polarization contains a distribution of Fourier components, it is appropriate to decompose J according to

$$J = \int_0^\infty J(\omega)\ d\omega/2\pi \quad . \qquad (5.6.25)$$

Only positive frequencies appear in Eq. (5.6.25), since our evaluation of the Poynting vector $\vec{S}^\varphi$, Eq. (5.6.6), made use of both positive and negative frequency components of $\vec{E}$ and $\vec{H}$.

If we write

$$P_i(\vec{K},t) = \frac{1}{2}\ [P_i(\vec{K},\omega^\varphi)\ e^{-i\omega^\varphi t} + P_i(\vec{K},\omega^\varphi)^*\ e^{i\omega^\varphi t}] \quad , \qquad (5.6.26)$$

or

$$P_i(\vec{K},t) = P_i^{(+)}(\vec{K},t) + P_i^{(-)}(\vec{K},t) \quad , \qquad (5.6.27)$$

it is appropriate to make the replacement

$$\langle P_i(\vec{K},\omega^\varphi)\ P_a(\vec{K},\omega^\varphi)\rangle$$

$$\rightarrow \int_0^\infty \frac{d\omega}{2\pi}\ 4 \int_{-\infty}^\infty e^{i\omega t}$$

$$\times\ \langle P_i^{(-)}(\vec{K},0)\ P_a^{(+)}(\vec{K},t)\rangle\ dt \quad , \qquad (5.6.28)$$

which reduces to the original value of the lefthand side when the form (5.6.26) appropriate to a single frequency is used. More generally, Eq. (5.6.28) describes the frequency spectrum of $P_i(\vec{K},t)$ for $\omega > 0$, and vanishes when $\omega < 0$. Indeed, we have used the positive frequency component $P_a^{(+)}(\vec{K},t)$ precisely to insure that the negative frequency component of the noise spectrum

$$J(\omega) = \frac{2e_i^{\varphi}\, e_a^{\varphi}}{\epsilon_0^2\, |E^{\theta}|^2} \int_{-\infty}^{\infty} e^{i\omega t} \langle P_i^{(-)}(\vec{K},0)\; P_a^{(+)}(\vec{K},t)\rangle \, dt \qquad (5.6.29)$$

vanishes.

The nonlinear polarization induced by the laser electric field peak amplitude $\vec{E}^{\theta}$ (for mode θ inside the crystal) can be written

$$P_a^{(+)}(\vec{K}^{\varphi},t) = \epsilon_0\, \chi_{ab}(\vec{K}^{\varphi} - \vec{K}^{\theta},t)\; E_b^{\theta}(\omega^{\theta})$$

$$\times\; e^{i\vec{K}^{\theta}\cdot\vec{z} - i\omega^{\theta}t} \quad . \qquad (5.6.30)$$

In Eq. (5.6.30) the matter via χ_{ab} supplies the wave vector $\vec{K}^{\varphi} - \vec{K}^{\theta}$ which, when added to the incident wave vector $\vec{K}^{\theta}$, yields the scattered wave vector $\vec{K}^{\varphi}$. Both signs of the frequency are permitted in χ_{ab} , since we assume that the magneto-polariton frequencies are less than the laser frequency ω^{θ} so that the product still retains only positive frequencies. The resulting expression for $J(\omega)$ is given by

$$J(\omega + \omega^{\theta}) = 2 \int_{-\infty}^{\infty} e^{i\omega t} \langle N^{\varphi\theta}(\vec{K},0)^{\dagger} N^{\varphi\theta}(\vec{K},t)\rangle \, dt \quad , \qquad (5.6.31)$$

where $\vec{K}$ is evaluated at $\vec{K}^{\varphi} - \vec{K}^{\theta}$ and

$$N^{\varphi\theta}(\vec{K},t) = e_i^{\varphi} x_{ij}(\vec{K},t) e_j^{\theta} \quad . \qquad (5.6.32)$$

The calculations have been entirely classical so far. The advantage of this procedure is that we have been able to deal with all the complicated geometrical optics and boundary condition aspects of the problem. The final result, given by Eq. (5.6.18) evaluated at $\omega = \omega + \omega^{\theta}$ for $P_{out}^{\varphi}(\omega + \omega^{\theta})/P_{out}^{\theta}$, is valid quantum mechanically, provided the operators in the correlation function for J are properly ordered and evaluated quantum mechanically. The ordering has already been chosen with appropriate malice aforethought. Also, Hermitian conjugation ($\dagger$) has been used in place of complex conjugation. The time correlation in Eq. (5.6.31) has not been symmetrized to make it Hermitian. However, the final result, $J(\omega + \omega^{\theta})$, can readily be shown to be real. The quantum mechanical evaluation of $J(\omega + \omega^{\theta})$ will be carried out with the help of the fluctuation-dissipation theorem summarized in the next section.

5.7 The Fluctuation-Dissipation Theorem[110]

If K is the hamiltonian of the universe, half of the fluctuation-dissipation theorem can be written in the form

$$\langle B(t)\, A(0)\rangle = \langle A(0)\, B(t + i\hbar\beta)\rangle \quad , \qquad (5.7.1)$$

where $B(t) = \exp(iKt/\hbar)$ is the time-dependent, Heisenberg operator for B and $\langle M\rangle$ means trace $(M\rho_e)$, where $\rho_e = \exp(-\beta K)/\text{trace}\,\exp(-\beta K)$ is the equilibrium density operator at a temperature determined by $\beta = 1/(kT)$. Equation (5.7.1) can be readily established by permuting operators inside the trace.

Equation (5.7.1) gives us the useful relation

$$\int_{-\infty}^{\infty} e^{i\omega t} \langle B(t)\, A(0)\rangle\, dt$$

$$= e^{\beta\hbar\omega} \int_{-\infty}^{\infty} e^{i\omega t} \langle A(0)\, B(t)\rangle\, dt \qquad (5.7.2)$$

between spectra calculated with different orders of the operators.

If one replaces K by $K - \lambda A e^{-i\omega t}$ (which adds an infinitesimal force of strength λ to the equation for the momentum p_A conjugate to A), the linear response of B per unit force applied to A is given by

$$T_{BA}(\omega) \equiv [\, \langle B(t)\rangle_{\lambda A} - \langle B\rangle\,]\, e^{i\omega t}/\lambda$$

$$= - \int_{0}^{\infty} e^{i\omega t}\, dt\, \langle (B(t), A)\rangle \quad , \qquad (5.7.3)$$

where $\langle B(t)\rangle_{\lambda A}$ is the trace of B against the density matrix ρ computed by first-order perturbation theory from ρ_e. The notation $(B,A) \equiv [B,A]/i\hbar$ is used.

A comparison of Eqs. (5.7.2) and (5.7.3) yields

$$\int_{-\infty}^{\infty} e^{i\omega t} \langle B(t)A(0)\rangle = -i\hbar\ [T_{BA}(\omega) - T_{AB}(-\omega)]\ [n(\omega) + 1] \quad , \tag{5.7.4}$$

and the use of Eq. (5.7.2) gives

$$\int_{-\infty}^{\infty} e^{i\omega t} \langle A(0)B(t)\rangle dt = -i\hbar[T_{BA}(\omega)-T_{AB}(-\omega)]n(\omega) \quad , \tag{5.7.5}$$

where $n(\omega) = [\exp(\beta\hbar\omega) - 1]^{-1}$.

One may verify that if

$$A^{\dagger} = B, \qquad B^{\dagger} = A \quad , \tag{5.7.6}$$

then

$$T_{AB}(\omega) = T_{BA}(\omega)^{*} \quad . \tag{5.7.7}$$

Alternatively, if the time reversed operators obey

$$A^{T} = \epsilon_A A, \quad B^{T} = \epsilon_B B, \quad \epsilon_A \epsilon_B = 1 \quad , \tag{5.7.8}$$

then Eq. (5.7.7) is obeyed even though neither A nor B are

Hermitian.

In view of Eq. (5.6.31), our operators will obey condition Eq. (5.7.6). Equation (5.7.5) can thus be rewritten in the simpler form

$$\int_{-\infty}^{\infty} e^{i\omega t} \langle A(0)\ B(t)\rangle\, dt = 2\hbar \,\mathrm{Im}\ T_{BA}(\omega) n(\omega) \quad . \qquad (5.7.9)$$

Note that it is the response of the time dependent variable $B(t)$ to a force on the time independent variable A that is measured by T_{BA}.

Since

$$n(-\omega) = -[n(\omega) + 1] \quad , \qquad (5.7.10)$$

the intensities of positive and negative frequency components in a correlation such as Eq. (5.7.9) will have the typical anti-Stokes/Stokes ratio. The spectrum Eq. (5.6.31) can now be evaluated in the form

$$J(\omega + \omega^{\theta}) = 4\hbar n(\omega) \mathrm{Im}\ T_{N^{+}N}(\omega) \quad , \qquad (5.7.11)$$

where N is an abbreviation for $N^{\varphi\theta}$, and J is understood to vanish for $\omega + \omega^{\theta} < 0$.

5.8 Raman Scattering by Magneto-Polaritons

A. The Nonlinear Polarization

The nonlinear polarization under the simplifications of Section 5.1 is obtainable from Eqs. (3.4.2) and (5.2.5):

$$P_i^{NL}(\vec{z},t) = \epsilon_0 \sum_{\mu,\nu} q^{\mu} \Upsilon_{ij}^{\mu\nu} G_j^{\nu} \quad . \tag{5.8.1}$$

If no simplifications were made, the nonlinear force exerted on the internal coordinates would have the form

$$G_i^{\mu} = -(R_{iA}-\delta_{iA})\rho^0 \frac{\partial\Sigma}{\partial\Lambda_A^{\mu}} - \rho^0 \delta_{iA} \frac{\partial\Sigma'}{\partial\Lambda_A^{\mu}}$$

$$- 2^{(2000)}K_{iB}^{\mu\nu} \hat{y}_j^{\nu} (R_{jB}-\delta_{jB})$$

$$- ^{(1100)}K_{iBC}^{\mu}[E_{BC} - \frac{1}{2} (\hat{u}_{b,c}+\hat{u}_{c,b})]$$

$$- ^{(1010)}K_{iB}^{\mu\alpha}[\hat{m}_j^{\alpha}(R_{jB}-\delta_{jB})+m_j^{S\alpha}(R_{jB}-\delta_{jB}-\frac{1}{2} (\hat{u}_{j,b}-\hat{u}_{b,j}))]$$

$$- ^{(1001)}K_{iBC}^{\mu\alpha}(\Gamma_{B;C}^{\alpha} - \hat{m}_{j,c}^{\alpha}\delta_{jB})$$

$$+ f_i^{\mu e} - q^{\mu}E_i - q^{\mu}(\frac{\partial\hat{\vec{u}}}{\partial t} \times \vec{B}^S)_i - m_K^{\mu S} B_{K,j}$$

$$- (\frac{\partial\hat{\vec{p}}^{\mu}}{\partial t} \times \vec{B}^S)_i - m^{\mu}(\ddot{y}_i^{\mu} - \frac{\partial^2 \hat{y}_i^{\mu}}{\partial t^2}) \quad , \tag{5.8.2}$$

where $\rho^0 \Sigma'$ represents that portion of the stored energy $\rho^0\Sigma$ above and beyond the quadratic terms. If the elastic degree of freedom is ignored and all multipole terms above the electric dipole term are discarded, the nonlinear force would have the form

$$G_i^\mu = -[\ 3\,^{(3000)}K_{ijk}^{\mu\nu\gamma}\,\tilde{y}_j^\nu\,\tilde{y}_k^\gamma + 2\,^{(2010)}K_{ijk}^{\mu\nu\alpha}\,\tilde{y}_j^\nu\,\hat{m}_k^\alpha$$

$$+ 2\,^{(2001)}K_{ijkl}^{\mu\nu\alpha}\,\tilde{y}_j^\nu\,\hat{m}_{k,l}^\alpha + {}^{(1020)}K_{ijk}^{\mu\alpha\beta}\,\hat{m}_j^\alpha\,\hat{m}_k^\beta$$

$$+ {}^{(1002)}K_{ijkla}^{\mu\alpha\beta}\,\hat{m}_{j,k}^\alpha\,\hat{m}_{l,a}^\beta + {}^{(1011)}K_{ijkl}^{\mu\alpha\beta}\,\hat{m}_j^\alpha\,\hat{m}_{k,l}^\beta\,] \;. \tag{5.8.3}$$

Since we will be dealing with the scattering of an input electromagnetic field (the laser) accompanied by the creation or destruction of a single magneto-polariton, all terms involving a product of three or more fields must be discarded as is reflected in Eq. (5.8.3). Moreover, two field terms involving a product of two magnetizations can be immediately discarded since one of these fields must be at an optical frequency and hence minute because of its inability to respond to such a high frequency. With this in mind, the last three terms of Eq. (5.8.3) may be discarded. Finally, we discard the third term on the righthand side of Eq. (5.8.3) in keeping with the simplification of ignoring gradient terms. The result for the nonlinear force is then

$$G_i^{\mu} = -[\ 3^{(3000)}K_{ijk}^{\mu\nu\lambda}\ \hat{y}_j^{\nu}\ \hat{y}_k^{\lambda} + 2^{(2010)}K_{ijk}^{\mu\nu\alpha}\ \hat{y}_j^{\nu}\ \hat{m}_k^{\alpha}\ ] \quad . \qquad (5.8.4)$$

We are considering a problem in which we have two input fields: the laser field at frequency ω^{Θ} which is an optical frequency, and the magneto-polariton field, thermally driven, and at frequency ω which is much lower than the laser frequency. We expand any field variable $\vec{Z}(\vec{y}^{\mu},\vec{m}^{\alpha},\vec{E}$, or $\vec{B})$ in a Fourier expansion

$$\vec{Z}(\vec{z},t) = \frac{1}{2} \sum_{m,n=-\infty}^{+\infty} \vec{Z}(\vec{z},t;m,n) \quad , \qquad (5.8.5)$$

where the individual terms have the form

$$\vec{Z}(\vec{z},t;m,n) = \vec{Z}(\vec{z};m,n)\ e^{-i(m\omega^{\Theta} + n\omega)t} \quad . \qquad (5.8.6)$$

Reality of the original variable $\vec{Z}$ is guaranteed by the requirement

$$\vec{Z}(\vec{z};-m,-n) = \vec{Z}^{*}(\vec{z};m,n) \quad . \qquad (5.8.7)$$

We have included the factor $\frac{1}{2}$ in Eq. (5.8.5) so that the magnitude of $\vec{Z}(\vec{z};m,n)$ represents the amplitude at its associated frequency.

If the expansion Eq. (5.8.5) is applied to Eq. (5.8.1) and the corresponding coefficients with the frequency $\omega^{\Theta} \pm \omega$, i.e., the $(m,n) = (1, \pm 1)$ coefficients are compared, we obtain the result

$$P_i^{NL}(1, \pm 1) = \epsilon_0 \sum_{\mu,\nu} q^{\mu} \Upsilon_{ia}^{\mu\nu}(\omega^{\varphi}) \, G_a^{\nu}(1, \pm 1) \quad , \qquad (5.8.8)$$

where all field variables are understood to be functions of $\vec{z}$ and

$$\omega^{\varphi} = \omega^{\Theta} \pm \omega \qquad (5.8.9)$$

is the anti-Stokes or Stokes scattered frequency. Similarly, the nonlinear driving force can be written in the form

$$G_a^{\nu}(1, \pm 1) = - 3^{(3000)}K_{ajK}^{\nu\lambda\xi} \, y_j^{\lambda}(1,0) \, y_K^{\xi}(0, \pm 1)$$
$$- {}^{(2010)}K_{ajK}^{\nu\lambda\alpha} \, y_j^{\lambda}(1,0) \, m_K^{\alpha}(0, \pm 1) \quad . \qquad (5.8.10)$$

Note in the above that we have discarded the $y_j^{\lambda}(0, \pm 1) \, m_K^{\alpha}(1,0)$ term since $\vec{m}^{\alpha}$ is minute at an optical frequency.

The high frequency component of the internal coordinate $\vec{y}^{\lambda}(1,0)$ is eliminated by means of Eq. (5.2.5) which at optical frequencies reduces to

$$y_j^{\lambda}(1,0) = \epsilon_0 \sum_{\gamma} \Upsilon_{jc}^{\lambda\gamma}(\omega^{\Theta}) \, q^{\gamma} \, E_c(1,0) \quad . \qquad (5.8.11)$$

Since the expression (5.8.10) is already bilinear in the field variables, we have omitted the last term in Eq. (5.2.5). When the above result is inserted into Eq. (5.8.10) and the latter in turn inserted into Eq. (5.8.8), we see that the nonlinear polarization is expressible in the form

$$P_i^{NL}(\omega^\varphi) = - \epsilon_0^2 \sum_{\substack{\mu,\nu \\ \gamma,\lambda}} q^\mu \, Y_{ia}^{\mu\nu}(\omega^\varphi) \, Y_{jc}^{\lambda\gamma}(\omega^\theta) \, q^\gamma$$

$$\{ \sum_\xi 3^{(3000)}K_{ajk}^{\nu\lambda\xi} \, Y_k^\xi(\omega) + \sum_\alpha {}^{(2010)}K_{ajk}^{\nu\lambda\alpha} m_k^\alpha(\omega) \} E_c(\omega^\theta) \; . \tag{5.8.12}$$

In the above, it is understood that ω can be a positive or negative frequency. With the aid of Eqs. (5.6.22), (5.6.24), (5.6.26) and (5.6.27), the nonlinear polarization may be reexpressed as

$$P_a^{(+)}(\vec{k}^\varphi,t) = \epsilon_0 \; [\sum_\mu A_{abc}^{\vec{k}\mu} \, Y_c^\mu \, (\vec{k},t)$$

$$+ \sum_\alpha C_{abc}^{\vec{k}\alpha} m_c^\alpha(\vec{k},t) + D_{abc} E_c(\vec{k},t)] \; E_b^\theta(\omega^\theta) \; e^{-i\omega^\theta t} \quad , \tag{5.8.13}$$

where $\vec{k} = \vec{k}^\varphi - k^\theta$ is the magneto-polariton wave vector. The coefficients $A_{abc}^{\vec{k}\mu}$ and $C_{abc}^{\vec{k}\alpha}$ are obtained by comparison with Eq. (5.8.12), and the term in E_c is the electronic response which we have hitherto not accounted for. If we remember that

$$\vec{y}^{\mu}(\vec{K},t)^{\dagger} = \vec{y}^{\mu}(-\vec{K},t) \quad , \tag{5.8.14}$$

with similar relationships for $\vec{m}^{\alpha}$ and $\vec{E}$, we can suppress the index $\vec{K}$ and write the second order susceptibility tensor of Eq. (5.6.30) as

$$\chi_{ab} = \sum_{\mu} A^{\mu}_{abc}\, y^{\mu}_{c} + \sum_{\alpha} C^{\alpha}_{abc}\, m^{\alpha}_{c} + D_{abc}\, E_{c} \quad . \tag{5.8.15}$$

With the aid of Eqs. (5.6.32), (5.7.11) and (5.8.15), the spectrum can be expressed by

$$\begin{aligned} J(\omega + \omega^{\theta}) = 4\hbar n(\omega)\ \mathrm{Im}\ [& (A^{\mu}_{ijk})^{*} A^{\nu}_{abc}\, T_{\nu c \mu^{\dagger} k^{\dagger}} \\ & + D^{*}_{ijk} D_{abc}\, T^{\dagger}_{ck} + (C_{ijk})^{*} C^{\beta}_{abc}\, T_{\beta c \alpha^{\dagger} k^{\dagger}} \\ & + (A^{\mu}_{ijk})^{*} D_{abc}\, T_{c \mu^{\dagger} k^{\dagger}} + (A^{\mu}_{ijk})^{*} C^{\beta}_{abc}\, T_{\beta c \mu^{\dagger} k^{\dagger}} \\ & + D^{*}_{ijk} A^{\nu}_{abc}\, T_{\nu c k^{\dagger}} + (C^{\alpha}_{ijk})^{*} A^{\nu}_{abc}\, T_{\nu c \alpha^{\dagger} k^{\dagger}} \\ & + D^{*}_{ijk} C^{\beta}_{abc}\, T_{\beta c k^{\dagger}} + (C^{\alpha}_{ijk})^{*} D_{abc}\, T_{c \alpha^{\dagger} k^{\dagger}}] \\ & e^{\varphi}_{a}\, e^{\theta}_{b}\, e^{\varphi}_{i}\, e^{\theta}_{j} \quad . \end{aligned} \tag{5.8.16}$$

The only quantities that remain to be calculated are the response functions T.

B. The Response Functions

In brief, $T_{\nu c\mu^+ k}$ is the linear response in y_c^ν to the application of an infinitesimal force F_k^μ which acts on the μ-th internal coordinate along the k-th direction. T_{ck^+} is the linear response in the electric field component E_c to the application of an infinitesimal external polarization P_k^{ext} along the k-th direction. $T_{\beta c\alpha^+ k^+}$ is the linear response in the sublattice magnetization m_c^β to the application of an infinitesimal external magnetic field $(B_k^\alpha)^{ext}$ which acts on the α sublattice magnetization. $T_{c\mu^+ k^+}$ is the linear response in the electric field E_c to the force F_k^μ. $T_{\beta c\mu^+ k^+}$ is the linear response in the sublattice magnetization m_c^β to the force F_k^μ. $T_{\nu ck^+}$ is the linear response in the internal coordinate y_c^ν to the polarization P_k^{ext}. $T_{\nu c\alpha^+ k^+}$ is the linear response in the internal coordinate y_c^ν to the magnetic field $(B_k^\alpha)^{ext}$. $T_{\beta ck^+}$ is the linear response in the sublattice magnetization m_c^β to the polarization P_k^{ext}. $T_{c\alpha^+ k^+}$ is the linear response in the electric field E_c to the magnetic field $(B_k^\alpha)^{ext}$.

If an electric field is driven by an infinitesimal external polarization P^{ext}, it obeys a wave equation of the form

$$\underline{\alpha}(\vec{K},\omega) \cdot \vec{E} = \vec{P}^{ext}/\epsilon_0 \quad , \qquad (5.B.17)$$

where the dyadic $\underline{\alpha}$ is obtained from Eq. (5.3.12) by making the replacement $\nabla \rightarrow i\vec{k}$. Here $\vec{P}^{ext} = \vec{P}^{ext}(\vec{k},\omega)$ and $\vec{E} = \vec{E}(\vec{k},\omega)$ are coefficients in an expansion such as in Eq. (5.6.22). The electric field response is therefore given by

$$T^{EP}_{ck^+} \equiv T_{ck^+} = E_c/P^{ext}_k = (\underline{\alpha})^{-1}_{ck}/\epsilon_0 \quad , \tag{5.8.18}$$

where the superscripts EP are meant as an aid in remembering that this is the linear response in the electric field to an external polarization. With the aid of Eqs. (5.4.13) and (5.6.11), the inverse dyadic is given by

$$(\underline{\alpha}^{-1})_{ck} = \sum_{\varphi=1,2} \frac{(\det \underline{k}_m) e^{\varphi}_c e^{\varphi}_k [(\hat{e}^{\varphi}\cdot\underline{k}_m\cdot\hat{e}^{\varphi})(\hat{s}^{\varphi}\cdot\underline{k}_m\cdot\hat{s}^{\varphi}) - (\hat{e}^{\varphi}\cdot\underline{k}_m\cdot\hat{s}^{\varphi})^2]^{-1}}{(ck/\omega)^2 - (n^{\varphi})^2}$$

$$- \frac{s_c s_k}{\hat{s}\cdot\underline{k}_e\cdot\hat{s}} \quad . \tag{5.8.19}$$

The dielectric tensor $\underline{k}_e$ should be thought of as containing an electronic contribution; i.e. the electric susceptibility of Eq. (5.2.15) is to be supplimented by the background susceptibility $\underline{x}^{el}$ produced by the essentially instantaneous electroniç response.

The Maxwell equation (3.4.11) expressed in terms of its Fourier components in space and time, can be written

$$i\,\vec{\kappa} \times \vec{E}(\vec{\kappa},\omega) = i\,\omega\vec{B}(\vec{\kappa},\omega) \quad . \qquad (5.8.20)$$

Thus, with the aid of Eq. (5.8.18) we have

$$B_i = (\kappa/\omega)\ \epsilon_{ijl}\ s_j\ T^{EP}_{la^+}\ P^{ext}_a \quad . \qquad (5.8.21)$$

The linear response in the magnetic field to an externally applied polarization is therefore given by

$$T^{BP}_{ia^+} \equiv B_i/P^{ext}_a = (\kappa/\omega)\ \epsilon_{ijl}\ s_j\ T^{EP}_{la^+} \quad . \qquad (5.8.22)$$

This response function will be of help in calculations to follow.

When an infinitesimal force of the form $F^{\mu}_i\, e^{-i\omega t}$ is added to the internal coordinate equation of motion Eq. (5.2.3), Eq. (5.2.5) is modified to

$$y^{\nu}_k = \epsilon_0 \sum_{\lambda} \Upsilon^{\nu\lambda}_{kc}\ [\ q^{\lambda}\ E_c - \sum_{\beta}\ {}^{(1010)}K^{\lambda\beta}_{cl}\ m^{\beta}_l + F^{\lambda}_c\ ] \quad . \qquad (5.8.23)$$

Eliminating y^{μ}_j from Eq. (5.2.6), (the magnetic equation of motion), with the aid of the above expression, we obtain

$$m^{\alpha}_i = (\underline{1} - \underline{k}^{-1}_m)^{\alpha}_{ik}\ B_k/\mu_0 + (\underline{x}_{me})^{\alpha}_{ik}\ \epsilon_0\ E_k$$

$$- (\epsilon_0/\mu_0) \sum_{\beta,\mu,\lambda} I^{\alpha\beta}_{ij}\ \epsilon_{jrt}\ m^{S\beta}_r\ {}^{(1010)}K^{\mu\beta}_{lt}\ \Upsilon^{\mu\lambda}_{lk}\ F^{\lambda}_k \quad ,$$

(5.8.24)

where

$$(1 - \underline{K}_m^{-1})_{ik}^{\alpha} \equiv \sum_{\beta} I_{ij}^{\alpha\beta} \epsilon_{jlk} m_l^{S\beta} \quad , \tag{5.8.25}$$

$$(x_{me})_{ik}^{\alpha} \equiv -1/\mu_0 \sum_{\beta,\mu,\lambda} I_{ij}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta} \, {}^{(1010)}K_{lt}^{\mu\beta} \, Y_{lk}^{\mu\lambda} q^{\lambda} \quad . \tag{5.8.26}$$

Eliminating $\vec{m}^{\beta}$ from Eq. (5.8.23) by means of the expression (5.8.24) yields for the internal coordinate

$$y_i^{\mu} = \epsilon_0 \, (\underline{x}_e)_{ik}^{\mu} E_k + (\underline{x}_{em})_{ik}^{\mu} B_k/\mu_0$$

$$+ \left(\sum_{\lambda} \epsilon_0 Y_{ik}^{\mu\lambda} + \sum_{\substack{\nu,\tau,\lambda \\ \alpha,\beta}} (\epsilon_0^2/\mu_0) Y_{ic}^{\mu\nu} \, {}^{(1010)}K_{ca}^{\nu\alpha} I_{aj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta} \, {}^{(1010)}K_{lt}^{\tau\beta} Y_{lk}^{\tau\lambda} \right) F_k^{\lambda} \, , \tag{5.8.27}$$

where

$$(\underline{x}_e^{\mu})_{ik} \equiv \sum_{\nu} Y^{\mu\nu} q^{\nu}$$

$$+ \sum_{\substack{\nu,\xi,\lambda \\ \alpha,\beta}} (\epsilon_0/\mu_0) Y_{ic}^{\mu\xi} \, {}^{(1010)}K_{cl}^{\xi\alpha} I_{lj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta} \, {}^{(1010)}K_{at}^{\nu\beta} Y_{ak}^{\nu\lambda} q^{\lambda} \, , \tag{5.8.28}$$

$$(\underline{x}_{em})_{ik}^{\mu} \equiv -\epsilon_0 \sum_{\lambda,\alpha,\beta} Y_{ic}^{\mu\lambda} \, {}^{(1010)}K_{cl}^{\lambda\alpha} I_{lj}^{\alpha\beta} m_r^{S\beta} \epsilon_{jrk} \quad . \tag{5.8.29}$$

The polarization with the external force $\vec{F}^{\lambda}$ present, is obtained with the aid of Eqs. (3.4.2), (5.2.15), (5.2.16) and (5.8.27)-(5.8.29):

$$P_i = \sum_\mu q^\mu y_i^\mu = \epsilon_0(\underline{x}_e)_{ik}E_k + (\underline{x}_{em})_{ik}B_k/\mu_0$$

$$+ \{\sum_{\mu,\lambda} \epsilon_0 q^\mu(\Upsilon_{ik}^{\mu\lambda} + \sum_{\substack{\nu,\tau\\ \alpha,\beta}} (\epsilon_0/\mu_0)\Upsilon_{ic}^{\mu\nu}{}^{(1010)} K_{ca}^{\nu\alpha} I_{aj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta}{}^{(1010)} K_{lt}^{\tau\beta}\Upsilon_{lk}^{\tau\lambda})\} F_k^\lambda \tag{5.8.30}$$

The term in F^λ acts as an extra P^{ext}, so that

$$T^{PF}_{i\lambda^+k^+} \equiv P_i^{ext}/F_k^\lambda$$

$$= \sum_\mu \epsilon_0 q^\mu(\Upsilon_{ik}^{\mu\lambda} + \sum_{\substack{\nu,\tau\\ \alpha,\beta}} (\epsilon_0/\mu_0)\Upsilon_{ic}^{\mu\nu}{}^{(1010)} K_{ca}^{\nu\alpha} I_{aj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta}{}^{(1010)} K_{lt}^{\tau\beta}\Upsilon_{lk}^{\tau\lambda}) \tag{5.8.31}$$

is the linear response in the external polarization P_i^{ext} to the application of the force F_k^λ.

When an infinitesimal external magnetic field is applied to the spin associated with sublattice β, the magnetic equation of motion (5.2.9) is modified to

$$m_i^\alpha = (\underline{1} - \underline{K}_m^{-1})_{ik}^\alpha B_k/\mu_0 + (\underline{x}_{me})_{ik}^\alpha \epsilon_0 E_k$$

$$+ \sum_\beta I_{ij}^{\alpha\beta} \epsilon_{jlk} m_l^{S\beta}(B_k^\beta)^{ext} , \tag{5.8.32}$$

where the definitions given by Eqs. (5.8.25) and (5.8.26) have been used. Note that this is the equation of motion obtained when a term $\sum_\beta \vec{m}^{T\beta} \cdot (\vec{B}^\beta)^{ext}$ is added to the lagrangian. When Eq. (5.8.32) is used to eliminate the sublattice magnetization from the internal coordinate equation of motion, Eq. (5.8.23) with $\vec{F}^\lambda = 0$, and when Eq. (3.4.2) is used, we obtain for the polarization, with the external field $(\vec{B}^\beta)^{ext}$ present, the expression

$$P_i = \sum_\mu q^\mu y_i^\mu$$

$$= \epsilon_0(\underline{x}_e)_{ik} E_k + (\underline{x}_{em})_{ik} B_k/\mu_0$$

$$- (\epsilon_0/\mu_0) \sum_{\substack{\mu,\lambda \\ \alpha,\beta}} q^\mu Y_{ic}^{\mu\lambda(1010)} K_{cl}^{\lambda\alpha} \epsilon_{jak} m_a^{S\beta} (B_k^\beta)^{ext} \quad . \tag{5.8.33}$$

Again the last term may be interpreted as an extra external polarization which, in this case, is due to the presence of the spin-phonon coupling. Thus,

$$T^{PB}_{i\beta^+k^+} \equiv P_i^{ext}/(B_k^\beta)^{ext}$$

$$= -(\epsilon_0/\mu_0 \sum_{\mu,\lambda,\alpha}) q^\mu Y_{ic}^{\mu\lambda(1010)} K_{cl}^{\lambda\alpha} I_{lj}^{\alpha\beta} \epsilon_{jak} m_a^{S\beta} \tag{5.8.34}$$

is the linear response in the external polarization P_i^{ext} to the application of the magnetic field $(B_k^\beta)^{ext}$.

The response functions appearing in Eq. (5.8.16) can now be evaluated. With the aid of Eqs. (5.8.18), (5.8.22), (5.8.27) and (5.8.31), the linear response in the internal coordinate to the force F_k^μ is given by

$$T_{\nu c\mu^+ k^+} \equiv y_c^\nu / F_k^\mu$$

$$= \epsilon_0 (\underline{x}_e)_{cl}^\nu T^{EP}_{lj^+} \; T^{PF}_{j\mu^+ k^+} + 1/\mu_0 (\underline{x}_{em})_{cl}^\nu T^{BP}_{lj^+} \; T^{PF}_{j\mu^+ k^+}$$

$$+ (\epsilon_0 \; Y_{ck}^{\nu\mu} + \sum_{\substack{\lambda,\tau \\ \alpha,\beta}} (\epsilon_0^2/\mu_0) Y_{ci}^{\nu\lambda^{(1010)}} K_{ia}^{\lambda\alpha} I_{aj}^{\alpha\beta} \epsilon_{jrt} m_r^{S\beta^{(1010)}} K_{lt}^{\tau\beta} Y_{lk}^{\tau\mu}) \quad . \tag{5.8.35}$$

The linear response in the electric field to the external polarization, T_{ck^+}, is given by Eq. (5.8.18). The linear response in the sublattice magnetization to the external magnetic field $(B_k^\alpha)^{ext}$ is given with the aid of Eqs. (5.8.18), (5.8.22), (5.8.32) and (5.8.34) by

$$T_{\beta c\alpha^+ k^+} \equiv m_c^\beta / (B_k^\alpha)^{ext}$$

$$= (\underline{1} - \underline{K}_m^{-1})_{cl}^\beta (1/\mu_0) T^{BP}_{lj^+} \; T^{PB}_{j\alpha^+ k^+}$$

$$+ \epsilon_0 (\underline{x}_{me})_{cl}^\beta \; T^{EP}_{lj^+} \; T^{PB}_{j\alpha^+ k^+}$$

$$+ \sum_\alpha I_{cj}^{\beta\alpha} \; \epsilon_{jlk} \; m_l^{S\alpha} \quad . \tag{5.8.36}$$

The linear response in the electric field to the external force F_κ^μ is given with the aid of Eqs. (5.8.18) and (5.8.31) by

$$T_{c\mu^+\kappa^+} \equiv E_c/F_\kappa^\mu = (E_c/P_j^{ext})\ (P_j^{ext}/F_\kappa^\mu)$$

$$= T^{EP}_{cj^+}\ T^{PF}_{j\mu^+\kappa^+} \quad . \tag{5.8.37}$$

The linear response in the sublattice magnetization to the external force F_κ^μ is given with the aid of Eqs. (5.8.18), (5.8.22), (5.8.24) and (5.8.31) by

$$T_{\beta c\mu^+\kappa^+} \equiv m_c^\beta/F_\kappa^\mu = 1/\mu_0\ (\underline{1} - \underline{K}_m^{-1})^\beta_{cj}\ T^{BP}_{jl^+}\ T^{PF}_{l\mu^+\kappa^+}$$

$$+ \epsilon_0(\underline{x}_{me})^\beta_{cj} T^{EP}_{jl^+} T^{PF}_{l\mu^+\kappa^+}$$

$$- (\epsilon_0/\mu_0) \sum_{\alpha,\lambda} I^{\beta\alpha}_{cj} \epsilon_{jrt} m_r^{S\alpha^{(1010)}} K^{\lambda\alpha}_{lt} Y^{\lambda\mu}_{l\kappa} \quad . \tag{5.8.38}$$

The linear response in the internal coordinate to the external polarization P_κ^{ext} is given with the aid of Eq. (5.8.27) with $\vec{F}^\lambda = 0$ and Eqs. (5.8.18) and (5.8.22) by

$$T_{\nu c\kappa^+} \equiv Y_c^\nu/P_\kappa^{ext}$$

$$= \epsilon_0(\underline{x}_e)^\nu_{cl} T^{EP}_{l\kappa^+} + 1/\mu_0(\underline{x}_{em})^\nu_{cl} T^{BP}_{l\kappa^+} \quad . \tag{5.8.39}$$

The linear response in the internal coordinate to the external magnetic field $(B^{\alpha}_{k})^{ext}$ is given with the aid of Eqs. (5.8.18), (5.8.22), (5.8.33) and (5.8.34) by

$$T_{\nu c\alpha^{+}k^{+}} \equiv Y^{\nu}_{c}/(B^{\alpha}_{k})^{ext}$$

$$= \epsilon_0(\underline{x}_e)^{\nu}_{cj}\ T^{EP}_{jl^{+}}\ T^{PB}_{l\alpha^{+}k^{+}}$$

$$+ 1/\mu_0(\underline{x}_{em})^{\nu}_{cj} T^{BP}_{jl^{+}}\ T^{PB}_{l\alpha^{+}k^{+}}$$

$$- (\epsilon_0/\mu_0) \sum_{\lambda,\beta} Y^{\nu\lambda(1010)}_{cr}\ K^{\lambda\beta}_{rl} I^{\beta\alpha}_{lj}\ \epsilon_{jak}\ m^{S\alpha}_{a} \quad . \tag{5.8.40}$$

The linear response in the sublattice magnetization to the external polarization P^{ext}_{k} is given with the aid of Eqs. (5.8.18), (5.8.22) and (5.8.24) with $F^{\lambda}_{k} = 0$ by

$$T_{\beta ck^{+}} \equiv m^{\beta}_{c}/P^{ext}_{k}$$

$$= 1/\mu_0(\underline{1} - \underline{K}^{-1}_{m})^{\beta}_{cj}\ T^{BP}_{jk^{+}} + \epsilon_0(\underline{x}_{me})^{\beta}_{cj}\ T^{EP}_{jk^{+}} \quad . \tag{5.8.41}$$

Finally, the linear response in the electric field to the external magnetic field $(B^{\alpha}_{k})^{ext}$ is given with the aid of Eqs. (5.8.18) and (5.8.34) by

$$T_{c\alpha^{+}k^{+}} \equiv E_c/(B^{\alpha}_{k})^{ext} = (E_c/P^{ext}_{j})\ (P^{ext}_{j}/(B^{\alpha}_{k})^{ext})$$

$$= T^{EP}_{cj^+} \, T^{PB}_{j\alpha^+k^+} \quad . \qquad (5.8.42)$$

Equations (5.8.35)-(5.8.42) combined with Eqs. (5.8.18), (5.8.22), (5.8.31) and (5.8.34) are to be inserted into the spectrum (5.8.16) which in turn is to be inserted into Eq. (5.6.18) to obtain the outside scattered power ratio from an anisotropic magnetic medium.

VI. SUMMARY AND CONCLUDING REMARKS

A complete macroscopic Lagrangian theory of linear and nonlinear electrodynamics has been constructed for an anisotropic magnetic insulator.

At first glance it might be supposed that an orientational average over the external vectors $\tilde{e}^J$ could be performed on the symmetry breaking term $\tilde{K}$ of Eq. (2.1.2) which would achieve rotational invariance for the Lagrangian. However, such an orientational average vanishes. Moreover, the variation of an orientationally averaged $\tilde{K}$ also appears to vanish. Thus, it seems that the Lagrangian theory must retain this symmetry breaking term.

In Chapter III where the results of Chapter II are applied to an elastic magnetic dielectric, it appears that fundamental quantities for the system such as the momentum density, the stress tensor, the energy density, the Poynting vector, the angular momentum density and the angular momentum flow tensor contain no direct contributions which arise from the symmetry breaking term. At present, it seems that the only function that the symmetry breaking term provides, is the generation of the precession equation and the consequent identification of the magnetization $\vec{m}(\vec{z},t)$ and its time derivative $\dot{\vec{m}}(\vec{z},t)$, as generalized coordinates and velocitites respectively. Since all the generalized coordinates and velocities describing a system must be identified in order to calculate the aforementioned fundamental quantities (the stress tensor, etc.), it is only in this indirect fashion that the symmetry breaking term has helped to determine these quantites.

In Chapter IV, the general theory is linearized. The resulting linearized equations of motion are the jump-off point for describing relevant phenomena on the linear level. It remains to apply these results to such problems as linear magnetoelasticity and magnetoelectricity. It is hoped that the spontaneous magnetic field and magnetic moment and the proper consideration of the rotational part of the elastic deformation will yield results not previously reported.

The linearized equations of motion are also the starting point for studying nonlinear effects by means of an iterative technique. The specific nonlinear effect of light scattering by magneto-polaritons is investigated in Chapter V.

Appendix A: Calculation of Unknown Multipliers

The matrix elements Θ_{ij} defined by Eq. (2.3.13) are explicitly given by $(i,j = 1,2,3)$

$$\Theta_{ij} \equiv \{\Theta_i , \int \Theta_j \, d^3\vec{z}\}$$

$$= \frac{1}{3\gamma} \sum_J \left[\frac{(S_{iJ}\,\epsilon_{jlk} - S_{jJ}\,\epsilon_{ilk})\, S_{lJ}\, m_k + 2(m_k\, S_{kJ})\,\epsilon_{ijl}\, S_{lJ}}{|\vec{m}|^2 - (m_a\, S_{aJ})^2} \right.$$

$$\left. - \frac{2(m_a\, S_{aJ}) S_{lJ}\, m_k (\epsilon_{jlk}[\, m_i - (m_b\, S_{bJ})\, S_{iJ}] - \epsilon_{ilk}[m_j - (m_c\, S_{cJ}) S_{jJ}])}{(|\vec{m}|^2 - (m_d\, S_{dJ})^2)^2} \right] , \tag{A.1}$$

and

$$\Theta_{i4} \equiv \{\Theta_i , \int \Theta_4 \, d^3\vec{z}\} = -2m_i \, , (i = 1,2,3) \quad . \tag{A.2}$$

The four simultaneous equations (2.3.11) and (2.3.12) are also explicitly written out as

$$\Theta_{12}\, v_2 + \Theta_{13}\, v_3 - 2m_1\, v_4 = -B_1^{eff} \quad , \tag{A.3a}$$

$$\Theta_{21}\, v_1 + \Theta_{23}\, v_3 - 2m_2\, v_4 = -B_2 eff \quad , \tag{A.3b}$$

$$\Theta_{31}\, v_1 + \Theta_{32}\, v_2 - 2m_3\, v_4 = -B_3^{eff} \quad , \tag{A.3c}$$

$$2m_1\, v_1 + 2m_2\, v_2 + 2m_3\, v_3 = 0 \quad . \tag{A.3d}$$

The above equations are solved in a straightforward manner yielding for the unknown multipliers

$$v_1 = \frac{m_2 B_3^{eff} - m_3 B_2^{eff}}{D} , \tag{A.4a}$$

$$v_2 = \frac{m_3 B_1^{eff} - m_1 B_3^{eff}}{D} , \tag{A.4b}$$

$$v_3 = \frac{m_1 B_2 eff - m_2 B_1^{eff}}{D} , \tag{A.4c}$$

$$v_4 = (\frac{1 + \Upsilon \theta_{12} m_3 - \Upsilon \theta_{13} m_2}{2m_1}) B_1^{eff}$$

$$+ \frac{\Upsilon \theta_{13}}{2} B_2^{eff} - \frac{\Upsilon \theta_{12}}{2} B_3^{eff} , \tag{A.4d}$$

where

$$D \equiv m_2 \theta_{13} + m_1 \theta_{32} + m_3 \theta_{21} = \frac{1}{\Upsilon} . \tag{A.5}$$

The antisymmetry of the matrix θ_{ij} has been used in reducing the above expressions. The denominator D was evaluated with the aid of Eq. (A.1), the property of orthogonality of the matrix S_{ij}, and some nontrivial algebra. Thus, the unknown multipliers have been determined to be

$$v_i = \Upsilon \epsilon_{ijk} m_j B_k^{eff} (i = 1,2,3) , \tag{A.6}$$

and v_4 is given by Eq. (A.4d).

It should be clear that in solving the simultaneous equations (A.3), we have also solved for the matrix (which is also antisymmetric) inverse to θ_{ij}. It follows from inspection of Eqs. (A.3) and (A.4) that this inverse may be expressed as

$$\theta^{-1}_{ij}(\vec{z}) = \gamma\ \epsilon_{ijk}\ m_k(\vec{z})\ ,\ i,j \leq 3\quad , \tag{A.7}$$

$$\theta^{-1}_{14}(\vec{z}) = \left(\frac{1 + \gamma\ \theta_{12}\ m_3 - \gamma\ \theta_{13}\ m_2}{2m_1} \right) \quad , \tag{A.8}$$

$$\theta^{-1}_{24}(\vec{z}) = \frac{\gamma\ \theta_{13}}{2} \quad , \tag{A.9}$$

$$\theta^{-1}_{34}(\vec{z}) = -\frac{\gamma\ \theta_{12}}{2} \quad . \tag{A.10}$$

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