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OPTICAL PULSE PROPAGATION AND GYROTROPIC EFFECTS
IN SPATIALLY DISPERSIVE MEDIA

by

Ashok Puri

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Abstract

OPTICAL PULSE PROPAGATION AND GYROTROPIC EFFECTS IN SPATIALLY DISPERSIVE MEDIA

by

Ashok Puri

Adviser: Professor Joseph L. Birman

This thesis is concerned with an investigation of topics in the electrodynamics of spatially dispersive media.

We examine the propagation of electromagnetic fields in the non-local, spatially dispersive media in which exciton polaritons are formed. Numerical estimates of energy-transport, group and signal velocities near exciton resonance are carried out. Studies on energy velocity are made on the assumption that the propagating modes in the medium are plane waves. As a next step, we examine Gaussian pulse propagation in spatially dispersive media, using a Fourier Transform method. We obtain expressions and results for velocity of peak propagation and distortion of the pulse.

As a problem of weak spatial dispersion, wave propagation in an optically active medium is examined from a phenomenological approach. We retain k-linear terms in the

inverse dielectric function. Additional boundary conditions are obtained in a mathematically consistent fashion. Reflectivity is calculated and numerical results are obtained.

The problem of surface waves in gyrotropic media is considered and the dispersion equation for surface waves is studied. An Attenuated Total Reflection (ATR) model experiment is formulated and analyzed quantitatively.

For a model gyrotropic medium the question of extinction of the incident field is examined using an integral equation approach. The dispersion equation, the extinction condition and the additional boundary conditions are obtained.

DEDICATION

To my beloved parents

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I am deeply indebted to Professor Joseph L. Birman for introducing me to the field of non-local optics and supervising the work done in this thesis. I am grateful to him for his advice and encouragement throughout this work.

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CHAPTER I

INTRODUCTION

1.1. Introduction

In this thesis we shall examine a number of problems relating to the electrodynamics of bounded, spatially dispersive media. In such media, one has the constitutive relation which relates dielectric displacement vector, $\vec{D}(\vec{r},\omega)$ and electric field, $\vec{E}(\vec{r},\omega)$ in a non-local fashion

$$\vec{D}(\vec{r},\omega) = \int \epsilon(\vec{r},\vec{r}',\omega)\,\vec{E}(\vec{r}',\omega)\,dr', \tag{1.1}$$

where the tensor, $\epsilon(\vec{r},\vec{r}',\omega)$ is the dielectric function. This dielectric function in general takes into account the inhomogenities in space i.e. effect of presence of surface etc. In the present study we concentrate on propagation of electromagnetic waves in the bulk, semi infinite medium. Then we shall assume that far enough from the surface all the points in the bulk medium are equivalent and the dielectric function will be taken to depend upon $(\vec{r}-\vec{r}')$ only i.e. it is translationally invarient. As an equivalent statement of our approximation, consider the Fourier Transform of the general non-local dielectric function, $\epsilon(\vec{r},\vec{r}',\omega)$ of Eq. (1.1). This is

$$\epsilon(\vec{K},\vec{K}',\omega) = \int \epsilon(\vec{r},\vec{r}',\omega)\; e^{i(-\vec{K}\cdot\vec{r} + \vec{K}'\cdot\vec{r}')}\,dr\,dr' \tag{1.2}$$

Assuming the translationally invarient dielectric function, $\epsilon(\vec{r},\vec{r}',\omega) = \epsilon(\vec{r}-\vec{r}',\omega)$ we find from Eq. (1.2),

$$\epsilon(\vec{K},\vec{K}',\omega) = \epsilon(\vec{K},\omega)\ \delta(\vec{K}-\vec{K}') \tag{1.3}$$

Finally, Eq. (1.3) implies that $\vec{K}=\vec{K}'$, thus Umklapp processes are absent and we obtain the wave vector dependent dielectric function, $\epsilon(\vec{K},\omega)$. One of the important consequences of the wave vector dependence of the dielectric function is that, additional modes arise for transverse (and longitudinal) electromagnetic propagation as solutions of Maxwell's equations. Recall that in the usual frequency dependent (ordinary dispersive) case of $\epsilon(\omega)$ the transverse solutions are given as solutions of

$$K^2 = \frac{\omega^2}{c^2}\ \epsilon(\omega) \tag{1.4}$$

For spatial dispersion Eq. (1.4) becomes an equation of higher algebraic degree in K. One of the many new features resulting from this is manifest in the solution of the elementary reflection problem (Fresnel equations) where Maxwell's boundary conditions are not enough to solve the problem at the interface of a semi infinite spatially dispersive medium and vacuum. Thus Maxwell's boundary conditions (MBC) plus additional boundary conditions (ABC) are required to solve reflection and transmission problems.[1-10]

In this thesis we study several problems involving a wave vector dependent dielectric function. We consider topics in which strong spatial dispersion and weak spatial dispersion are manifest.

In Chapter II we study the propagation of electromagnetic fields in non-local spatially dispersive media in which exciton-polaritons are formed. In the vicinity of an exciton resonance, this corresponds to strong spatial dispersion. As is well known, there are several velocities, which have been associated with an electromagnetic wave propagating in dispersive media: the phase velocity, $V_P = \omega/K$; the group velocity, $V_G = \frac{d\omega}{dK}$; the signal velocity, V_S and the energy velocity, $V_E = |\vec{S}_E|/U$ where $\vec{S}_E$ is the Poynting flux vector and U is the energy density. Signal velocity, V_S can be determined in many cases, from the time when the main signal at frequency ω_L arrive at an interior point in the medium, with some predetermined detectable amplitude. These different velocities have been introduced and studied in frequency dispersive[11(a),13] as well as spatially dispersive media.[8] There has been considerable interest in the subject in recent years because of advances in experimental picosecond spectroscopy which have made possible picosecond time of flight experiments on materials like GaAs, CdSe, CuCl and CdS[14-17] in which exciton polaritons are formed. In this Chapter our objective is two-fold; first we study the propagation of electromagnetic energy in spatially dispersive media. Numerical estimates of energy, group and signal velocities are carried out near exciton resonance for material with GaAs parameters.[18] These studies are performed considering electromagnetic fields in the medium to be plane waves. Next we use a Fourier Transform method to study Gaussian pulse propagation in spatially dispersive media. We extend the analysis given by Garrett and McCumber[19] for the local, single wave case, to the multi-wave coupled polariton case, assuming normal incidence on a semi infinite dielectric medium. Frequency ranges close to exciton resonance and far from exciton resonance are studied. Numerical results are reported for the power spectrum, $P(z,\omega)$ and the amplitude profile, $f(z,t)$ using material parameters for CdS[20]. The pulse shape is studied for various pulse widths. Finally we draw conclusion about the velocity of the peak of the pulse propagating in spatially dispersive medium in the vicinity of exciton resonance and demonstrate the condition for the pulse to remain gaussian.

Chapters III and IV deal with an optically active gyrotropic medium which corresponds to weak wave vector dependence. In Chapter V we study a model optically active medium which exhibits spatial dispersion.

In the case of weak spatial dispersion one can expand the dielectric function $\epsilon(\vec{K},\omega)$ or inverse dielectric function $\epsilon^{-1}(\vec{K},\omega)$ in a power series in $\vec{K}$, and break off after the second or third term:

$$\epsilon_{ij}(\vec{K},\omega) = \epsilon_{ij}(\omega) + i\gamma_{ij\ell}(\omega) K_\ell + \alpha_{ij\ell m}(\omega) K_\ell K_m \qquad (1.5)$$

$$\epsilon^{-1}_{ij}(\vec{K},\omega) = \epsilon^{-1}_{ij}(\omega) + i\,\delta_{ij\ell}(\omega)\,K_\ell + \beta_{ij\ell m}(\omega)\,K_\ell K_m \qquad (1.6)$$

It is well known that linear terms in the dielectric function correspond to optical activity.[1,21] In Chapter III, we investigate the problem of wave propagation in gyrotropic media near a dipole transition frequency.[22] In the expression (1.6), we retain K -linear terms. The phenomenological constitutive relation is obtained, and the problem of additional boundary conditions is solved. The structure of the electromagnetic fields in the gyrotropic half space is obtained. Finally, we study the problem of reflection and refraction on gyrotropic half space.

In Chapter IV we study the surface wave dispersion relation in optically active crystals. In the ordinary frequency dispersive media, allowed surface modes can be determined by matching allowed modes inside and outside the crystal at the boundary of the medium using Maxwell's boundary conditions. This procedure gives a surface wave dispersion equation relating the allowed mode energies and the wave vector parallel to the surface. In optically active bounded crystals, the general mode structure has been recently studied by Pattanayak and Birman.[23] We briefly review the mode structure, we write down the dispersion relation obtained by the above authors in an approximate form. We formulate an experiment using the Attenuated Total Reflection (ATR) model geometry in order to detect these surface waves.

Finally in Chapter V, we investigate the problem of the extinction of electromagnetic fields in gyrotropic media which exhibit spatial dispersion. When an electromagnetic wave is incident on a material medium bounded by a closed surface, the incident wave is completely extinguished at every point in the medium and is replaced by another wave propagating with different velocity and in a different direction (refracted wave). This is known as the Ewald-Oseen extinction theorem.[11(b), 25, 26] The electric field in the medium is represented as superposition of incident field and the radiated field of excited dipoles. Birman and Sein[4] generalized the integral equation technique to spatially dispersive media. We employ their generalization to obtain the dispersion relation, extinction condition and additional boundary conditions required to solve the reflection and refraction from a half space which is optically active and which exhibits spatial dispersion.[27]

CHAPTER II

PULSE PROPAGATION IN SPATIALLY DISPERSIVE MEDIA

2.1 Introduction

The propagation of electromagnetic pulses in dispersive media has recently received a great deal of attention. It is well known[13] that the group velocity

$$V_G = \frac{d\omega}{dK} = \frac{c}{n(\omega)+\omega\frac{dn}{d\omega}} \tag{2.1}$$

describes the propagation of electromagnetic wave packets in a linear, dispersive, isotropic and non-absorbing media. For an absorbing dielectric, difficulties arise in relating V_G to the velocity of energy propagation. In this case the wave vector, $\vec{K}$ is complex and V_G is often taken as $d\omega/dK_r$, where K_r is the real part of the wave vector $\vec{K}$. In the region of strong dispersion V_G becomes greater than C or negative. The common belief is that the concept of group velocity breaks down.[11(a)] Sommerfeld and Brillouin[12] have shown that for step pulses which begin at some instant of time $t=0$ and then follows a sinusoidal shape i.e.

$$f(t) = \theta(t)\, \text{Sin}\,\omega t\,, \tag{2.2}$$

the original pulse becomes distorted as it propagates in the medium. Precursors of the pulse travel with various speeds (Sommerfeld Precursor; Brillouin Precursor; Exciton Precursor) and the main part of the pulse arrives at the "signal velocity" $V_S < C$.

In an absorbing dielectric the energy velocity V_E has been analysed by Loudon.[13] It is defined as the ratio of the Poynting flux $|\vec{S}_E|$ and total electromagnetic energy density, U.

Garrett and McCumber[19] have shown that for the case $\Gamma\tau \gg 1$, the pulse propagates with group velocity V_G even though $V_G > C$, $V_G = \pm\infty$ or $V_G < 0$. Here τ is the half width of the pulse and Γ is the half width of the absorption spectrum.

The purpose of this Chapter is to study pulse propagation in non-local, spatially dispersive media. The electrodynamics of spatially dispersive media has been a subject of study for the last 25 years.[1-10] It is well known that in crystals like CdS, GaAs CdSe and CuCl, exciton-polaritons are formed when light in a spectral region near the exciton enters the crystal. One characteristic effect of spatial dispersion is that at any frequency there is more than one travelling wave: each one with different phase velocity. These are coupled exciton-polaritons. The physical polariton is the correct linear combination of these waves with the coefficient of the linear combination determined by relevant boundary conditions.

Advances in picosecond optical measuring techniques have opened up the possibility of time of flight measurements of light pulses in semiconductors. In order to investigate the propagation of exciton-polaritons in spatially dispersive media, a direct approach of "injecting" an optical pulse in the medium has been used. Transit-time and pulse shape have been determined as the pulse propagates through the medium.[14-17] Recently there has been a time of flight experiment on an absorbing dielectric (GaP:N),[28(a)] which claims to verify the theoretical predictions of Garrett and McCumber.[19] However, the experimental resolution in that experiment is not completely sufficient to compare with the theory[19] and further work is awaited.[28(b)]

Section (2.2) deals with the steady state pulse propagation (plane waves) in dispersive media in terms of energy transport velocity V_E . First we give a brief review of Loudon's[13] formulation of energy transport velocity in a frequency dispersive dielectric. Next we present a formulation of propagation of energy transport in a spatially dispersive medium.[18]

In Section (2.3) we study the propagation of a Gaussian pulse in dispersive media. First we give a brief review of the formulation of Garrett and McCumber[19] for a frequency dispersive dielectric. In spatially dispersive media, away from an exciton resonance, propagation is mainly on the photon-like branch, and the Garrett and McCumber analysis can be carried over in the weak spatial dispersion limit. Close to the exciton resonance we are not able to carry out analytical work so that we give results of numerical study in various frequency limits.[20]

Finally Section (2.4) gives a brief discussion. Energy, group and signal velocities in non-local media are compared for model parameters of GaAs. Numerical results for for Gaussian pulse propagation in spatially dispersive media are presented. Finally, comparison is made with the recent time of flight experiments.

2.2 Energy Transport in Dispersive Media

In this section we study the propagation of electromagnetic energy through the dielectric medium. Energy associated with the electromagnetic field propagating in the dielectric medium is partially associated with the electromagnetic wave and the rest is carried by the excitations in the medium. Following Loudon,[11] first we shall present here a theory of energy propagation in absorbing local dielectric in the framework of single oscillator model. As a next step we shall generalize it to spatially dispersive media.[18]

2.2.1 Propagation of Electromagnetic Energy in Frequency Dispersive Media

Here we shall obtain the expression for velocity of energy transport, V_E in absorbing media. Let us consider absorbing, frequency dispersive medium, which will be represented by damped, non-interacting harmonic oscillators. An equation of motion of a typical oscillator in presence of electric field $\vec{E}$ is given by

$$M(\ddot{\vec{r}} + \Gamma\dot{\vec{r}} + \omega_0^2\vec{r}) = e\vec{E} \ , \qquad (2.3)$$

where r, M, Γ, ω_0 and e denote displacement, mass, damping constant, natural frequency and effective charge of the oscillator respectively. Changing the variables in Equation (2.3) by defining $\vec{\xi} = \sqrt{M}\,\vec{r}$, one can write

$$\ddot{\vec{\xi}} + \Gamma \dot{\vec{\xi}} + \omega_0^2 \vec{\xi} = \frac{1}{\sqrt{M}} \vec{E} . \tag{2.3a}$$

The induced polarization $\vec{P}$ is given by

$$\vec{P} = \frac{e\vec{\xi}}{\sqrt{M}\,V_a} + \frac{(\epsilon_\infty - 1)}{4\pi} \vec{E} , \tag{2.4}$$

where V_a is the volume of the oscillator and ϵ_∞ is the background dielectric constant. For an isotropic dielectric medium $\vec{\xi}$, $\vec{E}$ and $\vec{P}$ all points in same direction, and complex dielectric constant is

$$\epsilon(\omega) = 1 + 4\pi \frac{P}{E} . \tag{2.5}$$

Thus considering $\vec{\xi}$, $\vec{E}$, $\vec{P}$ as plane waves i.e. $e^{i(kz-\omega t)}$, we can write down the expression for the dielectric function as

$$\epsilon(\omega) = \epsilon_\infty + \frac{4\pi\alpha_0\omega_0^2}{(\omega_0^2 - \omega^2 - i\omega\Gamma)} , \tag{2.6}$$

where $4\pi\alpha_0 = 4\pi e^2 / M\omega_0^2 V_a$ is the oscillator strength. The refractive index is defined from ϵ by

$$\epsilon^{1/2}(\omega) = \eta' + i\chi \ , \tag{2.7}$$

where η' and χ are real and imaginary parts of refractive index respectively. Using Equations (2.6) and (2.7) we obtain

$$\eta'^2 - \chi^2 = \epsilon_\infty + \frac{4\pi\alpha_0(\omega_0^2-\omega^2)}{(\omega_0^2-\omega^2)^2+\Gamma^2\omega^2} \tag{2.8a}$$

$$2\eta'\chi = \frac{4\pi\alpha_0\omega\Gamma}{(\omega_0^2-\omega^2)^2+\Gamma^2\omega^2} \ . \tag{2.8b}$$

Using Maxwell's equations in Gaussian units we can write

$$\frac{c}{4\pi}\int_\sigma \vec{E}\times\vec{H}\cdot\hat{n}\,ds = -\frac{1}{4\pi}\int_V (\dot{\vec{E}}\cdot\vec{E} + \vec{H}\cdot\dot{\vec{H}} + 4\pi\,\vec{E}\cdot\dot{\vec{P}})\,dV \ , \tag{2.9}$$

where V is the volume and σ is the surface surrounding the volume. Using Equations (2.3a) and (2.4) we can write the term $\vec{E}\cdot\dot{\vec{P}}$ appearing in the right hand side of Equation (2.9) as

$$\vec{E}\cdot\dot{\vec{P}} = \vec{E}\cdot\left[\frac{(\epsilon_\infty-1)}{4\pi}\dot{\vec{E}} + \frac{\dot{\vec{\xi}}}{\sqrt{M}\,V_a}\right]$$

$$= \frac{d}{dt}\left[\frac{(\epsilon_\infty-1)}{8\pi}E^2 + \frac{1}{2V_a}(\dot{\xi}^2+\omega_0^2\xi^2)\right] + \frac{\Gamma\dot{\xi}^2}{V_a} \ . \tag{2.10}$$

We define the total energy density as

$$U = \frac{1}{2V_a}(\dot{\xi}^2 + \omega_0^2 \xi^2) + (\epsilon_\infty E^2 + H^2)\frac{1}{8\pi} . \tag{2.11}$$

Then we can write Equation (2.9) as

$$\int_\sigma \vec{S}_E \cdot da + \int_V \frac{\Gamma \dot{\xi}^2}{V_a} dv = -\int \dot{U} dv , \tag{2.12}$$

where $\vec{S}_E$ is defined as the Poynting flux. Here the left hand side of Equation (2.12) represents the rate of loss of energy. The first term represents the propagation of energy across the surface σ and the second term represents the loss of energy in volume V because of damping. The integration on the right hand side of Equation (2.12) represents the rate of change of energy stored in volume V . From Equation (2.12) we can write

$$\vec{\nabla} \cdot \vec{S}_E + \frac{d}{dt}(U_E + U_M) = -\frac{\Gamma \dot{\xi}^2}{V_a} , \tag{2.13}$$

where $\vec{S}_E$, U_E and U_M are given by,

$$\vec{S}_E = \frac{c}{4\pi} \vec{E} \times \vec{H} \tag{2.14a}$$

$$U_E = \frac{1}{8\pi}(\epsilon_\infty E^2 + H^2) \tag{2.14b}$$

$$U_M = V_a^{-1}(\dot{\xi}^2 + \omega_0^2 \xi^2) . \tag{2.14c}$$

Here U_E and U_M are electromagnetic and mechanical energy respectively.

Considering harmonic time dependence for $\vec{\xi}$, $\vec{P}$, $\vec{E}$ and $\vec{H}$, and taking the time average of the Poynting flux S_E and total energy density U, by using $H=(n'+i\chi)E$, and Equation (2.3a), one obtains,

$$\langle \vec{S}_E \rangle = \frac{cn'}{8\pi} |E|^2 \tag{2.15a}$$

$$\langle U \rangle = \frac{|E|^2}{8\pi} \left(\frac{2\omega n' \chi}{\Gamma} + n'^2 \right) . \tag{2.15b}$$

The energy transport velocity V_E defined as ratio of $|\langle \vec{S}_E \rangle|$ and $\langle U \rangle$ is given by,

$$V_E = c/(n' + 2\omega\chi/\Gamma) . \tag{2.16}$$

In the limit $\chi \longrightarrow 0$, V_E goes to c/n' , the phase velocity.

2.2.2 <u>Transport of Energy in Spatially Dispersive Media</u>

Here we shall study energy transport in spatially dispersive medium. Consider a model non-local semi-infinite medium occupying $z \geqslant 0$. An electromagnetic plane wave is incident normally on the medium and it gives rise to two transverse exciton-polariton waves propagating in the medium. The equation of motion for displacement in spatially dispersive medium can be taken as

$$\ddot{\vec{\xi}} + \Gamma \dot{\vec{\xi}} + \omega_0^2 \vec{\xi} - D\nabla^2 \vec{\xi} = \frac{e_T^* \vec{E}}{\sqrt{M^*}} , \qquad (2.17)$$

where $D = \hbar\omega_0 / M^*$, M^* is the exciton mass and e_T^* is the transverse effective charge. The dielectric function corresponding to Equation (2.17) for a non-local semi infinite medium in "dielectric approximation" is written as,

$$\epsilon(\vec{K},\omega) = \epsilon_\infty + \frac{4\pi\alpha_0 \omega_0^2}{(\omega_0^2 - \omega^2 - i\omega\Gamma + DK^2)} . \qquad (2.18)$$

Here we have considered that the dielectric function, which properly corresponds to an infinite medium, can be used to describe the semi infinite spatially dispersive medium. Our physical justification is that in the present study we shall concentrate on bulk propagation, so that non-translationally invarient contribution to $\epsilon(\vec{r}, \vec{r}')$ (or Umklapp effects) will be neglected. Also note that the wave number of a volume propagating wave is determined using the translationally invarient part of $\epsilon(\vec{r}, \vec{r}')$.

The physical polariton in a non-local medium is represented as

$$\vec{E}(z,\omega) = \sum_{j=1}^{2} \vec{E}_j \, e^{i K_j z} , \qquad (2.19)$$

where K_j is a solution of $(K_j/K_0)^2 = \epsilon(K_j,\omega)$; $K_0 = \omega/c$ and $\vec{E}_j$ are determined by relevant boundary conditions. Note that and correspond to Upper (UP) and Lower (LP) Polaritons respectively. Maddox and Mills,[29] and Bishop and Maraduddin[30] have shown that the Poynting flux $\vec{S}$ in the non-local medium is the sum of the electromagnetic Poynting vector $\vec{S}_E$ and mechanical (excitonic in our case) Poynting vector $\vec{S}_M$. The Poynting flux satisfies the equation[30]

$$\vec{\nabla}\cdot(\vec{S}_E + \vec{S}_M) + \frac{d}{dt}(U_E + U_M) = -\frac{\Gamma}{V_a}\dot{\xi}^2 - \frac{D}{V_a}\delta(z)\dot{\vec{\xi}}\cdot\frac{\partial\vec{\xi}}{\partial z}, \qquad (2.20)$$

where $\vec{S}_E$, $\vec{S}_M$, U_E and U_M are given by

$$\vec{S}_E = \frac{c}{4\pi}(\vec{E}\times\vec{H}) \qquad (2.21a)$$

$$\vec{S}_M = -(D/V_a)\sum_{\beta}\hat{x}_\beta\,\theta(z)\,\dot{\vec{\xi}}\cdot\frac{\partial\vec{\xi}}{\partial x_\beta} \qquad (2.21b)$$

$$U_E = (1/8\pi)(\epsilon_\infty E^2 + H^2) \qquad (2.21c)$$

$$U_M = V_a^{-1}[\dot{\xi}^2 + \omega_0^2\xi^2 + D(\vec{\nabla}\xi)^2]\,. \qquad (2.21d)$$

Using Equations (2.17) and (2.19) in (2.21) and taking a time average one obtains

$$|\langle\vec{S}_E\rangle| = \frac{c}{8\pi}\,\mathrm{Re}\left[\left(\sum_{j=1}^{2}E_j\,e^{ik_jz}\right)\left(\sum_{j=1}^{2}E_j^*n_j^*\,e^{-ik_j^*z}\right)\right] \qquad (2.22a)$$

$$|\langle \vec{S}_M \rangle| = \frac{\omega}{2D} \epsilon_\infty \frac{\Omega_p}{4\pi} \, Re\left[\left(\sum_{j=1}^{2} \frac{E_j e^{iK_j z}}{(K_j^2 - \gamma^2)} \right)\left(\sum_{j=1}^{2} \frac{K_j^* E_j^* e^{-iK_j^* z}}{(K_j^{*2} - \gamma^{*2})} \right)\right] \quad (2.22b)$$

$$\langle U_E \rangle = \frac{1}{16\pi} \, Re\left[\epsilon_\infty \left(\sum_{j=1}^{2} E_j e^{iK_j z} \right)\left(\sum_{j=1}^{2} E_j^* e^{-iK_j^* z} \right) + \left(\sum_{j=1}^{2} n_j E_j e^{iK_j z} \right)\left(\sum_{j=1}^{2} n_j^* E_j^* e^{-iK_j^* z} \right)\right] \quad (2.22c)$$

$$\langle U_M \rangle = \frac{\epsilon_\infty \Omega_p^2}{16\pi D^2} \left(\omega_0^2 + \omega^2 + D|\gamma|^2 \right) Re\left[\left(\sum_{j=1}^{2} \frac{E_j e^{iK_j z}}{(K_j^2 - \gamma^2)} \right) \left(\sum_{j=1}^{2} \frac{E_j^* e^{-iK_j^* z}}{(K_j^{*2} - \gamma^{*2})} \right)\right] . \quad (2.22d)$$

where $n_j = K_j / K_0$ are refractive indices, Ω_p is the plasma frequency $\Omega_p^2 = 4 e_T^{*2} / \epsilon_\infty M^* Va$ and $\gamma = [-(\omega_0^2 - \omega^2 - i\omega\Gamma)/D]^{1/2}$.

The energy velocity of the physical polariton is given by

$$V_E = |\langle \vec{S}_E + \vec{S}_M \rangle| / \langle U_E + U_M \rangle \quad . \quad (2.23)$$

As the next step one can write various contributions to by extracting Upper and Lower polariton contributions from $|\langle \vec{S}_E \rangle|$ and $|\langle \vec{S}_M \rangle|$, and forming $|\langle \vec{S}_{UP} \rangle|$ and $|\langle \vec{S}_{LP} \rangle|$ respectively. Similarly one can write $\langle U_{UP} \rangle$ and $\langle U_{LP} \rangle$ respectively. These expressions are given by

$$|\langle \vec{S}_{UP}\rangle| = \frac{c}{8\pi}\,\mathrm{Re}\left(E_1 E_1^* n_1^*\, e^{i(K_1-K_1^*)z}\right)$$

$$+\frac{\omega}{2D}\,\frac{\epsilon_\infty \Omega_p^2}{4\pi}\,\mathrm{Re}\left[\frac{E_1 E_1^* K_1^*\, e^{i(K_1-K_1^*)z}}{(K_1^2-\gamma^2)(K_1^{*2}-\gamma^{*2})}\right] \qquad \text{(2.24a)}$$

$$\langle U_{UP}\rangle = \frac{1}{16\pi}\,\mathrm{Re}\left[\epsilon_\infty\left(E_1^* E_1\, e^{i(K_1-K_1^*)z}\right) + E_1 E_1^* n_1 n_1^*\, e^{i(K_1-K_1^*)z}\right]$$

$$+\frac{\epsilon_\infty \Omega_p^2}{16\pi D^2}\left(\omega^2+\omega_0^2+D|\gamma|^2\right)\mathrm{Re}\left[\frac{E_1 E_1^*\, e^{i(K_1-K_1^*)z}}{(K_1^2-\gamma^2)(K_1^{*2}-\gamma^{*2})}\right] \qquad \text{(2.24b)}$$

$$V_{E(UP)} = |\langle S_{UP}\rangle| / \langle U_{UP}\rangle \qquad \text{(2.24c)}$$

$$|\langle \vec{S}_{LP}\rangle| = \frac{c}{8\pi}\,\mathrm{Re}\left(E_2 E_2^* n_2^*\, e^{i(K_2-K_2^*)z}\right)$$

$$+\frac{\omega}{2D}\,\frac{\epsilon_\infty \Omega_p^2}{4\pi}\,\mathrm{Re}\left[\frac{E_2 E_2^* K_2^*\, e^{i(K_2-K_2^*)z}}{(K_2^2-\gamma^2)(K_2^{*2}-\gamma^{*2})}\right] \qquad \text{(2.24d)}$$

$$\langle U_{LP}\rangle = \frac{1}{16\pi}\,\mathrm{Re}\left[\epsilon_\infty\left(E_2^* E_2\, e^{i(K_2-K_2^*)z}\right) + E_2 E_2^* n_2 n_2^*\, e^{i(K_2-K_2^*)z}\right]$$

$$+\frac{\epsilon_\infty \Omega_p^2}{16\pi D^2}\left(\omega^2+\omega_0^2+D|\gamma|^2\right)\mathrm{Re}\left[\frac{E_2 E_2^*\, e^{i(K_2-K_2^*)z}}{(K_2^2-\gamma^2)(K_2^{*2}-\gamma^{*2})}\right] \qquad \text{(2.24e)}$$

$$V_{E(LP)} = \langle S_{LP} \rangle / \langle U_{LP} \rangle \quad . \tag{2.24f}$$

where $V_{E(UP)}$ and $V_{E(LP)}$ are Upper and Lower Polariton contribtuions to energy velocity. Using the material parameters of GaAs, $\hbar\omega_0 = 1.515$ ev, $4\pi\alpha_0 = 0.0013$, $M^* = 0.6$ Me, Me = 0.5 Mev, $\epsilon_\infty = 12.55$, $\Gamma = 1\ cm^{-1}$, $\Omega_p^2/\omega_0^2 = 10^{-3}$, $Z = 3.7\ \mu$, these contributions of energy velocity are plotted in Fig. 1 as dashed curves against reduced frequency, $(\omega-\omega_0)/\omega_0$ in the resonance region. On the same Figure is plotted the group velocity, V_G for Upper and Lower polariton branches repsectively. The magnitude of signal velocity, V_S is also shown in the pseudo-gap region. We shall discuss Fig. 1 in more detail in Section 2.4.

2.3 Propagation of a Gaussian Pulse in Dispersive Media

In this section we review the theory of propagation of a Gaussian pulse in dispersive media. Propagation of a steady state Gaussina pulse through absorbing media was studied theoretically by Garrett and McCumber.[19] They demonstrated that under conditions $\Gamma\tau \gg 1$, or $\Gamma\tau \sim 1$, where Γ being the half width of the absorption spectrum and τ being half width of the pulse, the pulse remains substantially Gaussian and the peak of the pulse propagates with group velocity, V_G even if $V_G = \pm\infty$. Experimentally verification was claimed by Chu and Wong[28(a)] in a time of flight experiment on GaP:N in region of anomalous dispersion. The sensitivity of their experiment was insufficient to resolve pulse shapes, however, and further work is expected.[28(b)] In this Section we shall briefly review Garrett and McCumber's analysis for absorbing media. As a next step we shall generalize it to spatially dispersive media. Close to exciton resonance we must use numerical techniques to compute the amplitude, $f(z,t)$ for Upper and Lower polaritons respectively. Finally, we shall make comparisons with recent time of flight experiments.

2.3.1 Pulse Propagation in Absorbing Media

Let us consider a Gaussian pulse incident on a semi infinite $(z \geqslant 0)$ absorbing medium. The source field is locate at $z=0$. The amplitude of the field at the point, z and time, t is given by

$$f(z,t) = \tau (2\pi)^{-1/2} \int_{-\infty}^{\infty} f(z,\omega)\, e^{-i\omega t}\, d\omega \quad , \tag{2.25}$$

where,

$$f(z,\omega) = e^{i K(\omega) z} \; e^{-\frac{1}{2}(\omega - \bar{\omega})^2 \tau^2} \quad . \tag{2.25a}$$

Here $\bar{\omega}$ is the central frequency of the Gaussian and τ^{-1} is the half width of the pulse, $K(\omega)$ is the solution of the dispersion equation, $K(\omega) = \frac{\omega}{c}\sqrt{\epsilon(\omega)}$; $\epsilon(\omega)$ is the complex dielectric function for the absorbing medium defined by Equation (2.6). The refractive index $n(\omega)$ is given by $n(\omega) = \sqrt{\epsilon(\omega)}$.

Now we consider the case for which the spectral width of the pulse is substantially smaller than the atomic line width i.e. $\Gamma\tau \gg 1$. Under this condition the major contribution of Equation (2.25) comes from ω in the neighborhood of central frequency $\bar{\omega}$ of incident pulse. Thus we can expand $K(\omega)$ in Taylor series as

$$K(\omega) = K(\bar{\omega}) + (\omega-\bar{\omega}) \frac{d}{d\omega} K(\omega)\Big|_{\bar{\omega}} + \frac{1}{2}(\omega-\bar{\omega})^2 \frac{d^2}{d\omega^2} K(\omega)\Big|_{\bar{\omega}} + \cdots, \quad (2.26)$$

where from Equation (2.6), for the oscillator model,

$$\frac{1}{m!} \frac{d^m}{d\omega^m} (\omega n(\omega))\Big|_{\bar{\omega}} = \frac{(-1)^m \omega_p \omega_0}{(\bar{\omega} - \omega_0 + i\Gamma)^{m+1}}, \quad m \geq 2 \quad (2.26a)$$

Here $\omega_p = (2\pi\alpha_0 \omega_0^2)/\epsilon_\infty$.

The series (2.26) diverges if $(\omega-\bar{\omega})^2 > ((\bar{\omega}-\omega_0)^2+\Gamma^2)$ so that this expansion is only useful when the important frequencies of Equation (2.25) are those for which $(\omega-\bar{\omega})^2 \ll [(\bar{\omega}-\omega_0)^2 + \Gamma^2]$. For small z, the important frequencies are those for which $(\omega-\bar{\omega})^2\tau^2 \lesssim 1$ and with $\Gamma\tau \gg 1$, the Taylor's series (2.26) converges rapidly. Putting Equation (2.26) in (2.25) we get

$$f(z,t) = \tau(2\pi)^{-1/2}\int_{-\infty}^{\infty} d\omega\; e^{-i\omega t}\cdot e^{i\left[K(\bar{\omega}) + (\omega-\bar{\omega})\frac{dK}{d\omega}\big|_{\bar{\omega}}\right.}$$

$$\left. + \frac{(\omega-\bar{\omega})^2}{2}\frac{d^2K}{d\omega^2}\Big|_{\bar{\omega}}\right]z \times e^{-(\omega-\bar{\omega})^2\tau^2/2} \quad (2.27)$$

Substituting $(\omega-\bar{\omega}) = u$, we get

$$f(z,t) = \left(1 - \frac{zi}{\tau^2}\frac{d^2K}{d\omega^2}\Big|_{\bar{\omega}}\right)^{-1/2} e^{i(K(\bar{\omega})z - \bar{\omega}t)} \times$$

$$\times \exp\left[-\left(t - z\frac{dK}{d\omega}\Big|_{\bar{\omega}}\right)^2 \Big/ 2\tau^2\left(1 - \frac{iz}{\tau^2}\frac{d^2K}{d\omega^2}\Big|_{\bar{\omega}}\right)\right] . \quad (2.28)$$

Using the model dielectric function given by Equation (2.6) we can write $f(z,t)$ which can be shown to be of Gaussian form with shift in center of the packet and changed "full width at half maximum" FWHM in general. Garrett and McCumber[19] have carried out exact numerical study of $|f(z,t)|$ and compared it with these approximate analytical results. Their results reveal that the pulse remains substantially Gaussian and the peak of the pulse propagates with group velocity, V_G even $V_G > c$ or $\pm\infty$. Recently Chu and Wong[28(a)] have studied pulse propagation in an absorb- medium (GaP:N). The lack of distortion they report, however, seems questionable in view of comment by Katz and Alfano.[28(b)]

The measure pulse velocity in GaP:N with laser tuned to an A-exciton line, using a picosecond time of flight technique. They report the pulse propagates through the material with little pulse distortion and its peak propagates with the group velocity. The group velocity is measured separately by measuring the absorption coefficient $\alpha(\omega)$ from transmission measurement $I = I_0 e^{-\alpha(\omega) z}$ and the Kramer-Kronig relations are numerically applied to obtain the real part of $\sqrt{\epsilon(\omega)}$. K_r is given by $\omega n'(\omega)/c$ and $\Delta t = \Delta \ell / (d\omega/dK_r)$. Chu and Wong have verified the predictions of Garrett and McCumber that the peak of the pulse propagates with group velocity V_G , even though it is $> C$, or even $\pm \infty$.

2.3.2 Pulse Propagation Through a Non-Local Medium

Let us consider a Gaussian pulse incident on the semi infinite $z \geq 0$ spatially dispersive medium. The amplitude of the field at the point, z and time t is given by

$$f(z,t) = \tau (2\pi)^{-1/2} \int_{-\infty}^{\infty} d\omega \, e^{-i\omega t} \sum_{j=1}^{2} f_j(z,\omega) , \tag{2.29}$$

where

$$f_j(z,\omega) = A_j(\omega) \cdot e^{i K_j(\omega) z} \, e^{-(\omega-\bar{\omega})^2 \tau^2/2} \tag{2.29a}$$

Here $\bar{\omega}$ is the central frequency of the Gaussian and τ^{-1} is the half width of the pulse, $j = 1, 2$ correspond to the

Upper and Lower polaritons (UP and LP) respectively, K_j is the solution of the dispersion equation for transverse electromagnetic waves; $K^2 = K_0^2\ \epsilon(\vec{K}, \omega)$; $\epsilon(\vec{K}', \omega)$ is the dielectric function for spatially dispersive medium and is given in the "dielectric approximation" by Equation (2.18).

In Equation (2.29a) $A_j(\omega)$ are coupling constants to be determined by the full set of boundary conditions (Maxwell's plus Additional Boundary Conditions).

For frequency regimes, $\omega/\omega_0 \gg 1$, $\omega/\omega_0 \ll 1$ and $\omega_0 < \omega < \omega_\ell$ there is effectively a single polariton propagating and the Garrett and McCumber analysis can be applied. We discuss these regimes in this section. Assuming $\Gamma\tau \gg 1$, we can carry out an expansion of the wave vector $K(\omega)$ around $\bar{\omega}$,

$$K(\omega) = K(\bar{\omega}) + (\omega-\bar{\omega})\left.\frac{dK}{d\omega}\right|_{\bar{\omega}} + \frac{(\omega-\bar{\omega})^2}{2}\left.\frac{d^2K}{d\omega^2}\right|_{\bar{\omega}} + \cdots \quad (2.30)$$

For the oscillator model this series converges[19] for $(\omega-\bar{\omega})^2\tau^2 \lesssim 1$ and $\Gamma\tau \gg 1$. Thus we expand up to second degree in Equation (2.30). Putting Equation (2.30) in Equation (2.29) we get,

$$f(z,t) = \frac{\tau}{\sqrt{2\pi}}\int_{-\infty}^{\infty} d\omega\ e^{-i\omega t}\sum_{j=1}^{2} A_j(\omega)\ e^{i\left[K_j(\omega) + (\omega-\bar{\omega})\left.\frac{dK}{d\omega}\right|_{\bar{\omega}}\right.}$$

$$+ \frac{(\omega-\bar{\omega})^2}{2} \left.\frac{d^2K_j}{d\omega^2}\right|_{\bar{\omega}} \Big] z \quad \times e^{-(\omega-\bar{\omega})^2\tau^2/2} .$$

Substituting $(\omega-\bar{\omega}) = u$, we get,

$$f(z,t) = \frac{\tau}{\sqrt{2\pi}} \sum_{j=1}^{2} A_j(\bar{\omega})\, e^{i(K(\bar{\omega})z - \bar{\omega}t)} \times$$

$$\times \int_{-\infty}^{\infty} du \exp\left[-iut - u^2\frac{\tau^2}{2} + i\left(u\left.\frac{dK_j}{d\omega}\right|_{\bar{\omega}} + u^2\left.\frac{d^2K_j}{d\omega^2}\right|_{\bar{\omega}}\right)z\right] \quad (2.31)$$

Carrying out the integration we obtain

$$f(z,t) = \sum_{j=1}^{2} \left(1 - \frac{zi}{\tau^2} \left.\frac{d^2K_j}{d\omega^2}\right|_{\bar{\omega}}\right)^{-1/2} A_j(\bar{\omega})\, e^{i(K(\bar{\omega})z - \bar{\omega}t)} \times$$

$$\times \exp\left[-\left(t - z\left.\frac{dK_j}{d\omega}\right|_{\bar{\omega}}\right)/2\tau^2\left(1 - \frac{zi}{\tau^2}\left.\frac{d^2K_j}{d\omega^2}\right|_{\bar{\omega}}\right)\right]$$

$$= \sum_{j=1}^{2} f_j(z,t) \quad (2.32)$$

Next we proceed to discuss various limiting cases. We employ an expansion of $K_j(\omega)$ for various cases, following Frankel and Birman.[8]

Case I:

Above the exciton resonance, i.e. in the limit $\omega/\omega_0 \gg 1$, the dispersion $K_j(\omega)$ is given by:

$$K_1(\omega) = \sqrt{\epsilon_\infty}\,\frac{\omega}{c} - \frac{c}{D\sqrt{\epsilon_\infty}}\,\frac{\beta^2\omega_0^2}{(2\beta\omega + i\Gamma)} \tag{2.33a}$$

$$K_2(\omega) = \frac{\omega}{\sqrt{D}} + \frac{i\Gamma}{2\sqrt{D}} - \frac{\omega_0^2}{\sqrt{D}\,(2\omega - i\Gamma)}\,, \tag{2.33b}$$

where $\beta = 4\pi\alpha_0 D/c^2$.

Putting (2.33a) in Equation (2.32) we find

$$f_1(z,t) = \frac{1}{\sqrt{M}}\,\exp\left(-(\Delta_1 + i\Delta_2)\right) A_1(\bar{\omega})\,, \tag{2.34}$$

where

$$M = \left[\left(1 + \frac{z}{\tau^2}\,\frac{8c\beta^4\omega_0^2}{D}\,\Gamma\,\frac{(12\beta^2\bar{\omega}^2 - \Gamma^2)}{((2\beta\bar{\omega})^2 + \Gamma^2)^3}\right)\right.$$

$$\left. + i\left(\frac{z}{\tau^2}\,\frac{8c\beta^4}{D}\,\omega_0^2\,\frac{((2\beta\bar{\omega})^2 - 6\beta\bar{\omega}\Gamma^2)}{((2\beta\bar{\omega})^2 + \Gamma^2)^3}\right)\right] \tag{2.35a}$$

$$\Delta_1 = \frac{(t-\alpha)^2}{2\tau_2^2} + \left(\frac{U}{\gamma} + V\right)\frac{1}{2\tau_1^2} + W \tag{2.35b}$$

$$\tau_2^2 = \gamma^{-1}\,\tau_1^2 \tag{2.35c}$$

$$\tau_1^2 = \tau^2 |M|^2 \tag{2.35d}$$

$$\gamma = \left(1 + \frac{Z}{\tau^2} \frac{8c\beta^4\omega_0^2\Gamma}{D} \frac{(12\beta^2\bar{\omega}^2 - \Gamma^2)}{((2\beta\bar{\omega})^2 + \Gamma^2)^3}\right), \tag{2.35e}$$

and α , U , V , W and Δ_2 are functions of $\bar{\omega}$, Z and parameters appearing in the dielectric function $\epsilon(\vec{K},\omega)$ in Equation (2.18).

Similarly putting (2.33b) in Equation (2.32) we obtain

$$f_2(z,t) = \frac{1}{\sqrt{M'}} \exp(-(\Delta_1' + i\Delta_2')) A_2(\bar{\omega}) \tag{2.36}$$

where,

$$M' = \left[\left(1 - \frac{Z}{\tau^2} \frac{8\omega_0^2}{\sqrt{D}} \Gamma \frac{(12\bar{\omega}^2 - \Gamma^2)}{((2\bar{\omega})^2 + \Gamma^2)^3}\right)\right.$$

$$\left. + i\left(\frac{Z}{\tau^2} \frac{8\omega_0^2}{\sqrt{D}} \frac{((2\bar{\omega})^3 - 6\bar{\omega}\Gamma^2)}{((2\bar{\omega})^2 + \Gamma^2)^3}\right)\right] \tag{2.37a}$$

$$\Delta_1' = \frac{(t-\alpha')^2}{2\tau_2'^2} + \left(\frac{U'}{\gamma'} + V'\right)\frac{1}{2\tau_1'^2} + W' \tag{2.37b}$$

$$\tau_2'^2 = \gamma'^{-1} \tau_1'^2 \tag{2.37c}$$

$$\tau_1'^2 = \tau^2 |M'|^2 \tag{2.37d}$$

$$\gamma' = \left[1 - \frac{Z}{C^2} \frac{8\omega_0^2}{\sqrt{D}} \Gamma \frac{(12\bar{\omega}^2 - \Gamma^2)}{((2\bar{\omega})^2 + \Gamma^2)^3} \right] , \tag{2.37e}$$

and α', U', V', W' and Δ_2' are functions of $\bar{\omega}$, Z and parameters appearing in dielectric function, $\epsilon(\vec{K}, \omega)$.

Case II:

In the gap region $\omega_0 < \omega < \omega_\ell$ there is one polariton mode propagating in the medium and the amplitude of this mode can be calculated by considering the approximate form of the dispersion equation in the gap:

$$K_2(\omega) = \frac{1}{\sqrt{D}} \left[(\omega - \tilde{\omega})(\omega + \tilde{\omega}) \right]^{1/2} , \tag{2.38}$$

where,

$$\tilde{\omega} = -\frac{i\Gamma}{2} + \omega_0 \left[1 + \Delta - \Gamma^2/2\omega_0^2 \right]^{1/2}$$

and

$$\Delta = D\epsilon_\infty / c^2$$

In the limit $\omega \approx \tilde{\omega}$,

$$K_2(\omega) = \left(\frac{2\tilde{\omega}}{D}\right)^{1/2} (\omega - \tilde{\omega})^{1/2} . \tag{2.39}$$

Using Equations (2.39) and (2.32) we can write the amplitude in the gap region as:

$$f_2(z,t) = \frac{1}{\sqrt{M'}} \exp\left[-(\Delta_1' + i\,\Delta_2')\right] A_2(\bar{\omega}) , \qquad (2.40)$$

where,

$$M' = \left[\left(1 - \frac{z}{\tau^2 2\sqrt{2D}} \frac{\xi(\gamma^3 - 3\gamma\delta^2) + \eta(\delta^3 - 3\gamma^2\delta)}{(\gamma^2 + \delta^2)^3}\right)\right.$$

$$\left. + i\left(\frac{z}{\tau^2 2\sqrt{2D}} \frac{\eta(\gamma^3 - 3\gamma\delta^2) - \xi(\delta^3 - 3\gamma^2\delta)}{(\gamma^2 + \delta^2)^3}\right)\right] \qquad (2.41a)$$

$$\Delta_1' = \frac{(t-\alpha')^2}{2\tau_2'^2} + \left(\frac{U'}{\gamma'} + V'\right)\frac{1}{2\tau_1'^2} + W' \qquad (2.41b)$$

$$\tau_2'^2 = \tau_1'^2 \gamma'^{-1} \qquad (2.41c)$$

$$\tau_1'^2 = \tau^2 |M'|^2 \qquad (2.41d)$$

$$\gamma' = \left(1 - \frac{z}{\tau^2 2\sqrt{2D}} \frac{\xi(\gamma^3 - 3\gamma\delta^2) + \eta(\delta^3 - 3\gamma^2\delta)}{(\gamma^2 + \delta^2)^3}\right), \qquad (2.41e)$$

and γ, δ, η, ξ, U', V', W' and Δ_2' are functions of $\bar{\omega}$, z and parameters appearing in dielectric function.

Case III:

Finally away from resonance (below resonance) i.e. $\omega/\omega_0 \ll 1$, there is only one propagatin polariton branch. In this limit the dispersion relation is approximated as

$$K_2(\omega) = q\frac{\omega}{\sqrt{D}} + \frac{i\omega^2\Gamma}{4q\omega_0^2\sqrt{D}} + \frac{\omega^3}{8q\omega_0^2\sqrt{D}} , \qquad (2.42)$$

where

$$q = \left[\frac{D\epsilon_\infty}{c^2}\left(1+\frac{4\pi\alpha_0}{\epsilon_\infty}\right) - \Gamma^2/4\omega_0^2\right]^{1/2}.$$

Again as in previous cases putting Equation (2.42) in (2.32) we obtain the amplitude of the polariton as:

$$f_2(z,t) = \frac{1}{\sqrt{M'}} \exp\left(-\left(\Delta_1' + i\Delta_2'\right)\right) A_2(\bar{\omega}), \qquad (2.43)$$

where,

$$M' = \left[\left(1+\frac{\Gamma z}{\tau^2}\frac{1}{2q\omega_0^2\sqrt{D}}\right) - i\left(\frac{3}{4}\frac{Z\bar{\omega}}{\tau^2 q\omega_0^2\sqrt{D}}\right)\right] \qquad (2.44a)$$

$$\Delta_1' = \frac{(t-\alpha')^2}{2\tau_2'^2} + \left(\frac{U'}{\gamma'} + V'\right) + W' \qquad (2.44b)$$

$$\tau_2'^2 = \gamma'^{-1}\tau_1'^2 \qquad (2.44c)$$

$$c_1'^2 = \tau^2 |M'|^2 \tag{2.44d}$$

$$\gamma' = \left[1 + \frac{z\Gamma}{2\tau^2 q \omega^2 \sqrt{D}} \right] , \tag{2.44e}$$

and α' , U' , V' , W', Δ_2' are functions of $\bar{\omega}$, z and parameters appearing in dielectric function $\epsilon\,(\vec{k},\omega)$.

2.3.3. <u>Pulse Propagation Close to ω_ℓ ($\omega \gtrsim \omega_\ell$)</u>

As noted earlier, in the case of non-local media, at each frequency there is more than one propagating mode. In the frequency region just above ω_ℓ there are two polariton modes which couple strongly to each other. Here we concentrate on this frequency region. We have been unable to obtain analytical results and thus we perform the study by numerical computation. We compute the power spectrum $P(z,\omega)$ where

$$\begin{aligned} P(z,\omega) &= 2\pi\,\tau^2 \,|\, f(z,\omega)|^2 \\ &\simeq 2\pi\tau^2 \left(|f_1(z,\omega)|^2 + |f_2(z,\omega)|^2\right) \\ &= 2\pi\tau^2 \Big[|A_1(\omega)|^2 \exp[-(\omega-\bar{\omega})^2\tau^2 - 2\,\mathrm{Im}\,K_1(\omega) z] \\ &\quad + |A_2(\omega)|^2 \exp[-(\omega-\bar{\omega})^2\tau^2 - 2\,\mathrm{Im}\,K_2(\omega) z] \Big] \end{aligned} \tag{2.45}$$

As an illustrative case, we consider CdS, and we used parameters: $\Gamma/2\omega_0 = 10^{-5}$, $\hbar\omega_0 = 2.55$ ev, $M^* = 0.9\ M_e$, $\epsilon_\infty = 8.0$ and $4\pi\alpha_0 = 0.0125$ respectively. We consider three cases, namely $\Gamma\tau \gg 1$, $\Gamma\tau = 1$ and $\Gamma\tau \ll 1$. For each case τ is taken as 0.1 psec, 13 psec and 10^{-9} sec respectively. The power spectrum, $P(z,\omega)$ is plotted for each case, against frequency for various crystal thicknesses, Z in Figs. 3, 4 and 5. Note that the dashed curves are the controus of $P(z,\omega)$ versus Z for various frequencies. Fig. 6 shows the power spectrum corresponding to $\Gamma\tau \ll 1$ very close to exciton resonance.

Next using Equation (2.29) the amplitude $|f(z,t)|$ is computed for various cases namely $\Gamma\tau \gg 1$, $\Gamma\tau = 1$ and $\Gamma\tau \ll 1$ respectively. Log $|f_1(z,t)|$ and Log $|f_2(z,t)|$ are plotted in each case against time for various crystal thicknesses z in Figs. 7, 8 and 9 respectively. For all the numerical studies the central frequency of the Gaussian, $\bar{\omega}$ is taken slightly above ω_ℓ i.e. $\bar{\omega} = \omega_0(1 + 10^{-3})$, so that the effect of both polariton branches UP and LP can be incorporated.

We also studied the pulse shape for the various cases mentioned above. For fixed value of crystal thickness, namely $z = 1\mu$, $|f(z,t)|$ is computed for Upper and Lower polaritons as function of time, as we sweep the laser frequency, $\bar{\omega}$ across the resonance. Figs. 10, 12 and 14 demostrate plots for various cases as we sweep laser frequency $\bar{\omega}$ from $\omega_0(1-10^{-2})$ to $\omega_0(1+10^{-2})$. Figs. 11, 13 and 15 show the corresponding plots for $\bar{\omega}$ very close to resonance, ranging from $\omega_0(1-10^{-3})$ to $\omega_0(1+10^{-3})$. A finer scale is used in those Figures. We shall discuss the Figs. 3-15 in more detail in Section 2.4.

2.4 Discussion

In this Chapter we studied the propagation of electromagnetic pulses in spatially dispersive media. In Section (2.2) energy transport in spatially dispersive media was studied. Various contributions to energy velocity, $V_E(UP)$ and $V_E(LP)$ below and above resonance are plotted in Fig. 1 for the material parameters of GaAs. On Fig. 1 the group velocity, V_G corresponding to Upper and Lower polariton branches is also plotted against reduced frequency. On the higher frequency side, $V_{E(UP)} \leq V_{G(UP)}$. On the lower requency side, $V_{E(LP)} \leq V_{G(LP)}$. Exactly at resonance, $V_E \sim 0.1\, V_G$. In the resonance region, the signal velocity, $V_S < V_G$. Fig. 2 shows the plot of C/V_S versus reduced frequency in the pseudo-gap region, $\omega_0 < \omega < \omega_\ell$. The signal velocity, V_S follows very close to group velocity, V_G in the pseudo-gap region. Analysis of the signal velocity in the spatially dispersive medium was given by Frankel and Birman. For large z they showed

$$\frac{c}{V_s} = \frac{c\chi}{(\Gamma/2)} \quad , \tag{2.45}$$

where χ is the imaginary part of the wave vector and Γ is the damping constant of the medium.

In Section (2.3) we presented results of an analysis of Gaussian pulse propagation in spatially dispersive media analogous to that of Garrett and McCumber in the local case. There we considered frequency regimes far from resonance: Cases I, and III, $(\omega/\omega_0) \gg 1$ and $\ll 1$ respectively: and also in the pseudo-gap region, $\omega_0 < \omega < \omega_\ell$. The pulse is characterized by $\Gamma\tau \gg 1$. Equations (2.34), (2.36), (2.40) and (2.43) show that in the various frequency regimes mentioned above, the Gaussian pulse propagates substantially as Gaussian both on Upper and Lower polariton branches. In general there will be a shift in the peak of the packet, and a change in full width at half maximum in general, as given in those equations.

Close to exciton resonance but above the longitudinal mode frequency (ω_ℓ), our numerical study (for CdS parameters) reveals the following (as given in Section (2.3)):

For $\Gamma\tau \gg 1$ we obtain a symmetric power spectrum as shown in Fig. 3. As z is changed, the shape of the power spectrum remains unchanged. A slight asymmetry in the power spectrum is observed for the case $\Gamma\tau = 1$ as shown in Fig. 4. For the case $\Gamma\tau \ll 1$, as shown in Fig. 5, as z is increased more and more asymmetry in the power spectrum can be noted. In this case an interesting "sharp cross over" is observed at ω_ℓ in the power spectrum from the Lower to Upper polariton. This is shown in Fig. 6.

For $\Gamma\tau \gg 1$, the amplitude plots $|f_1(z,t)|$ and $|f_2(z,t)|$ designated as UP and LP respectively, show very little variation in the shape of the packet as crystal thickness z is changed from $z = 10^{-6}$ cm to 10^{-4} cm. (See Fig. 7). This case corresponds to many oscillations in time as the packet is wide in time domain.

For $\Gamma\tau = 1$, the amplitude plots of $|f_1(z,t)|$ and $|f_2(z,t)|$ show very little variation in the shape of the packet as we increase the crystal thickness z, as shown in Fig. 8. This case corresponds to relatively few oscillations in time.

In the limit $\Gamma\tau \ll 1$, from the qualitative point of view, the packet gets more distorted as it propagates in the crystal as shown in Fig. 9. Notice here $|f_1(z,t)|$ or UP becomes broadened and distorted as it moves in the crystal i.e. as z is increased. Also $|f_2(z,t)|$ or LP even for $z = 10^{-6}$cm does not appear Gaussian: it is rather flat and as it moves in the crystal more oscillations develop.

In the limit $\Gamma\tau \gg 1$, the pulse shape remains Gaussian as we sweep across the resonance; this is shown in Figs. 10 and 11. The pulse width remains substantially constant on both Upper and Lower polariton branches. Similar behavior is noted for $\Gamma\tau = 1$, as shown in Figs. 12 and 13. In this case very little variation in pulse width is noted as we sweep across resonance (for example a 26 p. sec. pulse shows maximum variation of 4 p. sec.). The $\Gamma\tau \ll 1$ limit corresponds to considerable pulse distortion as demonstrated in Figs. 14 and 15. There is a great deal of structure in the Lower polariton branch above resonance as seen in these Figures.

As a physical description we can understand that for the case $\Gamma\tau \gg 1$, the pulse shows minimum distortion because it is wide in time and as it propagates in the medium it imparts energy to the medium and the dipoles of the medium are excited. As the dipoles relax back they still see some part of the pulse. Hence in turn, the pulse gets back the part of the energy, thus very little distortion in the pusle is created. On the other hand, for $\Gamma\tau \ll 1$ case pulse is much narrower in time, relative to $\Gamma\tau \gg 1$ case and as it propagates through the medium it imparts energy to the dipoles and when these dipoles relax back they do not see any part of the pulse. The pulse has already moved in time. Thus pulse looses energy at each moment as it moves in the medium and more distortions occur for $\Gamma\tau \ll 1$ case.

Our analysis shows that the velocity of motion of the peak of the pulse follows the group velocity as shown in Fig. 16 in the resonance region in the cases $\Gamma\tau \gg 1$ and $\Gamma\tau = 1$. This can be inferred from the shift in the cneter of the packet as $\bar{\omega}$ sweeps across resonance. In these cases the pulse shape remains substantially Gaussian, with very little variation in pulse width.

We can compare our results in part with some recent experiments. It seems that our numerical results for Gaussian pulse propagation in excitonic polariton (spatially dispersive) for media laser frequencies in the resonance region agree with the time-of-flight experiments (Ref. 14-17). These experiments seem to correspond to the case $\Gamma\tau \approx 1$; and they find that the pulse shape remains substantially Gaussian, and that the peak of the pulse travels at the "classical" group velocity computed from $V_{G_j} = (d\omega_j/dK_r)$ in each branch. Here we say "seem to" because experimental conditions are not always adequately given. Also in agreement is the "cross-over" of power propagating from Lower to Upper branch polariton as the laser sweeps through resonance frequency from below.

CHAPTER III

WAVE PROPAGATION IN BOUNDED GYROTROPIC MEDIUM NEAR RESONANCE

3.1 Introduction

In recent years the optics of nongyrotropic material media has attracted a great deal of attention. This is due in part to observations[2] that spatial dispersion (wave-vector-dependent terms in the dielectric function) leads to the appearance of new modes in the crystal. In a finite crystal these elctromagnetic modes can interfere with each other. The problem of reflection and refraction at the interface between such media and vacuum is also complicated due to the constitutive relation for such media being of an integral type. Hence the electric field inside the medium obeys an integro-differential equation. Several approximations are usually made to solve the optics of such media; common to all of them is the assertion that the wave vector $\vec{K}$ is small i.e. the wavelength is larger than lattice spacing. In nongyrotropic terms of $\vec{K}$ and keeps only the second order term in $\vec{K}$, the first order vanishing due to symmetry. In gyrotropic crystals i.e. in crystals which lack centre of inversion symmetry, or horizontal reflection symmetry plane, $\vec{K}$ -linear terms are included in the expansion of the dielectric function. This then leads to optical activity. One of the striking features of the optics of such media in connection with the appearance of new modes is the possibility of having three waves in such media near optical resonance. Although the three wave effect has been predicted by Ginzburg in 1958,[31] such an effect was earlier discussed by Gibbs in 1882.[32] Gibbs dismissed the third mode as he pointed out that the refractive index of the third wave is too large to be considered in the framework of macroscopic electromagnetic theory. Ginzburg, however, pointed out that near some exciton resonance frequency the refractive indices of all three waves may attain such values that they can be treated in the domain of Maxwell's theory. He suggested one should start with the inverse dielectric function and keep $\vec{K}$ -linear terms in the expansion of the inverse dielectric function. He obtained a dispersion relation which has three solutions at any given frequency.

In the case of gyrotropic media several forms of additional boundary conditions have been proposed, in both microscopic and macroscopic appraches.[33] In order to treat the problem of reflection and refraction on a gyrotropic half space, Ginzburg and Agranovich[34] keep quadratic terms in K in addition to the K -linear terms in the expansion of the (wave vector dependent) inverse dielectric function. Such a procedure not only increases the number of additional waves but also raises the question of convergence.[35]

In this Chapter we develop a macroscopic theory of gyrotropy keeping only K -linear terms in the expansion of the inverse dielectric function.[22] In Section (3.2) we start with the molecular optics approach[11(b)] and obtain the inverse dielectric function for a gyrotropic medium, which was proposed phenomenologically by Ginzburg.[36] In Section (4.3) we obtain the constitutive relation and the additional boundary condition (ABC). Section (4.4) deals with the structure of electromagnetic fields inside the gyrotropic medium. In Section (4.5) we solve the problem of reflection and refraction from optically active half space. Finally Section (4.6) is devoted to a brief discussion.

3.2 Basic Equations of Molecular Optics

We start from the basic equations of molecular optics as discussed in Born and Wolf,[11(b)]

$$\vec{E}_L(\vec{r}_i) = \vec{E}^{(i)}(r_i) + \sum_{j \neq i} \nabla_i \times (\nabla_i \times (\vec{p}(\vec{r}_j)\, G(R_{ij})), \tag{3.1}$$

where

$$G(R_{ij}) = \frac{e^{i k_0 R_{ij}}}{R_{ij}}, \tag{3.2}$$

$$k_0 = \omega/c, \quad R_{ij} = |\vec{r}_i - \vec{r}_j|, \tag{3.3}$$

and $\vec{E}^{(i)}$ is the external electric field incident on the medium, $\vec{p}_j(\vec{r}_j)$ is the dipole moment of the j th dipole and $\vec{E}_L(\vec{r}_i)$ is the local field at the i th molecule. We have considered for simplicty a non-magnetic medium. The magnetic field is given by the expression

$$\vec{H}_L(\vec{r}_i) = \vec{H}^{(i)}(\vec{r}_i) + \sum_{j \neq i} (-i K_0) \nabla_i \times \left(\vec{p}(\vec{r}_j) G(R_{ij}) \right), \quad (3.4)$$

where $\vec{H}^{(i)}$ is the incident magnetic field and $\vec{H}_L(\vec{r}_i)$ is the local magnetic field at the i^{th} dipole.

A recent important review of the molecular optics approach from a modern point of view was given by Van Kranendonk and Sipe.[37] They were able to relate gyrotropy to a tight binding quantum theory of molecular structure and excitations.

We assume here that gyrotropy is due to the lack of inversion symmetry in the individual constituents (molecules) comprising the medium. We take them to be isotropic. As is well known the dipole moment then depends not only on the electric field but also on its spatial derivatives. In such a medium the dipole moment is therefore related to the local electric field in the following manner

$$\vec{p}(r_j) = \alpha(\omega)\, \vec{E}_L(\vec{r}_j) + \beta(\omega)\, \vec{\nabla}_j \times \vec{E}_L(r_j), \quad (3.5)$$

where $\alpha(\omega)$ is essentially the mean polarizability of the molecule and $\beta(\omega)$, related to the gyration vector, depends on the properties of the medium. In the usual formulation of molecular optics the second term on the right hand side Equation (3.5) is not present. In the long wavelength regime in which we are interested we may to a good approximation, treat the dipole distributions to be continuous and make the transition from discrete to continuous variables in Equations (3.1), (3.4) and (3.5).

In the continuous distribution limit Equations (3.1), (3.2) and (3.5) become

$$\vec{E}_L(\vec{r}) = \vec{E}^{(i)}(\vec{r}) + \int_{\sigma}^{\Sigma} \vec{\nabla} \times (\vec{\nabla} \times \vec{P}(r')G(R))\,d^3r', \tag{3.6}$$

$$\vec{H}_L(\vec{r}) = \vec{H}^{(i)}(\vec{r}) - iK_0 \int_{\sigma}^{\Sigma} \vec{\nabla} \times (\vec{P}(r')\,G(R))\,d^3r', \tag{3.7}$$

and

$$\vec{P}(\vec{r}) = N\alpha(\omega)\vec{E}_L(\vec{r},\omega) + N\beta(\omega)\vec{\nabla} \times \vec{E}_L(\vec{r}), \tag{3.8}$$

where

$$G(R) = \frac{e^{iKR}}{R} \quad ; \quad R = |\vec{r} - \vec{r}\,'|, \tag{3.9}$$

and N is the number density of dipoles which we assume to be uniform. In Equations (3.6) and (3.7) the integration is taken over the interior of the crystal, V is the volume occupied by the crystal, Σ is the surface bounding the volume and σ is the surface surrounding the small volume excluded at the point, r at which the fields are being calculated. Equations (3.6) and (3.7) are identical in form with the usual molecular optics equations and therefore we obtain the usual relationship between the macroscopic fields and the local fields which are

$$\vec{E}(\vec{r},\omega) = \vec{E}_L(\vec{r},\omega) - \frac{4\pi}{3}\vec{P}(\vec{r},\omega) \quad , \tag{3.10}$$

$$\vec{H}(\vec{r},\omega) = \vec{H}_L(\vec{r},\omega) \tag{3.11}$$

where $\vec{E}$ and $\vec{H}$ are the macroscopic Maxwell electric and magnetic field respectively. If we now eliminate $E_L(\vec{r},\omega)$ between Equations (3.8) and (3.10) we find the following constitutive relation between $\vec{P}$ and $\vec{E}$

$$\vec{P}(\vec{r},\omega) = \alpha_0(\omega)\vec{E}(\vec{r},\omega) + \beta_0(\omega)\vec{\nabla}\times\vec{E}(\vec{r},\omega) + \frac{4\pi}{3}\beta_0(\omega)\vec{\nabla}\times P, \tag{3.12}$$

where

$$\alpha_0(\omega) = \frac{N\alpha(\omega)}{\left(1 - \frac{4\pi}{3}N\alpha(\omega)\right)} \quad , \tag{3.13}$$

$$\beta_0(\omega) = \frac{N\beta(\omega)}{\left(1 - \frac{4\pi}{3}N\alpha(\omega)\right)} \quad . \tag{3.14}$$

Equation (3.12) may be rewritten in many different ways. By introducing the dielectric induction vector in the usual way we get the relation

$$D(\vec{r},\omega) = \epsilon_0(\omega)\,\vec{E}(\vec{r},\omega) + \frac{4\pi}{3}\beta_0(\omega)\,\vec{\nabla}\times\vec{D}(\vec{r},\omega)$$

$$+ \frac{8\pi}{3}\beta_0(\omega)\,\vec{\nabla}\times\vec{E}(\vec{r},\omega), \tag{3.15}$$

where

$$\vec{D}(\vec{r},\omega) = \epsilon_{\infty}\vec{E}(\vec{r},\omega) + 4\pi\vec{P}(\vec{r},\omega) \tag{3.16a}$$

$$\epsilon_0(\omega) = 1 + 4\pi\alpha_0(\omega) \quad . \tag{3.16b}$$

Near resonance the dielectric displacement vector $\vec{D}$ is large compared to the electric field $\vec{E}$. Therefore if one neglects the third term on the r.h.s of Equation (3.15) compared to the second term, one obtains the equation

$$\vec{D}(\vec{r},\omega) \cong \epsilon_0(\omega)\vec{E}(\vec{r},\omega) + \frac{4\pi}{3}\beta_0(\omega)\vec{\nabla}\times\vec{D}(\vec{r},\omega) . \tag{3.17}$$

This implies the operator equation

$$\epsilon^{-1}(\vec{r},\omega) = \epsilon_0^{-1}(\omega)\,\vec{I} + \epsilon_1^{-1}(\omega)\,\vec{\nabla}\times \quad , \tag{3.17a}$$

where

$$\epsilon_1^{-1}(\omega) = \frac{4\pi}{3}\frac{\beta_0(\omega)}{\epsilon_0(\omega)} \quad .$$

A constitutive relation of type Equation (3.17) has been proposed phenomenologically by Ginzburg.[36]

3.3 The Constitutive Relation and Additional Boundary Condition

Here we consider an isotropic, non-magnetic and linear gyrotropic medium. Equation (3.17) give us

$$\vec{E}(\vec{r},\omega) = \epsilon_0^{-1}(\omega)\, \vec{D}(\vec{r},\omega) + \epsilon_1^{-1}(\omega)\, \vec{\nabla}\times\vec{D}(\vec{r},\omega). \tag{3.18}$$

We now relate the dielectric displacement and polarization field in the usual manner as described by Equation (3.16a) It can be shown from Equations (3.18) and (3.16a) that

$$\vec{P}(\vec{r},\omega) + \frac{4\pi\gamma(\omega)}{\epsilon_\infty}\vec{\nabla}\times\vec{P}(\vec{r},\omega) = \alpha(\omega)\vec{E}(\vec{r},\omega) - \gamma(\omega)\vec{\nabla}\times\vec{E}(\vec{r},\omega), \tag{3.19}$$

where

$$4\pi\alpha(\omega) = \epsilon_0(\omega) - \epsilon_\infty \tag{3.20}$$

$$4\pi\gamma(\omega) = \epsilon_0(\omega)\,\epsilon_1^{-1}(\omega)\,\epsilon_\infty \quad . \tag{3.21}$$

Using Maxwell's eqations, we can write Equation (3.19) as

$$\vec{P}(\vec{r},\omega) + \frac{4\pi\gamma(\omega)}{\epsilon_\infty}\vec{\nabla}\times\vec{P}(\vec{r},\omega) = \alpha(\omega)\vec{E}(\vec{r},\omega) - iK_0\gamma(\omega)\vec{H}(\vec{r},\omega). \tag{3.22}$$

In order to simplify the analysis we shall from now on deal with a one dimensional situation. That is, consider propagation in the z direction. We replace $\vec{\nabla}$ by $\hat{z}\frac{\partial}{\partial z}$ in Equation (3.22). We then obtain the following system of differential euqations

$$P_x(z,\omega) - \frac{4\pi}{\epsilon_\infty}\gamma(\omega)\frac{\partial P_y}{\partial z}(z,\omega) = \alpha(\omega)E_x(z,\omega) - iK_0\gamma(\omega)B_x(z,\omega) \quad (3.23)$$

$$P_y(z,\omega) - \frac{4\pi}{\epsilon_\infty}\gamma(\omega)\frac{\partial P_x}{\partial z}(z,\omega) = \alpha(\omega)E_y(z,\omega) - iK_0\gamma(\omega)B_y(z,\omega). \quad (3.24)$$

Equations (3.23) and (3.24) are differential constitutive relations for a linear gyrotropic medium near resonance. Using Equations (3.230 and (3.24) it can be shown that

$$\Pi(z,\omega) + \frac{4\pi i\gamma(\omega)}{\epsilon_\infty}\frac{\partial}{\partial z}\Pi(z,\omega)$$

$$= \alpha(\omega)\left(E_y(z,\omega) - iE_x(z,\omega)\right) - iK_0\gamma(\omega)\left(B_y(z,\omega) - iB_x(z,\omega)\right), \quad (3.25)$$

where we have

$$\Pi(z,\omega) = P_y(z,\omega) - iP_x(z,\omega) \quad (3.26)$$

We now follow the approach described by Gakhov.[38] By introducing a new function, namely $\Pi(z,\omega)$ we have the boundary value problem for the system of differential equations (3.23) and (3.24). Considering a pill-box construction at $z = 0$ we can write down Equation (3.25) as,

$$\gamma(\omega)\,\Pi\,(0_-,\omega) - \gamma(\omega)\,\Pi\,(0_+,\omega) = 0 , \tag{3.27}$$

where 0_- and 0_+ are points outside and inside the surface. Since $\Pi(0_-,\omega) = 0$, we obtain the additional boundary condition as

$$P_y(0^+,\omega) - i\,P_x(0^+,\omega) = 0 . \tag{3.28}$$

Thus Equation (3.28) gives us an additional boundary condition derived from Maxwell's equations and constitutive relations. It is to be noted that the boundary condition (3.28) is a particular case of the effective boundary condition proposed by Agranovich and Ginzburg.[39]

3.4 Structure of Electromagnetic Fields Inside the Gyrotropic Half Space

We now investigate the nature of electromagnetic fields inside a gyrotropic half space whose constitutive relation is given in Section (3.3) and we determine how the boundary conditions (3.28) affect the mode structure. From Maxwell's equations we obtain the folowing equation after eliminating the magnetic field

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E}(\vec{r},\omega) = K_0^2\,D(\vec{r},\omega) . \tag{3.29}$$

For transverse fields we have

$$\vec{\nabla}\cdot\vec{E}(\vec{r},\omega) = \vec{\nabla}\cdot\vec{D}(\vec{r},\omega) = 0 \quad . \tag{3.30}$$

Thus we can write down

$$\nabla^2\begin{pmatrix}\vec{D}(\vec{r},\omega)\\ \vec{E}(\vec{r},\omega)\end{pmatrix} + \frac{4\pi\gamma(\omega)}{\epsilon_\infty}\nabla^2\begin{pmatrix}\vec{\nabla}\times\vec{D}(\vec{r},\omega)\\ \vec{\nabla}\times\vec{E}(\vec{r},\omega)\end{pmatrix} + K_0^2\epsilon_0\begin{pmatrix}\vec{D}(\vec{r},\omega)\\ \vec{E}(\vec{r},\omega)\end{pmatrix} = 0 \quad . \tag{3.31}$$

Equation (3.31) represents a system of partial differential equations for the vector components of the fields. By using a vector identity and straightforward algebra we obtain the following higher order uncoupled equation for $\vec{E}$ and $\vec{D}$.

$$(\nabla^2 + K_0^2\epsilon_0)^2\begin{pmatrix}\vec{D}\\ \vec{E}\end{pmatrix} + \left(\frac{4\pi\gamma}{\epsilon_\infty}\right)^2\nabla^6\begin{pmatrix}\vec{D}\\ \vec{E}\end{pmatrix} = 0 \quad . \tag{3.32}$$

We may rewrite Equation (3.32) as

$$\prod_{j=1}^{3}(\nabla^2 + K_j^2)\,\vec{E}(\vec{r},\omega) = 0 \,. \tag{3.33}$$

The dispersion relation obtained is

$$(K_0^2\epsilon_0 - K^2)^2 - K^6\left(\frac{4\pi\gamma}{\epsilon_\infty}\right)^2 = 0 \,, \tag{3.34}$$

or we can write as

$$K^6 - \left(\frac{\epsilon_\infty}{4\pi\gamma}\right)^2 K^4 + 2\left(\frac{\epsilon_\infty}{4\pi\gamma}\right)^2 K_0^2\epsilon_0 K^2 - \left(\frac{\epsilon_\infty}{4\pi\gamma}\right)^2 K_0^2\epsilon_0^2 = 0 \,. \tag{3.35}$$

The dispersion relation (3.35) is a sixth order in K and third order in K^2. In Section (3.6) we shall discuss the dispersion curves obtained from Equation (3.35). Equation (3.31) leads to the following polarization condition

$$\vec{\nabla} \times \vec{E}_j(\vec{r},\omega) = \frac{(K_0^2\epsilon_0 - K_j^2)}{(4\pi\gamma/\epsilon_\infty)K_j^2}\ \vec{E}_i(\vec{r},\omega) . \tag{3.36}$$

Using Equation (3.34) in (3.36) we find

$$\vec{\nabla} \times \vec{E}_j(\vec{r},\omega) = \pm K_j\ \vec{E}_j(\vec{r},\omega) \tag{3.37}$$

In order to study the polarization property of the electric field explicitly we choose the special case of normal incidence at a half space for which the problem is one dimensional. Further following the work of Ref. (40) we write solution as

$$\vec{E}(\vec{r},\omega) = \sum_{j=1}^{3} E_j(\vec{r},\omega) \tag{3.38}$$

where E_j satisfy the Helmholtz equations

$$(\nabla^2 + K_j^2)\ \vec{E}_j(\vec{r},\omega) = 0 . \tag{3.39}$$

Since the solutions of Equation (3.39) are simple plane waves Equation (3.38) becomes

$$\vec{E}(z,\omega) = \sum_{j=1}^{3} E_j(z,\omega) = \sum_{j=1}^{3} \hat{E}_j \, e^{i K_j z} \quad . \tag{3.40}$$

On substituting Equation (3.40) into Equation (3.37) we obtain

$$i \, \hat{z} \times \hat{E}_j = \pm \hat{E}_j \quad . \tag{3.41}$$

Thus the polarization assignments are

$$\hat{E}_j = (\hat{x} - i\,\hat{y})\, L_j \quad , \quad j = 1, 2 \tag{3.42}$$

$$\hat{E}_3 = (\hat{x} + i\,\hat{y})\, R \tag{3.43}$$

where L_j and R are amplitudes of left and right circularly polarized waves.

In other words the electric field now consists of two left circularly polarized waves whose amplitudes are coupled and one right circularly polarized wave, each having different phase velocities. From the Maxwell equations it is now easy to show that

$$\vec{P}(z,\omega) = \sum_{j=1}^{3} \hat{P}_j \, e^{i K_j z} \tag{3.44}$$

where

$$\hat{P}_j = (n_j^2 - 1)\, \hat{E}_j \tag{3.45}$$

and

$$n_j = K_j / K_0 \tag{3.46}$$

is the refractive index associated with the j th wave. On using Equations (3.38), (3.44) and (3.45) we find from Equation (3.28) that

$$\sum_{j=1}^{2} (n_j^2 - 1) L_j = 0 \quad , \tag{3.47}$$

which implies that the amplitudes of the two left circularly polarized waves are related by the boundary condition (3.28).

3.5 Refraction and Reflection Problem

We treat the case when a plane wave is incident from the vacuum side onto the gyrotropic half space $(z \geq 0)$. We denote the fields in the vacuum i.e. for $z < 0$ as follows

$$\vec{E}_1 = \vec{E}_{INC} + \vec{E}_{Ref}$$

$$= \hat{x} E_0 e^{i K_0 z} + (\hat{x} E_{Rx} + \hat{y} E_{Ry}) e^{-i K_0 z} \tag{3.48}$$

$$\vec{H}_1 = \hat{y} E_0 e^{-i K_0 z} + (\hat{x} E_{Ry} - \hat{y} E_{Rx}) e^{-i K_0 z} . \tag{3.49}$$

Fields inside the gyrotropic half space i.e. for $z \geq 0$ are denoted

$$\vec{E}_{IN} = \sum_{j=1}^{2} (\hat{x} - i\hat{y}) L_j e^{iK_0 n_{\ell_j} z} + (\hat{x} + i\hat{y}) R e^{iK_0 n_r z} \tag{3.50}$$

$$\vec{H}_{IN} = \sum_{j=1}^{2} (\hat{y} + i\hat{x}) n_{\ell_j} L_j e^{iK_0 n_{\ell_j} z} + (\hat{y} - i\hat{x}) n_r R e^{iK_0 n_r z}, \tag{3.51}$$

where $n_{\ell_j} = n_j$ and $n_r = n_3$. The subscripts ℓ and r refer to the left and right circular polarization respectively. The boundary condition (3.28) becomes

$$\sum_{j=1}^{2} (n_{\ell_j}^2 - 1) L_j = 0 . \tag{3.52}$$

In order to obtain the amplitudes of the reflected and refracted fields we use the continuity of $\hat{z} \times \vec{E}$ and $\hat{z} \times \vec{H}$ and obtain the following set of equations. Continuity of $\hat{z} \times \vec{E}$ at $z = 0$ gives

$$E_0 + E_{R_x} = \sum_{j=1}^{2} L_j + R \tag{3.53}$$

and

$$E_{R_y} = -i \left(\sum_{j=1}^{2} L_j - R \right) . \tag{3.54}$$

Continuity of $\hat{z} \times \vec{H}$ at $z = 0$ gives

$$E_0 - E_{R_x} = \sum_{j=1}^{2} n_{\ell_j} L_j + n_r R \tag{3.55}$$

and

$$E_{Ry} = i\left[\sum_{j=1}^{2} n_{\ell_j} L_j - n_r R\right] . \tag{3.56}$$

We have five unknowns namely E_{Rx}, E_{Ry}, L_1, L_2, and R, and we have five sets of homogeneous Equations (3.52) - (3.56). Thus we can solve for the unknowns and obtain

$$R = \frac{E_0}{(n_r+1)} \tag{3.57}$$

$$L_1 = \left(\frac{1-n_{\ell_2}}{1+n_{\ell_1}}\right)\frac{E_0}{(n_{\ell_1}-n_{\ell_2})} \tag{3.58}$$

$$L_2 = \left(\frac{1-n_{\ell_1}}{1+n_{\ell_2}}\right)\frac{E_0}{(n_{\ell_2}-n_{\ell_1})} \tag{3.59}$$

$$E_{R_x} = \frac{1}{2}\left[\left(\frac{1-n_{\ell_1}}{1+n_{\ell_1}}\right)\left(\frac{n_{\ell_2}-1}{n_{\ell_2}+1}\right) + \left(\frac{1-n_r}{1+n_r}\right)\right] E_0 \tag{3.60}$$

$$E_{R_y} = -i\left[\left(\frac{n_{\ell_1}+n_{\ell_2}}{1+n_{\ell_2}}\right)\left(\frac{1}{1+n_{\ell_1}}\right) - \frac{1}{(1+n_r)}\right] E_0 . \tag{3.61}$$

The refectivity $R(\omega)$ is defined as

$$R(\omega) = \sqrt{(|E_{Rx}|^2 + |E_{Ry}|^2)/E_0^2} . \tag{3.62}$$

Thus we obtain

$$R(\omega) = \frac{1+n_r^2}{(1+n_r)^2} - \frac{2(n_{\ell_1}+n_{\ell_2})(1+n_{\ell_1}n_{\ell_2})}{(1+n_{\ell_1})^2(1+n_{\ell_2})^2} . \tag{3.63}$$

3.6 Discussion

We have numerically calculated dispersion curves in order to illustrate the type and magnitude of the effects which can arise near resonance is an excitonic-polariton insulator due to gyrotropy. From these dispersion curves we calculated reflectivity as a function of frequency.

In order to avoid tensorial complications in our illustration, we consider the isotropic case as discussed in Sections (3.3) and (3.4). We write the inverse dielectric function as

$$\epsilon_{ij}^{-1}(K,\omega) = \epsilon_0^{-1}(\omega)\,\delta_{ij} + i\,\epsilon_1^{-1}(\omega)\;\varepsilon_{ij\ell}K_\ell \tag{3.64}$$

$$\epsilon_0(\omega) = \epsilon_\infty - \frac{\alpha_0}{(S+i\delta)} \tag{3.65a}$$

$$\epsilon_1(\omega) = -\frac{\alpha_1}{(S+i\delta)}\,, \qquad \alpha_1 = (\alpha_0^2/\alpha_{\parallel}) \tag{3.65b}$$

$$S = (\omega-\omega_0)/\omega_0\,, \quad \delta = \Gamma/2\omega_0\,, \tag{3.65c}$$

Where ϵ_∞ is the background dielectric constant, α_0 some effective oscillator strength coupling, and $\alpha_{\parallel}$ a gyrotropic parameter.

As a "model" substance we considered an insulator with parameter values like those of CdS, although we do not intend to imply this theory will apply CdS. The B-exciton in CdS has been desribed in a K -linear approximation as gyrotropic.[41] For the gyrotropy parameter $\alpha_{\parallel}$ we chose 0.01 or 10^{-7} cm.

In Fig. 17 the dispersion curves are shown for different values of the damping parameter δ . The real part of K is plotted near resonance. Recall there are three propagating modes in the medium: two are left circularly polarized, and coupled, the third is right circularly polarized. One of the coupled modes has negative phase velocity. We can understand physically the concept of negative phase velocity as the mode propagating with positive phase but in $-z$ direction. It is coupled with a mode with positive phase velocity and propagating in $+z$ direction. In Fig. 18 the imaginary part of K is plotted near resonance.

In Fig. 19 we plot the modulus of reflectivity as a function of frequency near resonance for different damping parameters. A noteworthy feature of these curves is the rather pronounced structure with local maxima and dips. It is not simple to correlate such structure with the dispersion curves (since the reflectivity is the resultant of all coupled modes) but one can note that peaks in reflectivity correspond to regions in frequency where group velocity on one or another branch is low, and/or attenuation ($\mathrm{Im}\, K$) is large. Notice for example that for $\delta = 5 \times 10^{-4}$ we observe three peaks in the reflection spectrum. As the damping is reduced, the number of peaks in the spectrum is reduced. For $\delta = 10^{-4}$ we observe only one peak. This peak shifts towards frequencies close to resonance as we decrease the damping from $\delta = 5\times10^{-4}$ to $\delta = 10^{-4}$. In Fig. 20 a plot of phase angle as function of frequency for different damping parameters is given, corresponding to the curves in Fig. 19.

In conclusion, we have studied the electrodynamics of bounded gyrotropic medium, near resonance from phenomenological point of view. Additional boundary conditions are obtained self-consistently. The problem of reflection from optically active half space is solved and numerical results are discussed.

CHAPTER IV

SURFACE WAVES IN BOUNDED GYROTROPIC MEDIA

4.1 Introduction

The characteristic of a surface electromagnetic mode is that it decays exponentially as we move away from the surface and it propagates along the surface with wave vector, u , where u is the component parallel to the surface of the full wave vector $\vec{K}$.

The electric field, $\vec{E}$ associated with such a mode is given by,

$$\vec{E}(\vec{r},\omega) = \vec{E}_0 e^{iux - \beta|z|} \tag{4.1}$$

Here the surface is taken at $z=0$ and $z>0$ corresponds to the semi infinite medium and $z<0$ corresponds to vacuum.

We shall recall in Section (4.2) that the dispersion relation for surface modes in the absence of spatial dispersion can be written as

$$u^2 = \frac{\omega^2}{c^2} \frac{\epsilon(\omega)}{(\epsilon(\omega)+1)} \quad , \tag{4.2}$$

with the condition $\epsilon(\omega)<-1$. Here $\epsilon(\omega)$ is defined by Equation (2.6). Consider for example $\Gamma=0$ case. We find that $\epsilon(\omega)<-1$ in the frequency region $\omega_0 < \omega < \omega_\ell$, which corresponds to the stopgap region for bulk propagation.

Surface plasmons have been observed experimentally from metal grating surfaces by Teng and Stern,[42] Powell and

Swan[43] detected these excitations by electron scattering from thin metal foils. Barker[44] was the first to observe surface plasmons by direct optical coupling to surface excitons using Attenuated Total Reflection (ATR) type geometry. Surface polaritons in a frequency dispersive medium (GaP)have been observed by Marschall and Fischer.[45]

When spatial dispersion is taken into account the dispersion equation for surface polaritons becomes dependent on the ABC's.[1-10] A surface polariton dispersion relation has been obtained by Maradudin and Mills[7] and also by Agarwal[46] using a translationally invarient model for dielectric runction, in the "dielectric approximation". A non-translationally invarient model for the dielectric function has been used by Frankel and Birman[8] to investigate surface polaritons near exciton resonance.

Bishop and Maradudin[47] have studied linear wave vector effects on the optical properties of crystals. They have formulated an Attenuated Total Reflection (ATR) experiment for surface polaritons in α -quartz. Falge and Otto[48] have made an experimental study of surface polaritons on α -quartz by the ATR method, in the infra-red optical region.

In this Chapter we study the surface waves corresponding to antisymmetric K -linear terms in the dielectric function which give rise to left and right circularly polarized light waves in the medium. The plan of the Chapter is as follows.

In Section (4.2) we review some properties of surface modes in a frequency dispersive medium. In Section (4.3) we obtain the surface wave dispersion relation in a bounded gyrotropic medium. Section (4.4) deals with the formulation of a ATR model experiment.[24] Finally Section (4.5) is devoted to brief discussion of these results.

4.2 Surface Modes in the Frequency Dispersive Medium

Consider a frequency dispersive medium free of charges and currents and also non-magnetic. Maxwell's equation for such a medium are give by,

$$\vec{\nabla}\cdot\vec{D} = 0 \tag{4.3a}$$

$$\vec{\nabla}\times\vec{E} = -\frac{1}{c}\frac{\partial}{\partial t}\vec{H} \tag{4.3b}$$

$$\vec{\nabla}\times\vec{H} = \frac{1}{c}\frac{\partial}{\partial t}\vec{D} \tag{4.3c}$$

$$\vec{\nabla}\cdot\vec{H} = 0 \ . \tag{4.3d}$$

The dielectric displacement, $\vec{D}$ is related to the electric field, $\vec{E}$ by a non-local constitutive relation in time as

$$\vec{D}(t) = \int_{-\infty}^{t} \epsilon(t-t')E(t')dt' , \tag{4.4}$$

which gives us $\vec{D}(\omega) = \epsilon(\omega)\ \vec{E}(\omega)$. Taking the curl of Equation (4.3b),

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} = -\frac{\epsilon(\omega)}{c^2}\frac{\partial^2 E}{\partial t^2}\ , \tag{4.5}$$

and considering fields to be time harmonic we get

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E}(\vec{r},\omega) + \epsilon(\omega)\frac{\omega^2}{c^2}\ \vec{E}(\vec{r},\omega) = 0\ . \tag{4.6}$$

As we have described earlier, surface modes are designated by Equation (4.1). Substituting Equation (4.1) in Equation (4.6) we get

$$-(u^2 - \beta^2) + \epsilon(\omega)\frac{\omega^2}{c^2} = 0$$

or

$$\beta = \sqrt{u^2 - \epsilon(\omega)\,\omega^2/c^2}\ . \tag{4.7}$$

Next we apply the Maxwell continuity conditions to the fields at the surface $z = 0$. Let us denote $\vec{E}^{(V)}$ and $\vec{E}^{(M)}$ the fields in the vacuum and the medium respectively. Continuity of the normal components of $\vec{D}$ and tangential components of $\vec{E}$ gives

$$E_x^{(V)} = \epsilon(\omega)\ E_z^{(M)} \tag{4.8}$$

$$E_x^{(V)} = E_x^{(M)} \tag{4.9}$$

Putting Equations (4.8) and (4.9) in Equation (4.3a) we get

$$E_x^{(V)} = -\frac{\beta^{(V)}}{iU} E_z^{(V)} \tag{4.10}$$

$$E_x^{(M)} = \frac{\beta^{(M)}}{iU} E_z^{(M)} \tag{4.11}$$

From Equations (4.10) and (4.11)

$$\epsilon(\omega) = \frac{E_z^{(V)}}{E_z^{(M)}}$$

$$= -(\beta^{(M)}/\beta^{(V)}) . \tag{4.12}$$

Putting the value of β from Equation (4.7) in (4.12),

$$\epsilon(\omega) = -\frac{\sqrt{U^2 - \epsilon(\omega)\,\omega^2/c^2}}{\sqrt{U^2 - \omega^2/c^2}} \tag{4.13}$$

or

$$U^2 = \frac{\omega^2}{c^2} \frac{\epsilon(\omega)}{(\epsilon(\omega)+1)} , \tag{4.2}$$

we note from Equation (4.13) that surface modes must satisfy $\epsilon(\omega) < 0$.

4.3 Surface Waves in an Optically Active Half Space

In this Section we examine surface waves at the interface between an optically active half space and vacuum following the recent analysis of Pattanayak and Birman.[23] The dielectric function for an homogeneous optically active medium[21] can be written as

$$\epsilon_{ij}(\vec{K},\omega) = \epsilon_0(\omega)\,\delta_{ij} + i\,\epsilon_1(\omega)\,\varepsilon_{ij\ell}K_\ell\,, \tag{4.14}$$

where $\vec{K}$ is the wave vector, ω is the frequency of the electromagnetic field, δ_{ij} is the Kroneker delta function, and $\varepsilon_{ij\ell}$ is the levi-cevita tensor. $\epsilon_0(\omega)$ and $\epsilon_1(\omega)$ are functions of frequency.

Consider the problem of surface waves at the interface between an optically active half-space and vacuum with $z<0$ vacuum and $z>0$ the optically active half space containing the medium. We represent the electromagnetic fields in the vacuum region $(z<0)$ using the angular spectrum representation

$$\vec{E}^{(v)}(\vec{r},\omega) = \iint_{-\infty}^{\infty} \hat{\vec{E}}^{(v)}(u,v)\, e^{i(ux+vy-w_0 z)}\,du\,dv, \tag{4.15}$$

where

$$w_0 = (K_0^2 - u^2 - v^2)^{1/2} \qquad \text{if} \quad u^2+v^2 < K_0^2\,,$$

$$W_0 = i(u^2+v^2-K_0^2)^{1/2} \qquad \text{if} \quad u^2+v^2 > K_0^2, \tag{4.16}$$

and $\hat{\vec{E}}^{(v)}$ has x, y and z components $\hat{E}_x^{(v)}$, $\hat{E}_y^{(v)}$, and $\hat{E}_z^{(v)}$ respectively. Note $\vec{E}^{(v)}$ and $\hat{\vec{E}}^{(v)}$ represent $\vec{E}^{(v)}(r, \omega)$ and $\hat{\vec{E}}^{(v)}(u, v)$ respectively.

Since the electric field is transverse in the vacuum region, define a vector $\vec{\eta} \equiv (u, v, -W_0)$ so that

$$\vec{\eta} \cdot \hat{\vec{E}}^{(v)} = 0. \tag{4.17}$$

The magnetic field in the vacuum region is given by

$$\vec{H}^{(v)}(\vec{r}, \omega) = \iint_{-\infty}^{\infty} \hat{\vec{H}}^{(v)}(u, v)\, e^{i(ux+vy-W_0 z)}\, du\, dv, \tag{4.18}$$

where

$$\begin{aligned} \hat{\vec{H}}^{(v)}(u,v) = {} & \hat{x}\left(\frac{uv}{K_0 W_0}\hat{E}_x^{(v)}(u,v) + \frac{v^2+W_0^2}{K_0 W_0}\hat{E}_y^{(v)}(u,v)\right) \\ & - \hat{y}\left(\frac{W_0^2+u^2}{K_0 W_0}\hat{E}_x^{(v)}(u,v) + \frac{uv}{K_0 W_0}\hat{E}_y^{(v)}(u,v)\right) \\ & + \hat{z}\left(\frac{u^2+W_0^2}{K_0 W_0}\hat{E}_x^{(v)}(u,v) + \frac{uv}{K_0 W_0}\hat{E}_y^{(v)}(u,v)\right). \end{aligned} \tag{4.19}$$

The electric field in the medium[23] has been shown to be

$$\vec{E}(\vec{r},\omega) = \iint_{-\infty}^{\infty} E_+(u,v)\, e^{i(ux+vy+w_+z)}\,du\,dv$$

$$+\iint_{-\infty}^{\infty} E_-(u,v)\, e^{i(ux+vy+w_-z)}\,du\,dv, \quad (4.20)$$

where

$$w_+ \equiv (k_0^2 n_+^2 - u^2 - v^2)^{1/2} \qquad \mathrm{Im}\ w_+ > 0$$

$$w_- \equiv (k_0^2 n_-^2 - u^2 - v^2)^{1/2} \qquad \mathrm{Im}\ w_- > 0 \quad (4.21)$$

$$\hat{\vec{E}}_+(u,v) = \left[\hat{x} + i\frac{(k_0 n_+ w_+ + iuv)}{(k_0^2 n_+^2 - u^2)}\hat{y}\right.$$

$$\left. + \frac{(uw_+ + ik_0 n_+ v)}{(u^2 - k_0^2 n_+^2)}\hat{z}\right]\hat{E}_x(u,v) \quad (4.22a)$$

$$\hat{\vec{E}}_-(u,v) = \left[\hat{x} - \frac{i(k_0 n_- w_- - iuv)}{(k_0^2 n_-^2 - u^2)}\hat{y}\right.$$

$$\left. + \frac{(uw_- - ik_0 n_- v)}{(k_0^2 n_-^2 - u^2)}\hat{z}\right]\hat{E}_{-x}(u,v) \quad (4.22b)$$

and the magnetic field in the medium is obtained from Maxwell's equations as $\vec{H}(r,\omega) = \frac{1}{iK_0} \vec{\nabla} \times \vec{E}(r,\omega)$. The continuity of $\hat{z} \times \vec{E}$ and $\hat{z} \times \vec{H}$ at the interface $z=0$ gives four homogeneous algebraic equations for the four unknowns $\hat{E}_x^{(v)}$, $\hat{E}_y^{(v)}$, $\hat{E}_{+x}$, and $\hat{E}_{-x}$. In order that a non-trivial solution exist, the secular determinant must vanish. Choosing the case $\nu = 0$ and ignoring magnetization effects ($\chi_{mag} = 1$), we obtain from

$$n_+ (W_+ + W_0)(W_- + n_-^2 W_0) + n_- (W_- + W_0)(W_+ + n_+^2 W_0) = 0 . \quad (4.23)$$

Equation (4.23) is the dispersion equation for wave propagation; we notice that in order to have modes decaying away from the interface, we must impose

$$u^2 > K_0^2 \quad (4.24a)$$

with

$$u^2 > \mathrm{Re}(K_0^2 n_\pm^2) . \quad (4.24b)$$

We now expand W_+, W_- and W_0 in Equation (4.23) in powers of K_0^2/u^2. Keeping only leading terms we obtain

$$u^4 - \frac{1}{4}\left(\frac{(n_+ + n_-)^2}{(1 + n_+ n_-)} + (1 + n_+ n_-)\right) K_0^2 u^2 + \frac{1}{4} n_+ n_- K_0^4 = 0 . \quad (4.25)$$

Although the dispersion Equation (4.25) for surface waves in an optically active medium is fourth order for large , the solution to Equation (4.25) that satisfies condition (4.24b) would bive the surface wave dispersion relation. We find that one of the solutions of Equation (4.25) does not satisfy Equation (4.24b). In order to obtain the surface wave dispersion relation quantitatively, we need to know values of n_+ and n_- as functions of frequency. For this purpose we note that n_+ and n_- are given by

$$n_+(\omega) = \frac{\mu'(\omega)}{2} + \sqrt{\epsilon'(\omega) + \mu'^2(\omega)} \tag{4.26a}$$

$$n_-(\omega) = -\frac{\mu'(\omega)}{2} + \sqrt{\epsilon'(\omega) + \mu'^2(\omega)} \; . \tag{4.26b}$$

Subtracting Equation (4.26a) and (4.26b) we obtain

$$\mu'(\omega) = (n_+(\omega) - n_-(\omega)) \; . \tag{4.27}$$

In order to obtain the functional relation of μ' with frequency, we assume the Chandrasekhar formula[1] for the rotation $\phi(\lambda)$ in an optically active medium,

$$\phi(\lambda) = \frac{K_2 \lambda^2}{(\lambda^2 - \lambda_0^2)^2} \; , \tag{4.28}$$

where K_2 is the coefficient of rotation $\lambda = \frac{\omega}{c} \cdot 2\pi$, λ_0 is the resonance wave length; $\mu'(\omega)$ is related to by

$$\mu'(\omega) = \frac{2c}{\omega}\,\phi(\omega)\,, \tag{4.29}$$

where

$$\phi(\omega) = \frac{K_2\,\omega^2\omega_0^4}{(\omega_0^2-\omega^2)^2}\,. \tag{4.29a}$$

Using Equation (4.28) and (4.29) we obtain Equation (4.27).

We have used the oscillator model for $\epsilon'(\omega)$, defined by Equation (2.6). For the sake of completeness, we plot $n_+(\omega)$ and $n_-(\omega)$ as functions of frequency in Fig. 21 using the parameters of quartz. In Figs. 22 and 23 we plot the surface wave dispersion for parameters of quartz and sodium-chlorate $(\lambda_0 = 900\text{ A})$ respectively.

4.4 An Attenuated Total Reflection (ATR) Experiment

In order to probe the surface excitations discussed in the previous section we describe an Attenuated Total Reflection (ATR) experiment in which external light is made to couple with decaying surface waves, via an air gap between the prisim and optically active medium. The geometry of the problem is shown in Fig. 24. We shall find the electromagnetic fields in each region and match them at the boundaries in order to determine the ATR spectra. In the prism we have,

$$\vec{E}_{\text{total } p}(\vec{r},\omega) = \vec{E}_{inc}(\vec{r},\omega) + \vec{E}_{Ref}(\vec{r},\omega) \tag{4.30}$$

$$\vec{E}_{inc}(\vec{r},\omega) = \int (\hat{x}\, E_{x_i} + \hat{z}\, E_{z_i})\, e^{iW_p z}\, e^{iux}\, du \tag{4.31a}$$

$$\vec{E}_{Ref}(\vec{r},\omega) = \int (\hat{x}\, E_{R_x} + \hat{y}\, E_{R_y} + \hat{z}\, E_{R_z})\, e^{-iW_p z}\, e^{iux}\, du \tag{4.31b}$$

$$W_p = (\epsilon_p K_0^2 - u^2)^{1/2} \tag{4.32}$$

$$\vec{H}_{\text{total } p}(\vec{r},\omega) = \int \Big[\hat{x}\,(E_{z_i} e^{iW_p z} - E_{R_z} e^{-iW_p z}) + \hat{y}$$

$$\Big\{ (E_{x_i} e^{iW_p z} - E_{R_x} e^{-iW_p z})$$

$$- \frac{u}{W_p}(E_{z_i} e^{iW_p z} + E_{R_z} e^{-iW_p z}) \Big\} + \hat{z}\, \frac{u}{W_p} E_{R_y} e^{-iW_p z} \Big]. \tag{4.33}$$

In the air film region $-\Delta < Z < 0$, we have,

$$\vec{E}_{\text{total air}}(\vec{r},\omega) = \int (\hat{\vec{E}}^{(v)} e^{iW_0 z} + \hat{\vec{\vec{E}}}^{(v)} e^{-iW_0 z})\, e^{iux}\, du \tag{4.34}$$

$$W_0 = (K_0^2 - u^2)^{1/2}, \tag{4.35}$$

where $\hat{\vec{E}}^{(v)}$ and $\hat{\vec{\vec{E}}}^{(v)}$ have x , y and z components as $\hat{E}_x^{(v)}$, $\hat{E}_y^{(v)}$, $\hat{E}_z^{(v)}$, $\hat{\hat{E}}_x^{(v)}$, $\hat{\hat{E}}_y^{(v)}$, $\hat{\hat{E}}_z^{(v)}$

respectively.

$$\vec{H}^{(v)}(\vec{r},\omega) = \int [(\hat{x} (\hat{E}_y^{(v)} e^{-iW_0 z} - \hat{\bar{E}}_y^{(v)} e^{iW_0 z}) - \hat{y} (\hat{E}_x^{(v)} e^{-iW_0 z} - \hat{\bar{E}}_x^{(v)} e^{iW_0 z})) \frac{W_0}{K_0} + \hat{z} (\hat{E}_x^{(v)} e^{-iW_0 z} - \hat{\bar{E}}_x^{(v)} e^{iW_0 z}) \times \frac{(u^2 + W_0^2)}{K_0}] e^{iux} du \; . \tag{4.36}$$

In Equation (4.3b) we have used the condition that in vacuum the $\hat{E}$ fields are transverse

$$\vec{\eta}_1 \cdot \hat{\vec{E}}^{(v)} = 0 \tag{4.37a}$$

$$\vec{\eta}_2 \cdot \hat{\bar{\vec{E}}}^{(v)} = 0 \; , \tag{4.37b}$$

where

$$\vec{\eta}_1 \equiv (u, 0, -W_0)$$

and

$$\vec{\eta}_2 \equiv (u, 0, W_0) \; .$$

In the optically active region,

$$\vec{E}(\vec{r},\omega) = \int \left(\hat{\vec{E}}_{+} e^{i(ux + W_{+}z)} + \hat{\vec{E}}_{-} e^{i(ux - W_{-}z)} \right) du \,, \tag{4.38}$$

where $\hat{\vec{E}}_{+}$ and $\hat{\vec{E}}_{-}$ are given by Equations (4.22a) and (4.22b) with $\nu = 0$ and

$$\vec{H} = -\frac{i}{K_0} \vec{\nabla} \times \vec{E} \,. \tag{4.39}$$

Applying the boundary condition (continuity condition)

$$\hat{z} \times \vec{E} \Big|_{(-\Delta)_p} = \hat{z} \times \vec{E} \Big|_{(-\Delta)_V} , \quad \hat{z} \times \vec{H} \Big|_{(-\Delta)_p} = \hat{z} \times \vec{H} \Big|_{(-\Delta)_V} ,$$

$$\hat{z} \times \vec{E} \Big|_{0^-_V} = \hat{z} \times \vec{E} \Big|_{0^+_{Gyro}} , \quad \hat{z} \times H \Big|_{0^-_V} = \hat{z} \times \vec{H} \Big|_{0^+_{Gyro}} .$$

and considering that the electric field is transverse in both prism and vacuum, we obtain eigth algebraic equations and we have eight unknowns, namely

$$E_{R_x} \,,\; E_{R_y} \,,\; \hat{E}^{(v)}_x \,,\; \hat{E}^{(v)}_y \,,\; \hat{\bar{E}}^{(v)}_x \,,\; \hat{\bar{E}}^{(v)}_y \,,\; \hat{E}_{+x} \,,\; \hat{E}_{-x} \,.$$

In matrix form we can write the equations as:

$$
\begin{pmatrix}
e^{iw_p\Delta} & 0 & -e^{iw_0\Delta} & 0 & -e^{-iw_0\Delta} & 0 & 0 & 0 \\
0 & -e^{iw_p\Delta} & 0 & e^{iw_0\Delta} & 0 & e^{-iw_0\Delta} & 0 & 0 \\
-\frac{u}{w_p}e^{iw_p\Delta} & 0 & 0 & -\frac{w_0}{w_p}e^{iw_0\Delta} & 0 & \frac{w_0}{w_p}e^{-iw_0\Delta} & 0 & 0 \\
-\frac{(u^2+w_p^2)}{w_p^2}e^{iw_p\Delta} & 0 & -\frac{w_0}{w_p}e^{iw_0\Delta} & 0 & \frac{w_0}{w_p}e^{-iw_0\Delta} & 0 & 0 & 0 \\
0 & 0 & 1 & 0 & 0 & 0 & -1 & -1 \\
0 & 0 & 0 & 1 & 0 & 0 & -a & -c \\
0 & 0 & 0 & \frac{w_0}{k_0} & 0 & -\frac{w_0}{k_0} & -\alpha_+ & -\alpha_- \\
0 & 0 & \frac{w_0}{k_0} & 0 & -\frac{w_0}{k_0} & 0 & \alpha_+ a & \alpha_- c
\end{pmatrix}
\begin{pmatrix}
E_{Rx} \\ E_{Ry} \\ \hat{E}_x^{\vee} \\ \hat{E}_y^{\vee} \\ \hat{\bar{E}}_x^{\vee} \\ \hat{\bar{E}}_y^{\vee} \\ \hat{E}_{+x} \\ \hat{E}_{-x}
\end{pmatrix}
=
\begin{pmatrix}
-E_{xi}e^{-iw_p\Delta} \\ 0 \\ -E_{zi}e^{-iw_p\Delta} \\ -\frac{u}{w_p}e^{-iw_p\Delta}E_{zi} - E_{xi}e^{-iw_p\Delta} \\ 0 \\ 0 \\ 0 \\ 0
\end{pmatrix}
\tag{4.40}
$$

Finally we specialize to the case of circularly polarized incident light, i.e. we put $E_{z_1} = i E_{x_1}$ in the above matrix. Also in Equation (4.40) take

$$a = \frac{-i K_0 n_+ W_+}{(k_0^2 n_+^2 - u^2)} \tag{4.41a}$$

$$b = \frac{i K_0 n_- W_-}{(k_0^2 n_-^2 - u^2)} \tag{4.41b}$$

$$u = \sqrt{\epsilon_p}\, \frac{\omega}{c} \sin\theta \tag{4.42}$$

and

$$\alpha_+ = i n_+ \,, \quad \alpha_- = i n_- \,. \tag{4.43}$$

The reflection coefficient $R(\omega)$ at the exit face of the prism boundary is given by

$$R(\omega) = \left(\frac{W_p^2 + u^2}{W_p^2}\right) E_{R_x}^2 + E_{R_y}^2 \,. \tag{4.44}$$

In Figs. 25 and 26 we plot $R(\omega)$ as a function of frequency for various angles of incidence θ . In this computation we take the prism dielectric constant $\epsilon_p = 15$ and the air gap $\Delta = 10^{-5}$ cm.

4.5 Discussion

For quartz, as shown in Fig. 22, when the reduced frequency, $(\omega-\omega_0)/\omega_0$ is increased from 0.14 to 0.21, an increase in surface wave dispersion, U from 10^6 to $3\times10^6\ cm^{-1}$ is noted. Similarly an increase in reduced frequency, $(\omega-\omega_0)/\omega_0$ from 0.1 to 0.14 corresponds to an increase in surface wave dispersion, U from $0.97\times10^6\ cm^{-1}$ to $2.1\times10^6\ cm^{-1}$ for the parameters of sodium chlorate ($NaClO_3$), as shown in Fig. 23. In Figs. 25 and 26 ATR spectra are shown for quartz and sodium chlorate ($NaClO_3$) respectively.

In our present analysis, we have considered antisymmetric K -linear terms in the dielectric function which give rise to left and right circularly polarized light waves in the medium. An ATR experiment on optically active medium would be interesting as it would provide a comparison to our computed ATR spectra and would give more insight into the coupling of fields at the interface of a gyrotropic medium.

CHAPTER V

EXTINCTION THEOREM FOR MODEL GYROTROPIC MEDIUM WITH SPATIAL DISPERSION

5.1 Introduction

The interaction of electromagnetic fields with bounded material media can be described by two methods discussed by Born and Wolf.[11] The first is based on Maxwell's partial differential equations corresponding to macroscopic fields supplemented by the usual boundary conditions. The origin of the second method lies in an integral equation formulation based on molecular optics. This method connects the local field acting on the dipole to both the external applied field and the field radiated by the remaining dipoles. This technique in the case of linear optics was developed by Ewald[25] for crysalline media and Oseen[26] for amorphous media. A major one of their conclusions can be formulated in the form of a theorem: when an electromagnetic field is incident on the material medium bounded by a closed surface, the incident field is completely extinguished at every point inside the medium and is replaced by another wave with different velocity and direction of propagation. This is known as Ewald-Oseen's extinction theorem. There have been various modifications and generalizations of the Ewald-Oseen's extinction theorem in recent years, even though the theorem has been known for some 70 years.[49]

Birman and Sein applied the integral equation technique to non-local media. They considered translationally invarient susceptibility in the "dielectric approximation".

They obtained from the solution of the integral equation, a dispersion equation, extinction theorem and additional boundary condition (ABC) necessary to describe the electrodynamics of spatially dispersive media. Later, Frankel and Birman[8] considered a non-translationally invarient susceptibility to describe the spatially-dispersive medium and employed the integral equation technique to study non-local media. Agarwal et al ,[49(i)] Pattanayak and Wolf[49(k)] and others[49(a-h),j] have discussed the extinction theorem in some generality.

In this Chapter we shall apply the method used by Birman and Sein,[4] to optically active, spatially dispersive medium. In Section (5.2) we give brief review of Ewald-Oseen's analysis.[11(b)] In Section (5.3) we apply Birman and Sein's generalization to optically active spatially dispersive medium.[27] Finally Section (5.4) is devoted to a brief discussion of the results.

5.2 Integral Equation Formulation

Consider an electromagnetic wave in homogeneous, isotropic and non-magnetic medium. The electric and magnetic fields at the j th dipole are given by

$$\vec{E}_j{}' = \vec{E}^{(i)} + \sum_{\ell}{}' \vec{E}_{j\ell} \tag{5.1a}$$

$$\vec{H}_j{}' = \vec{H}^{(i)} + \sum_{\ell}{}' \vec{H}_{j\ell} \tag{5.1b}$$

where the sum extends over all the dipoles except the j^{th} dipole. At the point, r_j where the j^{th} dipole is situated, the fields due to the ℓ^{th} dipole are given by

$$\vec{E}_{j\ell} = \vec{\nabla} \times \vec{\nabla} \times \vec{\Pi}_e \tag{5.2a}$$

$$\vec{H}_{j\ell} = \frac{1}{c} \vec{\nabla} \times \dot{\vec{\Pi}}_e \quad , \tag{5.2b}$$

where Π_e is the electric Hertz vector defined by

$$\vec{\Pi}_e = \frac{\vec{p}_\ell(t - R_{j\ell}/c)}{R_{j\ell}} \quad , \tag{5.2c}$$

here $\vec{p}_\ell$ is the dipole moment of the ℓ^{th} dipole and $R_{j\ell}$ is the distance between ℓ^{th} and j^{th} dipole. In the limit of the wavelength, λ much larger than spacing between dipoles; we can treat the distribution of dipoles to good approximation to be continuous i.e. the moment of the dipoles can be considered as a continuous function of position and time. Thus we can write Equation (5.1a) as

$$\vec{E}_L(\vec{r},t) = \vec{E}^{(i)}(\vec{r},t) + \int_{\sigma(r)}^{\Sigma} \vec{\nabla} \times \vec{\nabla} \times \left(\frac{\vec{P}(\vec{r}',t - R/c)}{R} \right) d\vec{v}' , \tag{5.3}$$

where the integral is taken over the interior of the crystal, V is the volume occupied by the crystal, Σ is the surface bounding the volume and $\sigma(r)$ is the surface surrounding the small volume excluded at the point, r which the field is being calculated.

The electric field in vacuum satisfies the equation,

$$(\nabla^2 + K_0^2)\,\bar{E}(\vec{r},\omega) = 0 \;, \tag{5.4}$$

where $K_0 = \omega/c$. The Green's function satisfies the equation

$$(\nabla^2 + K_0^2)\,G(R) = -4\pi\,\delta(r-r') \tag{5.5a}$$

$$G(R) = \frac{e^{iK_0R}}{R} \;, \qquad R = |\vec{r}-\vec{r}'| \;. \tag{5.5b}$$

Considering harmonic time dependence of the fields and using Equations (5.5b) we can write Equation (5.3) as

$$\vec{E}_L(\vec{r},\omega) = \vec{E}^{(i)}(\vec{r},\omega) + \int_{\sigma(r)}^{\Sigma} \vec{\nabla}\times\vec{\nabla}\times\left(\vec{P}(\vec{r}',\omega)\,G(r,r',\omega)\right)d\vec{r}' \;. \tag{5.6}$$

Next using the identity due to Hoek[11(c)]

$$\vec{\nabla}\times\vec{\nabla}\times\int_{\sigma}^{\Sigma} G(r,r',\omega)\,\vec{P}(\vec{r}')\,dr' - \frac{8\pi}{3}\vec{P}(\vec{r}) = \int_{\sigma}^{\Sigma} \vec{\nabla}\times\vec{\nabla}\times\left(G(r,r')P(r')\right)dr' \;, \tag{5.7}$$

we can write Equation (5.6) as

$$\vec{E}_L(\vec{r},\omega) = \vec{E}^{(i)}(\vec{r},\omega) + \vec{\nabla}\times\vec{\nabla}\times\int_{\sigma}^{\Sigma} G(\vec{r},\vec{r}')\,\vec{P}(r',\omega)\,dr' - \frac{8\pi}{3}\vec{P}(\vec{r},\omega) \;. \tag{5.8}$$

The polarization $\vec{P}(r,\omega)$ in the medium satisfies the equation

$$(\nabla^2 + K^2)\, \vec{P}(\vec{r},\omega) = 0 \quad . \tag{5.9}$$

Using Equations (4.5a) and (4.9) we can write

$$\vec{P}\, G = \frac{1}{(K^2 - K_0^2)} \left(\vec{P}\, \nabla^2 G - G\, \nabla^2 \vec{P} \right). \tag{5.10}$$

Using Green's theorem

$$\int_\sigma^\Sigma \vec{P}\, G\, d\vec{r}\,' = \frac{1}{(K^2 - K_0^2)} \left[\int_\Sigma \left(\vec{P} \frac{\partial G}{\partial n'} - G \frac{\partial \vec{P}}{\partial n'} \right) ds' - \int_\sigma \left(\vec{P} \frac{\partial G}{\partial n'} - G \frac{\partial \vec{P}}{\partial n'} \right) ds' \right]. \tag{5.11}$$

The second surface integral on the right hand side goes to $-4\pi \vec{P}(\vec{r})$ as the radius, a of the excluded sphere whose surface area is σ goes to zero. Thus we can write Equation (5.8) using Equation (5.11) as

$$\vec{E}_L(\vec{r},\omega) = \vec{E}^{(i)}(\vec{r},\omega) + \frac{1}{(K^2 - K_0^2)} \vec{\nabla} \times \vec{\nabla} \times \int \left(\vec{P} \frac{\partial G}{\partial n'} - G \frac{\partial \vec{P}}{\partial n'} \right) ds' + \frac{4\pi \vec{P}}{(K^2 - K_0^2)} - \frac{8\pi}{3} \vec{P}. \tag{5.12}$$

Inserting $\vec{P}(\vec{r},\omega) = N\alpha(\omega)\, \vec{E}_L(\vec{r},\omega)$ in (5.12) we can rewrite Equation (5.12) as

$$\vec{P}(\vec{r},\omega) = N\alpha(\omega)\left[\vec{E}^{(i)}(\vec{r},\omega) + \frac{1}{(K^2-K_0^2)}\vec{\nabla}\times\vec{\nabla}\times\int_{\Sigma}\left(P\frac{\partial G}{\partial n'} - G\frac{\partial P}{\partial n'}\right)ds'\right]$$

$$+ \frac{4\pi}{3}N\alpha(\omega)\left(\frac{n^2+2}{n^2-1}\right)\vec{P}(\vec{r}',\omega) . \qquad (5.13)$$

Clearly we have two terms. One, according to Equation (5.9), propagates with velocity $C/n(\omega)$. The second term represents a wave which like G is propagated with the vacuum velocity of light, C. Thus we can observe that Equation (5.13) splits into two groups, each vanishing separately,

$$\frac{4\pi}{3}N\alpha(\omega) = \left(\frac{n^2-1}{n^2+2}\right) \qquad (5.14)$$

$$\vec{E}^{(i)}(\vec{r},\omega) + \vec{\nabla}\times\vec{\nabla}\times\int_{\Sigma}\left(\vec{E}\frac{\partial G}{\partial n'} - G\frac{\partial \vec{E}}{\partial n'}\right)ds' = 0 . \qquad (5.15)$$

Equation (5.15) represents the extinction of the incident wave $\vec{E}^{(i)}(\vec{r},\omega)$ at every point inside the medium by interference with part of the dipole field. The incident wave is replace by another wave, $\vec{E}_L$ given by

$$\vec{E}_L(\vec{r},\omega) = \frac{1}{N\alpha(\omega)}\vec{P}(\vec{r},\omega) = \frac{4\pi}{3}\left(\frac{n^2+2}{n^2-1}\right)\vec{P}(\vec{r},\omega) , \qquad (5.16)$$

with velocity $C/n(\omega)$. Finally we can connect the effective local field, $\vec{E}_L(\vec{r},\omega)$ with Maxwell's field, $\vec{E}(\vec{r},\omega)$ which is given by

$$\vec{E}(\vec{r},\omega) = \frac{4\pi}{(n^2-1)} \vec{P}(\vec{r},\omega) \quad . \tag{5.17}$$

Equations (4.16) and (4.17) imply

$$\vec{E}_L(\vec{r},\omega) = \vec{E}(\vec{r},\omega) + \frac{4\pi}{3} \vec{P}(\vec{r},\omega) \quad . \tag{5.18}$$

Thus using the integral equation formulation we obtain conditions for: extinction of the electomagnetic fields, the Lorentz-Lorenz relation and finally the connection between local fields and macroscopic fields in the frequency dispersive media.

In the next section we shall apply the generalization of the integral equation technique used by Birman and Sein to optically active, spatially dispersive media.

5.3 Application to Optically Active Medium with Spatial Dispersion

When an electromagnetic wave impinges upon an optically active medium from vacuum, in general two modes are excited in the medium. These are left and right circularly polarized waves. In this section we consider an optically active medium which exhibits spatial dispersion. We employ the integral equation framework to obtain the dispersion relation, the extinction theorem and the additional boundary conditions required to solve the reflection problem.

5.3.1 Constitutive Relation

Let us denote $\vec{E}(\vec{r},t)$ as macroscopic electric field and $\vec{P}(\vec{r},t)$ the dielectric polarization at $(\vec{r},t)$. The polarization and electric field are related by a phenomenological constitutive relation

$$\vec{P}(\vec{r},\omega) = \int \overleftrightarrow{\chi}(\vec{r}-\vec{r}',\omega)\,\vec{E}(\vec{r}',\omega)\,dr' , \tag{5.19}$$

where we take an "isotropic" model for which only diagonal terms in the susceptibility tensor are non zero, and the kernel,

$$\overleftrightarrow{\chi}(\vec{r}-\vec{r}',\omega) = \overleftrightarrow{\chi}(R,\omega) = \chi^{N}(R,\omega)\,\mathbb{I} - \chi^{G}(R,\omega)\vec{\nabla}_{r}\times \tag{5.20}$$

is a sum of a non-gyrotropic part, $\chi^{N}(R,\omega)$ and a gyrotropic part, χ^{G}. Both of the latter are considered as resonance type of oscillator model,

$$\chi^{N}(R,\omega) = \chi_0\,\delta(R) + \chi_1\,G_+(R,\omega) \tag{5.21a}$$

$$\chi^{G}(R,\omega) = \chi_0'\,\delta(R) + \chi_1'\,G_+(R,\omega) . \tag{5.21b}$$

Here $R = (\vec{r}-\vec{r}')$, $G_+(R,\omega) = [\exp(iK_+R)]/R$, $\mathbb{I}$ is the unit dyadic and k_+ is a complex wave number which is defined as the pole of the Fourier transform of $\chi(R,\omega)$:

$$\chi(R,\omega) \xrightarrow{F.T.} \chi(K,\omega) = \frac{(\epsilon_\infty - 1)}{4\pi} + \frac{F/D}{(k^2 - K_t^2)} , \qquad (5.22)$$

and $K_t^2 = -(\omega_0^2 - \omega^2 - i\omega\Gamma) / D$. Here ϵ_∞ is the background dielectric constant, Γ is the damping constant, ω_0 is the exciton resonance frequency; $D = \hbar\omega_0/M^*$, where M^* is the effective exciton mass; and $F = \alpha_0\omega_0^2$, where α_0 is the oscillator strength. χ_0 and χ_1 appearing in Equation (5.21) are given by

$$\chi_0 = [(\epsilon_\infty - 1)/4\pi] (2\pi)^{3/2} \qquad (5.23a)$$

$$\chi_1 = \pi F / D (2\pi)^{1/2} \qquad (5.23b)$$

χ_0' and χ_1' appearing in Equation (5.21b) have a similar form as Equation (5.23).

5.3.2 The Integral Equation: Polarization Framework

The basic equation of molecular optics, which relates local field, $\vec{E}_L(\vec{r},\omega)$ and induced polarization $\vec{P}(\vec{r},\omega)$, is given by Equation (5.8),

$$\vec{E}_L(\vec{r},\omega) = \vec{E}^{(i)}(\vec{r},\omega) + \vec{\nabla}\times\vec{\nabla}\times\int_\sigma^\Sigma G(\vec{r},\vec{r}')\,\vec{P}(\vec{r}',\omega)\,d\vec{r}' - \frac{8\pi}{3}\vec{P}(\vec{r},\omega) , \qquad (5.8)$$

where $\vec{E}^{(i)}(\vec{r},\omega)$ is the incident electric field. If we assume that the Lorentz-Lorenz relation Equation (5.18) applies, we can write Equation (5.8) as

$$\vec{E}(\vec{r},\omega)+4\pi\vec{P}(\vec{r},\omega)=\vec{E}^{(i)}(\vec{r},\omega)+\vec{\nabla}\times\vec{\nabla}\times\int_{\sigma^{(1)}}^{\Sigma}\vec{P}(\vec{r}',\omega)\,G(R)\,d\vec{r}' \quad (5.24)$$

Next, multiplying both sides by $(2\pi)^{-3/2}\,\chi(\vec{r}''-\vec{r})$, defined by Equations (5.20) and (5.21), and integrating over r we obtain

$$\vec{P}(\vec{r}'',\omega)+\frac{4\pi\chi_0}{(2\pi)^{3/2}}\vec{P}(\vec{r}'',\omega)-\frac{4\pi}{(2\pi)^{3/2}}\chi_0'\,\vec{\nabla}''\times\vec{P}(\vec{r}'',\omega)$$

$$+\frac{4\pi\chi_1}{(2\pi)^{3/2}}\int^{\Sigma}P'(\vec{r},\omega)\,G_+(\vec{r}''-\vec{r})\,d\vec{r}-\frac{4\pi\chi_1'}{(2\pi)^{3/2}}\int^{\Sigma}G_+(\vec{r}''-\vec{r})\vec{\nabla}\times P\,d\vec{r}$$

$$=\frac{\chi_0}{(2\pi)^{3/2}}\vec{E}^{(i)}(\vec{r}'',\omega)-\frac{\chi_0'}{(2\pi)^{3/2}}\vec{\nabla}''\times E^{(i)}(\vec{r}'',\omega)+$$

$$+\frac{\chi_1}{(2\pi)^{3/2}}\int^{\Sigma}G_+(\vec{r}''-\vec{r})\,\vec{E}^{(i)}(\vec{r},\omega)\,d\vec{r}-\frac{\chi_1'}{(2\pi)^{3/2}}\int^{\Sigma}G_+(\vec{r}''-\vec{r})\,\vec{\nabla}\times\vec{E}^{(i)}d\vec{r}$$

$$+\frac{\chi_0}{(2\pi)^{3/2}}\vec{\nabla}''\times\vec{\nabla}''\times\int_{\sigma}^{\Sigma}\vec{P}(\vec{r}',\omega)\,G(\vec{r}'-\vec{r}'')\,d\vec{r}'$$

$$-\frac{\chi_0'}{(2\pi)^{3/2}}\vec{\nabla}''\times\vec{\nabla}''\times\vec{\nabla}''\times\int_{\sigma}^{\Sigma}\vec{P}(\vec{r}',\omega)\,G(\vec{r}'-\vec{r}'')\,d\vec{r}'$$

$$+\frac{\chi_1}{(2\pi)^{3/2}}\int^{\Sigma}G_+(\vec{r}''-\vec{r})\,\vec{\nabla}\times\vec{\nabla}\int_{\sigma}^{\Sigma}\vec{P}(\vec{r}',\omega)G(\vec{r}'-\vec{r})\,d\vec{r}'d\vec{r}$$

$$-\frac{\chi_1'}{(2\pi)^{3/2}}\int^{\Sigma}G_+(\vec{r}''-\vec{r})\vec{\nabla}\times\vec{\nabla}\times\vec{\nabla}\times\int_{\sigma}^{\Sigma}\vec{P}(\vec{r}',\omega)G(\vec{r}'-\vec{r})\,d\vec{r}'d\vec{r} \quad (5.25)$$

The integral equation (5.25) has both longitudinal and transverse solutions. We concentrate here on transverse solutions. We note that the incident wave $\vec{E}^{(i)}(\vec{r},\omega)$ satisfies Equation (5.4). In the optically active medium, let us assume the total polarization is a linear combination of plane waves, each satisfying wave equation with undetermined wave vector, K_j . Thus

$$\vec{P}(\vec{r},\omega) = \sum_{j=1}^{S} \alpha_j \vec{P}_j(\vec{r},\omega) . \tag{5.26}$$

In order to solve Equation (5.25) one has to transform expressions from volume to surface integrals. The typical basic integrals involved are,

$$\vec{F}(\vec{r}'') = \int G_+(\vec{r}''-\vec{r})\, \vec{P}(\vec{r})\, d\vec{r} \tag{5.27a}$$

$$\vec{E}(\vec{r}'') = \int G_+(\vec{r}''-\vec{r})\, \vec{\nabla}_r \times \vec{P}(\vec{r})\, d\vec{r} , \tag{5.27b}$$

we write $\vec{E}(\vec{r}'')$ in the following form

$$\begin{aligned}
E_i(\vec{r}'') &= \int G_+(\vec{r}''-\vec{r})\, (\vec{\nabla}_r \times \vec{P})_i\, d\vec{r} \\
&= \int G_+(\vec{r}''-\vec{r})\, \epsilon_{ilm}\, \partial_\ell P_m\, d\vec{r} \\
&= -\int \epsilon_{i\ell m} [\partial_\ell G_+(\vec{r}''-\vec{r})] P_m(\vec{r})\, d\vec{r} + \int \epsilon_{i\ell m}\, \partial_\ell [G_+(\vec{r}''-\vec{r})\, P_m(\vec{r})]\, d\vec{r}
\end{aligned}$$

$$= \int \epsilon_{i\ell m}\left[\partial_\ell'' G_+(\vec{r}''-\vec{r})\right] P_m(\vec{r})\, d\vec{r} + \int \epsilon_{i\ell m}\, \partial_\ell \left[G_+(\vec{r}''-\vec{r}) P_m(\vec{r})\right] d\vec{r}$$

$$= \epsilon_{i\ell m}\, \partial_\ell'' \int G_+(\vec{r}''-\vec{r})\, P_m(\vec{r})\, d\vec{r} + \epsilon_{i\ell m} \int \partial_\ell \left[G_+(\vec{r}''-\vec{r}) P_m(\vec{r})\right] d\vec{r}$$

$$= \epsilon_{i\ell m}\, \partial_\ell''\, F_m(r'') \quad + \quad \int (\text{Total Derivative}) \, .$$

Thus we obtain,

$$\vec{E}(\vec{r}'') = \vec{\nabla}'' \times \vec{F}(\vec{r}'') \, . \tag{5.27c}$$

provide the contribution of the total derivative term vanishes: it is satisfied for the polarization vanishing on the surface. Details of the evaluation of the inegral (5.27a) are given in appendices (B) and (D) of Ref. (4). Thus substituting Equations (5.26) and (5.27c) in Equation (5.25) and carrying out the integrals, we obtain terms propagating with K_J, K_0 and K_+ respectively. The separate vanishing of these terms produce:

(i) The dispersion relation

(ii) Condition for extinction of incident field in the medium

(iii) Additional Boundary Conditions (ABC).

In the following sections we shall describe the expressions obtained in each case.

5.3.3. Dispersion Equation, Mode Structure

Requiring the vanishing of the terms propagating with K_j results is

$$-\sum_{j=1}^{S}\Big[P_j^{\ell}\Big(1+\frac{4\pi\chi_0}{(2\pi)^{3/2}}+\frac{4\pi\chi_1}{(2\pi)^{3/2}}\frac{1}{(K_j^2-K_+^2)}-4\pi\chi_0'\, i\,\epsilon_{i\ell m}K_m^{(j)}$$

$$-\frac{4\pi\chi_1'\, i\,\epsilon_{i\ell m}K_m^{(j)}}{(K_j^2-K_+^2)}-\frac{4\pi\chi_0}{(2\pi)^{3/2}}\frac{K_j^2}{(K_j^2-K_0^2)}-\frac{4\pi\chi_1}{(2\pi)^{3/2}}\frac{K_j^2}{(K_j^2-K_0^2)(K_j^2-K_+^2)}$$

$$+\frac{4\pi\chi_0'}{(2\pi)^{3/2}}\frac{K_j^2\, i\,\epsilon_{i\ell m}K_m^{(j)}}{(K_j^2-K_+^2)}+\frac{4\pi\chi_1'}{(2\pi)^{3/2}}\frac{K_j^2\, i\,\epsilon_{i\ell m}K_m^{(j)}}{(K_j^2-K_0^2)(K_j^2-K_+^2)}\Big)\Big]. \qquad (5.28)$$

This implies

$$(\nabla^4-\alpha\nabla^2+\beta)\,\vec{P}+(\eta\nabla^2+\delta)\,\vec{\nabla}\times\vec{P}=0, \qquad (5.29)$$

where,

$$\alpha=\Big(-K_0^2\Big(1+\frac{4\pi\chi_0}{(2\pi)^{3/2}}\Big)-K_+^2\Big) \qquad (5.30a)$$

$$\beta=\Big(K_+^2K_0^2\Big(1+\frac{4\pi\chi_0}{(2\pi)^{3/2}}\Big)-K_0^2\,\frac{4\pi\chi_1}{(2\pi)^{3/2}}\Big) \qquad (5.30b)$$

$$\eta=K_0^2\,4\pi\chi_0'/(2\pi)^{3/2} \qquad (5.30c)$$

$$\delta = \left(\frac{4\pi\chi_1'}{(2\pi)^{3/2}} K_0^2 - \frac{4\pi\chi_0'}{(2\pi)^{3/2}} K_+^2 K_0^2 \right) . \tag{5.30d}$$

We can write Equation (5.29) to a higher order uncoupled equation

$$(\nabla^4 - \alpha\nabla^2 + \beta)^2 \vec{P} = -(-\eta\nabla^2 + \delta)^2 \nabla^2 \vec{P} , \tag{5.31}$$

considering plane waves as propagatin modes,[40] we obtain

$$(K^4 + \alpha K^2 + \beta)^2 = (\eta K^2 + \delta)^2 K^2 , \tag{5.32}$$

or

$$K^8 + a K^6 + b K^4 + c K^2 + d = 0 , \tag{5.33}$$

where

$$a = (-\eta^2 + 2\alpha) \tag{5.34a}$$

$$b = (\alpha^2 + 2\beta - 2\eta\delta) \tag{5.34b}$$

$$c = (2\alpha\beta - \delta^2) \tag{5.34c}$$

$$d = \beta^2 . \tag{5.34d}$$

Equation (5.33) is the dispersion relation, which gives 4 propagating modes in the medium. Using Equations (5.29) and (5.31) we can write the mode structure as

$$\vec{\nabla} \times \vec{P}_j = \pm K_j \vec{P}_j \tag{5.35a}$$

Let us consider propagation in the z direction,

$$K_j \hat{z} \times \vec{P}_j = \pm K_j \vec{P}_j$$

or

$$\hat{z} \times \vec{P}_j = \pm \vec{P}_j \tag{5.35b}$$

Equation (5.35b) implies that two of the propagating modes are left circularly polarized, and two are right circularly polarized, the mode structure of the electric field can be written as

$$\vec{E}_j = (\hat{x} - i\,\hat{y})\, L_j \qquad j = 1, 2 \tag{5.36a}$$

$$(\hat{x} + i\,\hat{y})\, R_j \qquad j = 3, 4 \ , \tag{5.36b}$$

and $\vec{P}_j = (n_j^2 - 1)\ \vec{E}_j$.

5.3.4 <u>Extinction Condition</u>

Requiring the vanishing of the bracket containing terms propagating at K_o produces

$$\frac{\chi_0}{(2\pi)^{3/2}}\vec{E}^{(i)} + \frac{4\pi}{(2\pi)^{3/2}}\frac{\vec{E}^{(i)}}{(K_0^2-K_+^2)} + \frac{\chi_0}{(2\pi)^{3/2}}\sum_{j=1}^{4}\frac{\vec{A}_0(\vec{P}_j)}{(K_j^2-K_0^2)} + \frac{4\pi\chi_1}{(2\pi)^{3/2}}\sum_{j=1}^{4}\frac{\vec{A}_0(\vec{P}_j)}{(K_j^2-K_0^2)(K_0^2-K_+^2)}$$

$$-\frac{\chi_0'}{(2\pi)^{3/2}}\vec{\nabla}\times\vec{E}^{(i)} + \frac{4\pi}{(2\pi)^{3/2}}\frac{\vec{\nabla}\times\vec{E}^{(i)}}{(K_0^2-K_+^2)} + \frac{\chi_0}{(2\pi)^{3/2}}\sum_{j=1}^{4}\frac{\vec{\nabla}\times\vec{A}_0}{(K_j^2-K_0^2)} + \frac{4\pi\chi_1}{(2\pi)^{3/2}}\sum_{j=1}^{2}\frac{\vec{\nabla}\times\vec{A}_0(P_j)}{(K_j^2-K_0^2)(K_0^2-K_1^2)} = 0 \tag{5.37}$$

where

$$\vec{A}_0(\vec{P}_j) = \vec{\nabla}\times\vec{\nabla}\times\vec{S}_c'(\vec{P}_j') \tag{5.38a}$$

$$\vec{S}_0'(\vec{P}_j) = \int^{\Sigma} ds'\left(\vec{P}_j(\vec{r}',\omega)\frac{\partial G}{\partial n}(\vec{r}'-r'') - G(\vec{r}'-\vec{r}'')\frac{\partial P_j(\vec{r}',\omega)}{\partial n}\right). \tag{5.38b}$$

Here n refers to the normal derivative and the integral is taken over the surface Σ. We can write Equation (5.37) as

$$E^{(i)}(\vec{r}) + \sum_{j=1}^{4}\frac{\vec{A}_c'(\vec{P}_j)}{(K_j^2-K_0^2)} = 0 \tag{5.39}$$

and

$$\vec{\nabla}\times E^{(i)}(\vec{r}') + \sum_{j=1}^{4}\frac{\vec{\nabla}\times\vec{A}_0'(\vec{P}_j')}{(K_j^2-K_0^2)} = 0 \tag{5.40}$$

Equation (5.39) corresponds to the condition for extinction of incident electric field in the medium. Equation (5.40) is the new term representing extinction of incident magnetic field in the medium.

5.3.5 Additional Boundary Condition

Finally, vanishing of the bracket containing terms propagatin at K_+ gives

$$\chi_1 \left(\frac{2}{\pi}\right)^{1/2} \sum_{j=1}^{4} \frac{\vec{S}_+(\vec{P}_j)}{(K_j^2 - K_+^2)} \left(\frac{K_j^2}{(K_j^2 - K_0^2)} - 1\right)$$

$$-\chi_1' \left(\frac{2}{\pi}\right)^{1/2} \sum_{j=1}^{4} \frac{\vec{\nabla} \times \vec{S}_+(\vec{P}_j)}{(K_j^2 - K_+^2)} \left(\frac{K_j^2}{(K_j^2 - K_0^2)} - 1\right)$$

$$+\frac{\chi_1}{(2\pi)^{3/2}} \frac{1}{(K_0^2 - K_+^2)} \left(\vec{S}_+(\vec{E}^{(i)}) + \sum_{j=1}^{4} \frac{\vec{S}_+(A_0(P_j))}{(K_j^2 - K_+^2)}\right)$$

$$-\frac{\chi_1'}{(2\pi)^{3/2}} \frac{1}{(K_0^2 - K_+^2)} \left(S_+(\vec{\nabla} \times \vec{E}^{(i)}) + \sum_{j=1}^{4} \frac{\vec{S}_+(\nabla \times A_0(P_j))}{(K_j^2 - K_+^2)}\right), \qquad (5.41)$$

where

$$\vec{S}_+(\vec{P}_j) = \int ds' \left(\vec{P}_j(\vec{r}',\omega) \frac{\partial G_+}{\partial n}(\vec{r}' - \vec{r}'') - G(\vec{r}' - \vec{r}'') \frac{\partial P(\vec{r}',\omega)}{\partial n}\right) \qquad (5.42a)$$

$$\vec{S}_+(\vec{A}_0(\vec{P}_j)) = \int ds' \left(\vec{A}_0(\vec{P}_j) \frac{\partial G_+}{\partial n}(\vec{r}' - \vec{r}'') - G(\vec{r}' - \vec{r}'') \frac{\partial}{\partial n} A_0(P_j)\right) \qquad (5.42b)$$

$$S_+(E^{(i)}) = \int ds' \left(\vec{E}^{(i)} \frac{\partial G_+}{\partial n}(\vec{r}' - \vec{r}'') - G(\vec{r}' - \vec{r}'') \frac{\partial E^{(i)}}{\partial n}\right). \qquad (5.42c)$$

Using the extinction conditions Equations (5.39) and (5.40) we find that the last two terms in Equation (5.41) vanish and we can write

$$\chi_1\left(\frac{2}{\pi}\right)^{1/2} \sum_{j=1}^{4} \vec{S}_+(\vec{P}_j) \frac{K_0^2}{(K_j^2-K_+^2)(K_j^2-K_0^2)}$$

$$-\chi_1'\left(\frac{2}{\pi}\right)^{1/2} \sum_{j=1}^{4} \frac{\vec{\nabla}\times\vec{S}_+(\vec{P}_j)}{(K_j^2-K_+^2)} \frac{K_0^2}{(K_j^2-K_0^2)} = 0, \tag{5.43}$$

which implies that the necessary condition is

$$\sum_{j=1}^{4} \frac{\vec{S}_+(\vec{P}_j)\ K_0^2}{(K_j^2-K_+^2)(K_j^2-K_0^2)} = 0 . \tag{5.44}$$

Equation (5.44) represents the additional boundary conditions required to solve the reflection and transmission problem at the boundary of the optically active-spatially dispersive half space.

5.3.6 Normal Incidence on a Plane Surface

Consider the problem of normal incidence along the z -direction on the semi infinite optically active, spatially dispersive half space. Following Ref. (4), we can write

$$\vec{S}_0(\vec{P}_j) = -2\pi\ \vec{P}_j(K_j,\omega)\ (K_0+K_j)\ \frac{e^{iK_0 r}}{K_0} \tag{5.45}$$

$$\vec{S}_+(\vec{P}_j) = -2\pi\ \vec{P}_j(K_j,\omega)\ (K_++K_j)\ \frac{e^{iK_+ r}}{K_+} \tag{5.46}$$

$$\vec{A}_0(\vec{P}_j) = \vec{\nabla} \times \vec{\nabla} \times \vec{S}_0(\vec{P}_j) = K_0^2 \, \vec{S}_0(\vec{P}_j) \, . \tag{5.47}$$

Using Equations (5.45) - (5.47) and the mode structure represented by Equations (5.36a) and (5.36b) we can write the extinction condition Equations (5.39) and (5.40) as

$$\vec{E}^{(i)}(K_0,\omega) = \frac{1}{2}\sum_{j=1}^{2}(n_j+1)(\hat{x}-i\hat{y})L_j + \frac{1}{2}\sum_{j=3}^{4}(n_j+1)(\hat{x}+i\hat{y})R_j \tag{5.48}$$

$$\vec{B}^{(i)}(K_0,\omega) = \frac{1}{2}\sum_{j=1}^{2}n_j(n_j+1)(\hat{x}-i\hat{y})L_j + \frac{1}{2}\sum_{j=3}^{4}n_j(n_j+1)(\hat{x}+i\hat{y})R_j \, . \tag{5.49}$$

Equations (5.48) and (5.49) represent the extinction conditions in terms of amplitudes of the modes. Again using Equations (5.45) - (5.47) the additional boundary condition Equation (5.44) becomes

$$\vec{P} - \frac{i}{K_+}\frac{d\vec{P}}{dz}\bigg|_{z=0} = 0 \, . \tag{5.50}$$

The components of Equation (5.50) can be written as

$$P_x - \frac{i}{K_+}\frac{dP_x}{dz} = 0 \tag{5.51a}$$

$$P_y - \frac{i}{K_+}\frac{dP_y}{dz} = 0 \, . \tag{5.51b}$$

Using Equations (5.51a) and (5.51b) we can write

$$\Pi^{\mp} - \frac{i}{K_+} \left.\frac{d\Pi^{\mp}}{dz}\right|_{z=0} = 0 \; , \qquad \Pi^{\mp} = P_y \mp i P_x \; . \tag{5.52}$$

Using the additional boundary conditions Equation (5.52) we can solve the reflection and transmission problem at the optically active, spatially dispersive half space.

Fields in the vacuum region, $z<0$ are given by

$$\vec{E}_{out}(z,\omega) = \hat{x} E_o e^{iK_0 z} + (\hat{x} E_x + \hat{y} E_y) e^{-iK_0 z} \tag{5.53}$$

$$H_{out}(z,\omega) = \hat{y} E_o e^{iK_0 z} + (\hat{x} E_y - \hat{y} E_x) e^{-iK_0 z} , \tag{5.54}$$

where E_o is the amplitude of the incident field and, E_x and E_y are the x and y components of amplitudes of reflected fields.

Fields in the optically active spatially dispersive half space, $z>0$ are given by

$$\vec{E}_{inside}(z,\omega) = \sum_{j=1}^{2} L_j (\hat{x} - i\hat{y}) e^{iK_j z} + \sum_{j=3}^{4} R_j (\hat{x} + i\hat{y}) e^{iK_j z} \tag{5.55}$$

$$\vec{H}_{inside}(z,\omega) = \sum_{j=1}^{2} L_j n_j (\hat{x} - i\hat{y}) e^{iK_j z} + \sum_{j=3}^{4} R_j n_j (\hat{x} + i\hat{y}) e^{iK_j z} . \tag{5.56}$$

The additional boundary conditions (5.52) can be written as

$$L_1 (n_1^2 - 1)\left(1 + \frac{n_1}{n_+}\right) + L_2 (n_2^2 - 1)\left(1 + \frac{n_2}{n_+}\right) = 0 \tag{5.57}$$

$$R_3(n_3^2-1)\left(1+\frac{n_3}{n_+}\right)+R_4(n_4^2-1)\left(1+\frac{n_4}{n_+}\right)=0\,. \tag{5.58}$$

Using the continuity of fields at $z=0$, and Equations (5.57) and (5.58) we obtain

$$\frac{E_{Rx}}{E_0}=\frac{\left[1-\dfrac{(n_2^2-1)}{(n_1^2-1)}\dfrac{(1+n_2/n_+)}{(1+n_1/n_+)}\right]}{\left[(n_2+1)-(n_1+1)\dfrac{(n_2^2-1)}{(n_1^2-1)}\dfrac{(1+n_2/n_+)}{(1+n_1/n_+)}\right]}$$

$$+\frac{\left[1-\dfrac{(n_4^2-1)}{(n_3^2-1)}\dfrac{(1+n_4/n_+)}{(1+n_3/n_+)}\right]}{\left[(n_4+1)-(n_3+1)\dfrac{(n_4^2-1)}{(n_3^2-1)}\dfrac{(1+n_4/n_+)}{(1+n_3/n_+)}\right]}-1 \tag{5.59}$$

$$\frac{E_{Ry}}{E_0}=i\,\frac{\left[1-\dfrac{(n_4^2-1)}{(n_3^2-1)}\dfrac{(1+n_4/n_+)}{(1+n_3/n_+)}\right]}{\left[(n_4+1)-(n_3+1)\dfrac{(n_4^2-1)}{(n_3^2-1)}\dfrac{(1+n_4/n_+)}{(1+n_3/n_+)}\right]}$$

$$\frac{-i\left[1-\frac{(n_2^2-1)}{(n_1^2-1)}\frac{(1+n_2/n_+)}{(1+n_1/n_+)}\right]}{\left[(n_2+1)-(n_1+1)\frac{(n_2^2-1)}{(n_1^2-1)}\frac{(1+n_2/n_+)}{(1+n_1/n_+)}\right]} \quad . \tag{5.60}$$

The reflection coefficient $R(\omega)$ is given by

$$R(\omega) = \sqrt{\left(|E_{Rx}|^2 + |E_{Ry}|^2\right)/E_0^2} \quad . \tag{5.61}$$

5.4 Discussion

We have employed an integral equation technique to obtain some rigorous results in a optically active, spatial dispersive media. We obtained a dispersion equation which gives 4 propagating modes, two of them are left and the other two are right circularly polarized. This clearly demonstrates the effect of spatial dispersion i.e. doubling of modes occurs in contrast to usual gyrotropy where one has one left and one other right circularly polarized light wave. Extinction conditions are obtaind. One of the extinction conditions corresponds to extinction of electric field as noted by Birman and Sein. The new term of Equation (5.40) corresponds to the extinction of the magnetic field. Finally, we obtain the "ABC's" required to solve the reflection problem from the vacuum-medium interface. We have analyzed the problem of normal incidence on vacuum-medium interface using these additional boundary conditions.

In conclusion we may point out that the analysis carried out above requries the integral equation formulation of optics based upon polarization picture and validity of the Lorentz-Lorenz relation. If we require that the Maxwells' equation approach is consistent with the integral equation approach, then the Lorentz-Lorenz relation comes out of the theory.[4]

CONCLUSIONS

In this thesis we have investigated several problems relating to spatial dispersion effects in crystal optics. In Chapter II we studied the pulse propagation in strong spatially dispersive media, that is, near a resonance of excitonic-polariton media. Expressions were obtained for energy velocity V_E for Upper and Lower polariton branches respectively. A numerical study was carried out using material parameters of GaAs. Comparison between calculated energy, group and signal velocities was examined in the vicinity of the exciton resonance. The energy propagation studies were made assuming that propagating modes are plane wave.

Next Gaussian pulse propagation studies in excitonic-polariton media were carried out in various frequency regions, for $\Gamma\tau \gg 1$, $\Gamma\tau = 1$ and $\Gamma\tau \ll 1$ cases. For $\Gamma\tau \gg 1$ and $\Gamma\tau = 1$ cases, peak of the pulse propagates with group velocity and pulse width shows very little variation.

Various problems related to weak spatial dispersion i.e. gyrotropy were investigated. First wave propagation in bounded gyrotropic medium was studied near resonance in Chapter III. The constitutive relation in the optically active medium was written down in an inverse dielectric function framework. The additional boundary condition (ABC) required to solve the reflection, refraction problem was obtained.

Dispersion relation and mode structure were obtained and the reflectivity problem from half space was solved and numerical results obtained for typical semiconducting medium.

Next in Chapter IV, using the general mode structure in the optically active medium surface wave dispersion relation was written and computed numerically. An Attenuated Total Reflection (ATR) model experiment to detect these surface waves was formulated.

Finally in Chapter V, the Extinction Theorem for model gyrotropic medium with spatial dispersion was investigated. The dispersion relations and the mode structure was obtained. The extinction conditon for electric and magnetic fields were obtained. Additional Boundary Conditions (ABC) required to solve the reflection problem were obtained and, finally, reflection coefficient, $R(\omega)$ was obtained.

Thus in summary we have examined in this thesis various wave vector dependent optical phenomenon, both corresponding to strong and weak spatial dispersion in bounded material media.

BIBLIOGRAPHY

1. V. M. Agranovich and V. L. Ginzburg, Spatial Dispersion in Crystal Optics and the Theory of Excitons, (Interscience, London, 1966).

2. S. I. Pekar, Zh. Eksp. Teor. Fiz. 33, 1022 (1957) [Sov. Phys. JETP 6 785 (1958)].

3. J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963).

4. J. L. Birman and J. J. Sein, Phys. Rev. B6, 2482 (1972); J. J. Sein Ph.D Thesis (New York University, 1969).

5. R. Zeyher, J. L. Birman and W. Brenig, Phys. Rev. B6, 4613 (1972).

6. G. S. Agarwal, D. N. Pattanayak and E. Wolf, Phys. Rev. Lett. 27, 1022 (1971); Phys. Rev. B10, 1477 (1974).

7. A. A. Maradudin and D. L. Mills, Phys Rev. B7, 2787 (1973).

8. M. J. Frankel and J. L. Birman, Phys Rev. B13, 2587 (1976), Phys. Rev. A15, 2000 (1977).

9. V. V. Hyzhnyakov, A. A. Maradudin and D. L. Mills, Phys. Rev. B11, 3149 (1975).

10. C. S. Ting, M. J. Frankel and J. L. Birman, Solid State Comm. 17, 1285 (1975).

11. M. Born and E. Wolf, Principles of Optics, (Oxford: Pergamon Press, 1965), (a) Sec. 1.3 (b) Sec. 2.4 (c) Sec. 8.3.1.

12. L. Brillouin, Wave Propagation and Group Velocity, (Academic Press, New York, 1960).

13. R. Loudon, J. Physics A3, 233 (1970).

14. R. G. Ulbrich and G. W. Fehrenbach, Phys. Rev. Lett. 43, 963 (1979).

15. Y. Masumoto, Y. Unuma, Y. Tanaka and S. Shionoya, J. Phys. Soc. Japan 47, 1844 (1979).

16. T. Itoh, P. Lavallard, J. Reydellet and C. Benoit a la Guillaume, Sol. State Comm. 37, 925 (1981).

17. Y. Sewaga, Y. Aoyagi, T. Baba and S. Namba, J. Phys. Soc. Japan 49, Suppl. A, 389 (1980).

18. A. Puri and J. L. Birman, Phys. Rev. Lett. 47, 173 (1981).

19. C. G. B. Garrett and D. E. McCumber, Phys. Rev. A1, 305 (1970).

20. A. Puri and J. L. Birman (Submitted to Phys. Rev. A).

21. L. D. Landau and E. M. Lifshitz, Electrodynamics of Continuous Media, (Pergamon Press, New York, 1960), p. 337.

22. D. N. Pattanayak, A. Puri and J. L. Birman, Phys. Rev. B24, 4279 (1981).

23. D. N. Pattanayak and J. L. Birman, Phys. Rev. B24, 1471 (1981).

24. A. Puri, D. N. Pattanayak and J. L. Birman, JOSA 72, 938 (1982).

25. P. P. Ewald, Ann. Phys. 49, 1 (1915).

26. C. W. Oseen, Ann. Phys. 48, 1 (1915).

27. A. Puri and J. L. Birman, Optics Comm. 37, 81 (1981).

28(a) S. Chu and S. Wong, Phys Rev. Lett. 48, 738 (1982).
(b) A. Katz and R. R. Alfano, Submitted to Phys. Rev. Lett. (Comments); Further work in progress.

29. R. Maddox and D. L. Mills, Phys. Rev. B11, 2229 (1975).

30. M. A. Bishop and A. A. Maradudin, Phys. Rev. B14, 3384 (1976).

31. V. L. Ginzburg, Soviet Physics JETP 7, p. 1096 (1958).

32. The Scientific Papers of J. Williard Gibbs, Vol. II, p. 205.

33(a) Yu. A. Tsvirko, Soviet Physics-Solid State 4, 622 (1962);
(b) Soviet Physics-Solid State 5, 1089 (1963);
(c) Ref. 1 and 31.

34. See p. 208 of Ref. 1.

35. See p. 1097 of Ref. 33(b).

36. Inverse dielectric function of type Eq. (3.17a) was proposed by Ginzburg in Ref. 31. For details see Sections 5 and 10 of Ref. 1.

37. J. Van Kranendonk and J. E. Sipes, in Progress in Optics, Vol. XV, editor E. Wolf (North Holland Publishing Co., 1977), pp. 245-350.

38. L. P. Gakhov, Boundary Value Problems, (Addison Wesley, 1966), Chap. 39, 38.3 and 38.4.

39. See Page 211 of Ref. 1.

40. G. S. Agarwal, A. J. Devaney and D. N. Pattanayak, Journal of Math. Phys. 14, 906 (1973).

41. E. L. Ivchenko and A. V. Selkin, Zh. Eksp. Teor. Fiz. 76, 1837 (1979).

42. Y. Y. Teng and E. A. Stern, Phys. Rev. Lett. 19, 511 (1967).

43. C. J. Powel and J. B. Swan, Phys. Rev. 118, 640 (1960).

44. A. S. Barker, Phys. Rev. Lett. 28, 892 (1972).

45. N. Marschall and B. Fischer, Phys. Rev. Lett. 28, 811 (1972).

46. G. S. Agarwal, Optics Comm. 6, 221 (1972).

47. M. F. Bishop and A. A. Maradudin, Solid State Comm. 23, 507 (1977).

48. H. J. Flage and A. Otto, Phys. Stat. Sol. (b) 56, 523 (1973).

49(a) A. Wierzbicki, Bul. Acad. Polonaise des Sciences 9, 833 (1961); Acta Phys. Pol. 21, 557 (1962), ibid 21, 575 (1962).

(b) N. Bloembergen and P. S. Pershan, Phys. Rev. 127, 206 (1962).

(c) B. A. Sotskii, Opt. Specrro. 14, 57 (1963).

(d) R. K. Bullough, J. Phys. A. (Proc, Phys. Soc.) Ser. 2, 1, 409 (1968).

(e) J. J. Sein, Ref. 4 above and Opt. Comm. 2, 170 (1970).

(f) E. Lalor, Opt. Comm. 1, 50 (1969).
(g) E. Lalor and E. Wolf, Phys. Rev. Lett. 26, 1274 (1971).
(h) T. Suzuki, J. Opt. Soc. Amer. 61, 1029 (1971).
(i) G. S. Agarwal, D. N. Pattanayak and E. Wolf, Opt. Comm. 4, 260 (1971).
(j) E. Lalor and E. Wolf, J. Opt. Soc. Amer. 62, 1165 (1972).
(k) D. N. Pattanayak and E. Wolf, Opt. Comm. 6, 217 (1972).

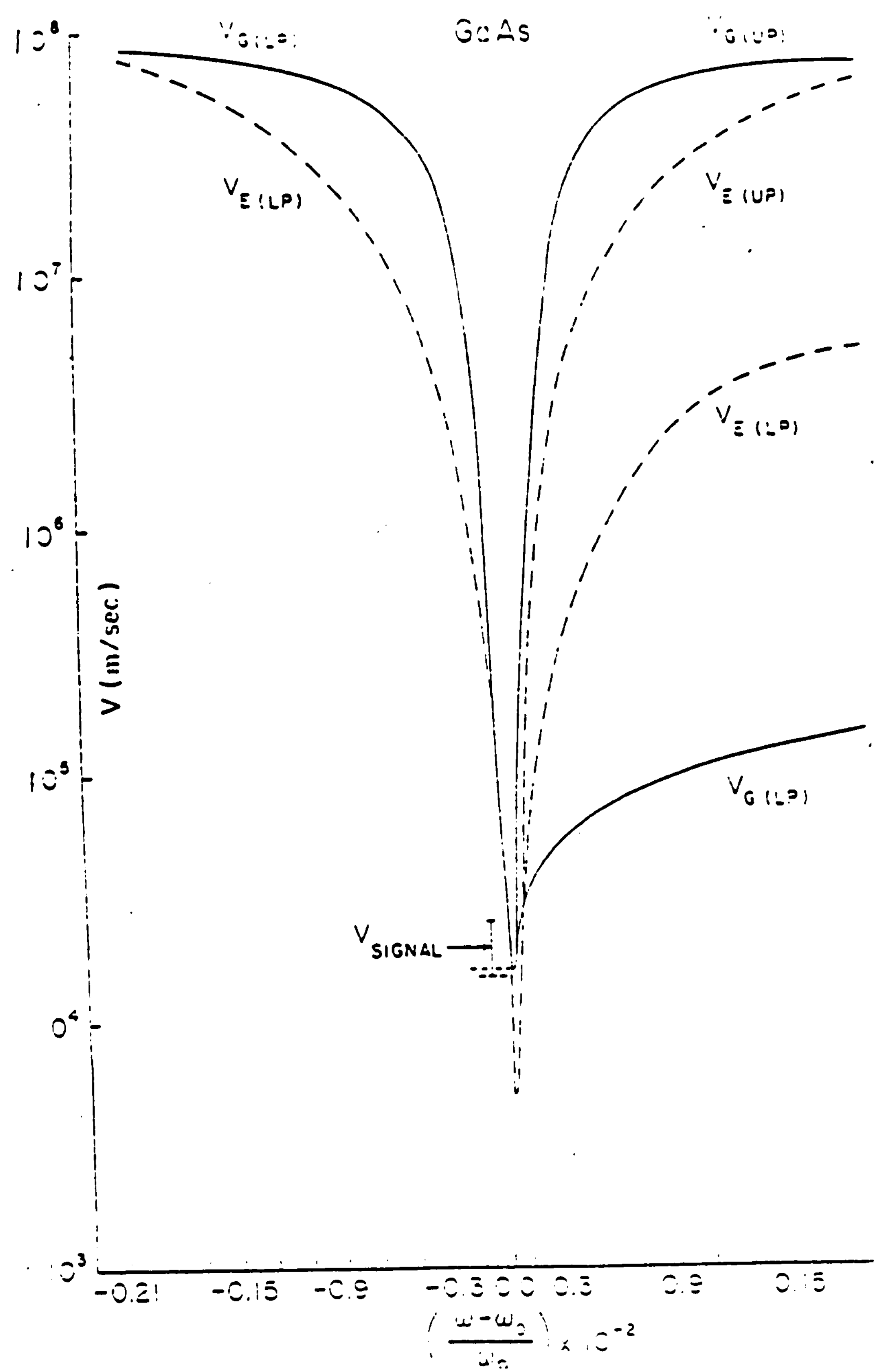
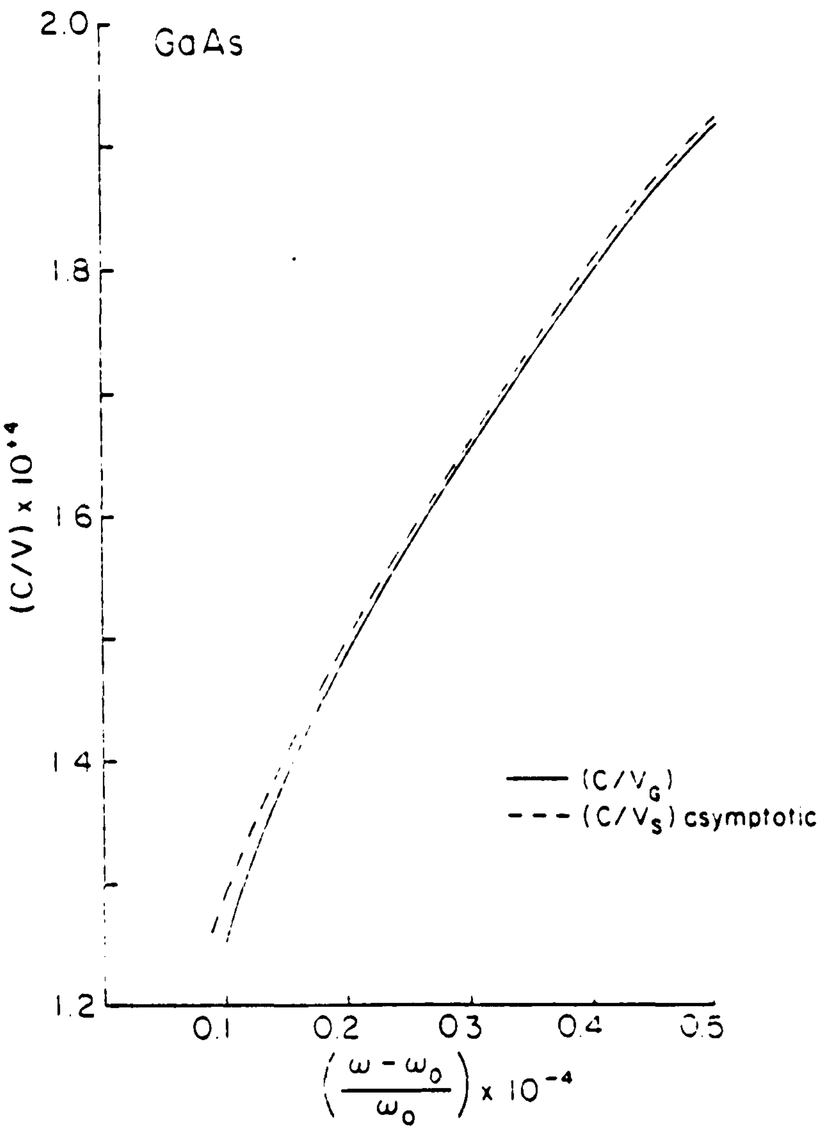

GaAs
$V_{G(LP)}$
$V_{G(UP)}$
$V_{E(LP)}$
$V_{E(UP)}$
$V_{E(LP)}$
$V_{G(LP)}$
V_{SIGNAL}
V (m/sec)
10^8
10^7
10^6
10^5
10^4
10^3
-0.21
-0.15
-0.9
-0.3
0.0
0.3
0.9
0.15
$\left(\frac{\omega-\omega_0}{\omega_0}\right) \times 10^{-2}$

Fig. 1



GaAs
2.0
1.8
1.6
1.4
1.2
$(C/V) \times 10^{4}$
0.1
0.2
0.3
0.4
0.5
$\left(\frac{\omega - \omega_0}{\omega_0}\right) \times 10^{-4}$
(C/V_G)
(C/V_S) asymptotic

Fig. 2

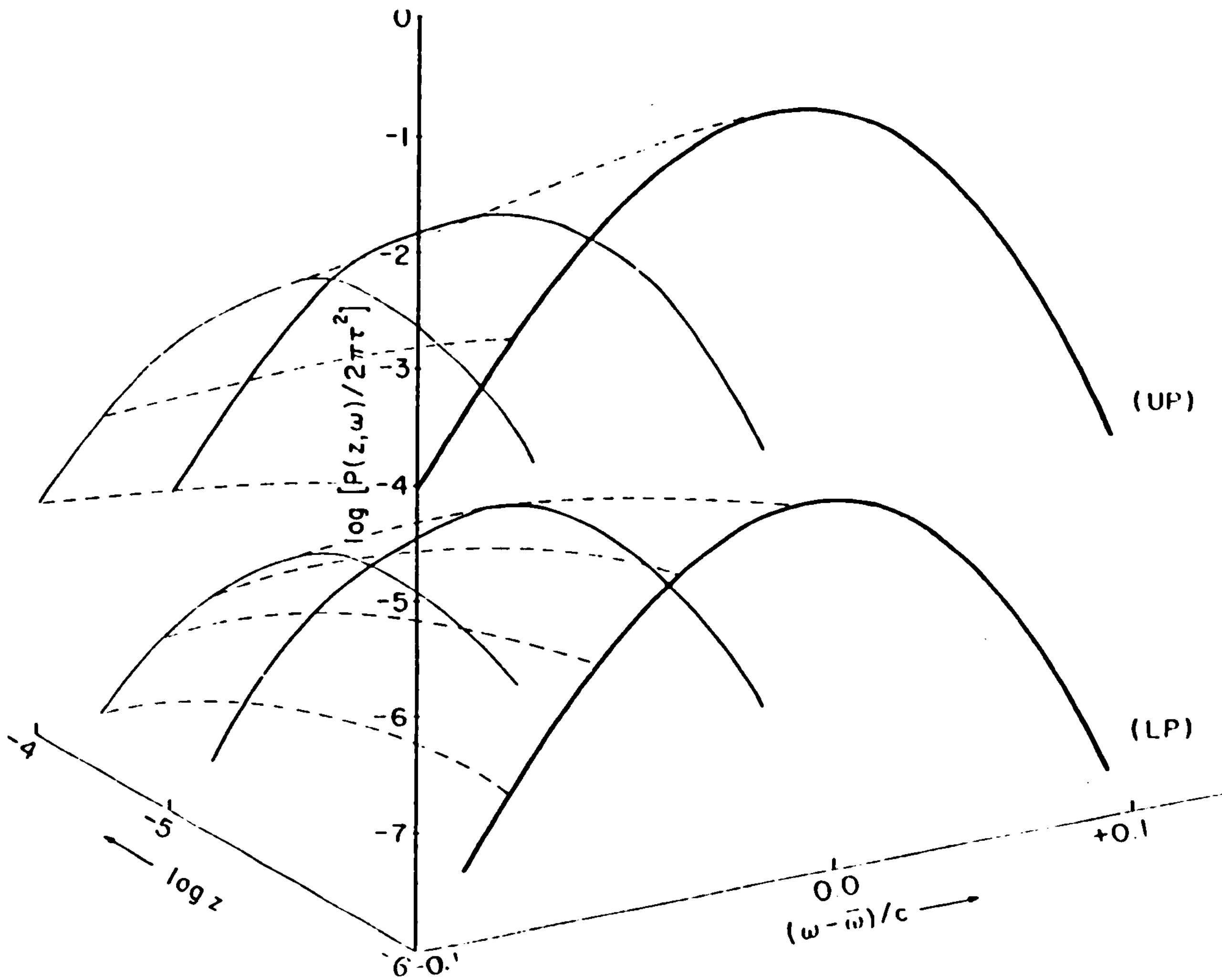


Fig. 3

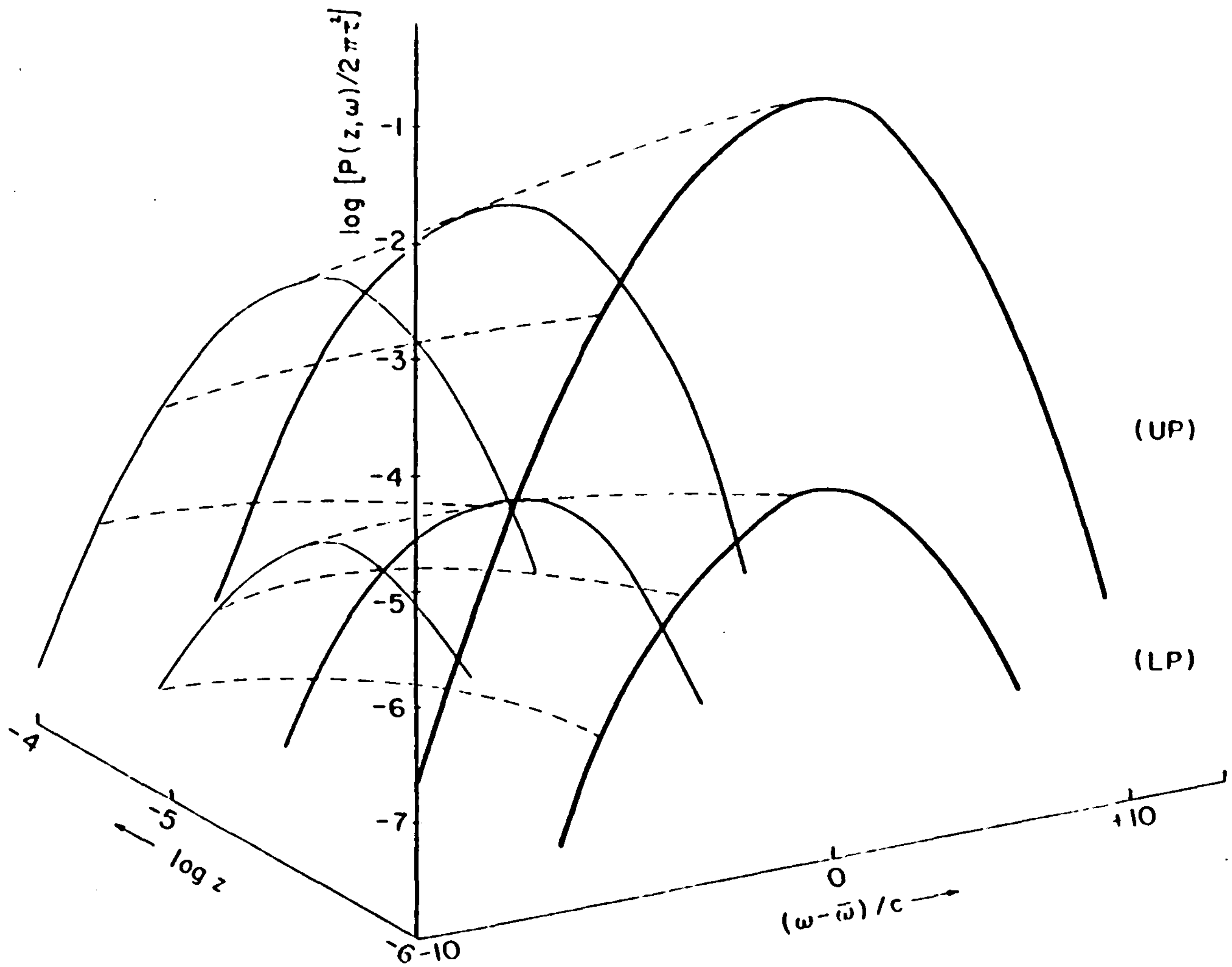
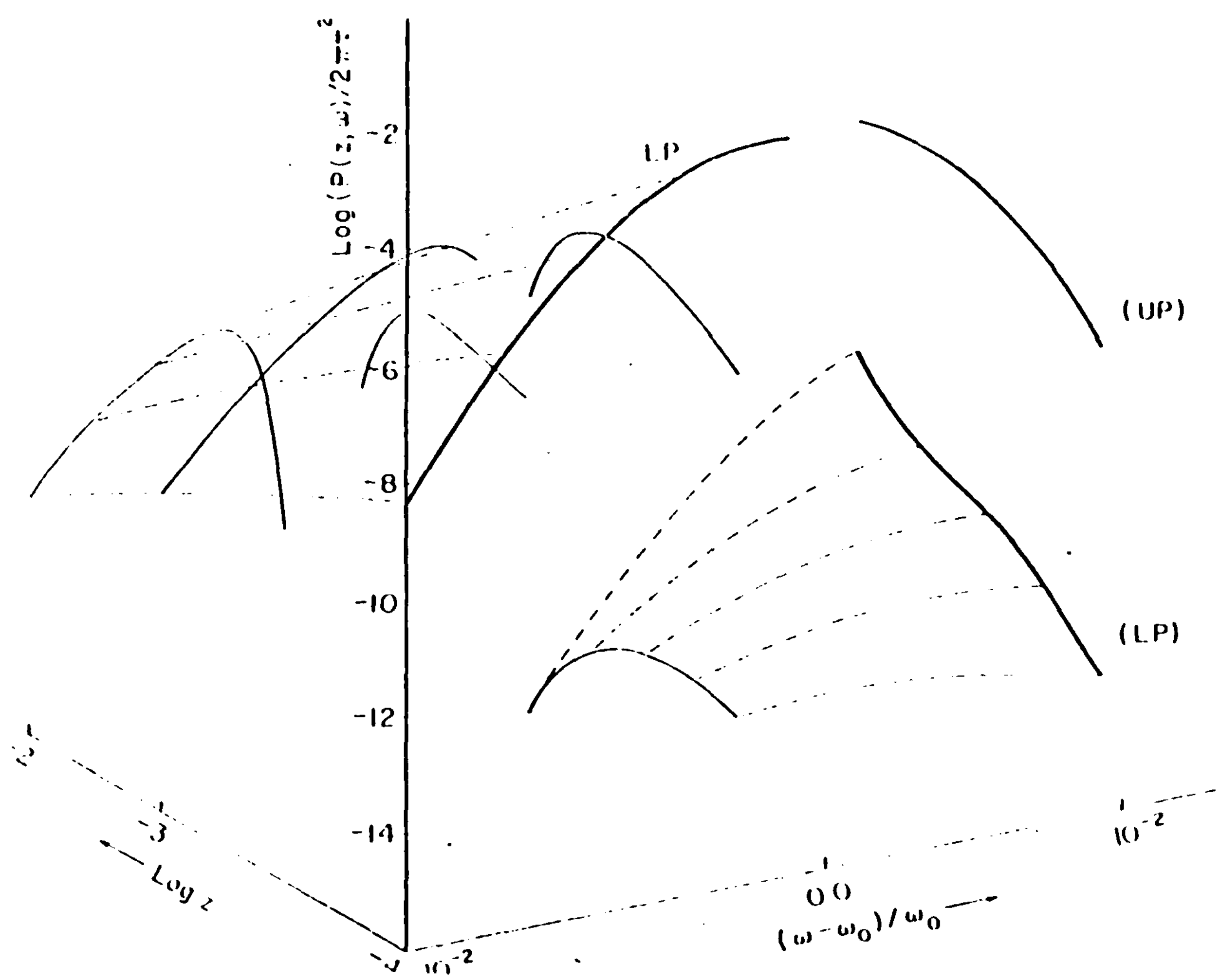
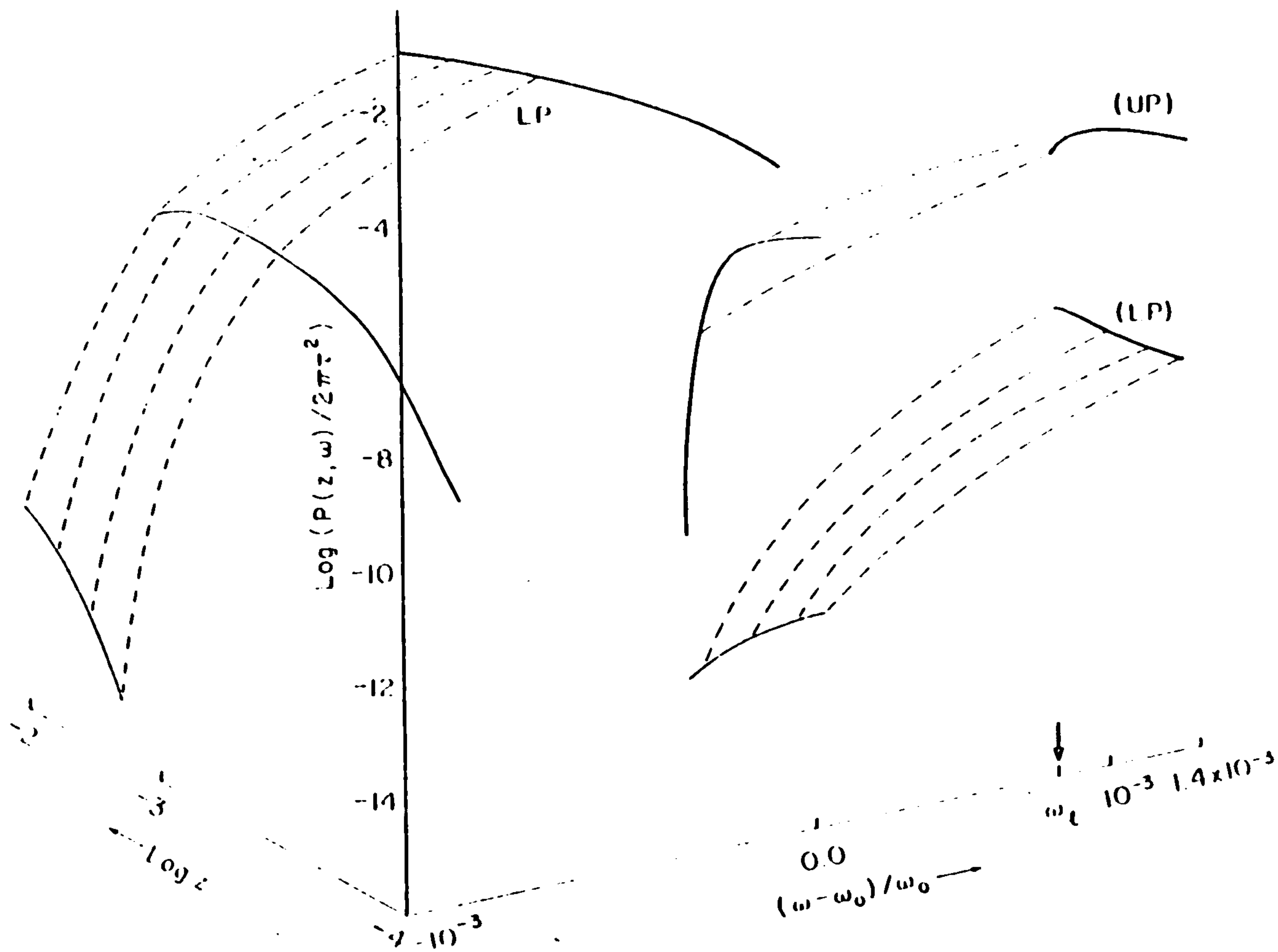


Fig. 4



Log (P(z, ω)/2πτ²
-2
-4
-6
-8
-10
-12
-14
LP
(UP)
(LP)
-3
Log z
0.0
10^{-2}
$(\omega-\omega_0)/\omega_0$

Fig. 5



-2
LP
(UP)
-4
(LP)
Log (P(z, ω)/2πτ²)
-8
-10
-12
-14
Log z
0.0
$(\omega-\omega_0)/\omega_0$
ω_c
10^{-3}
1.4×10^{-3}

Fig. 6

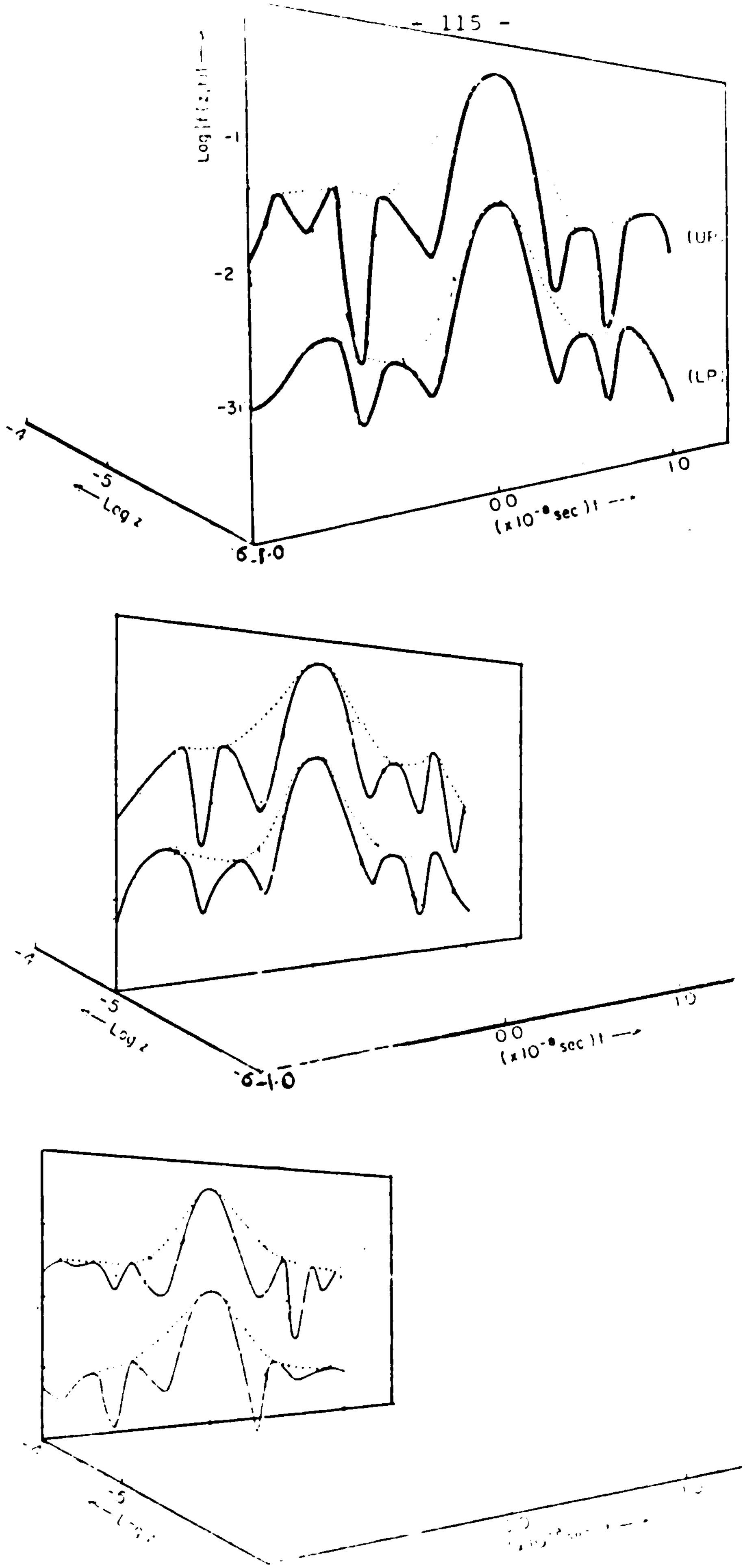


Fig. 7

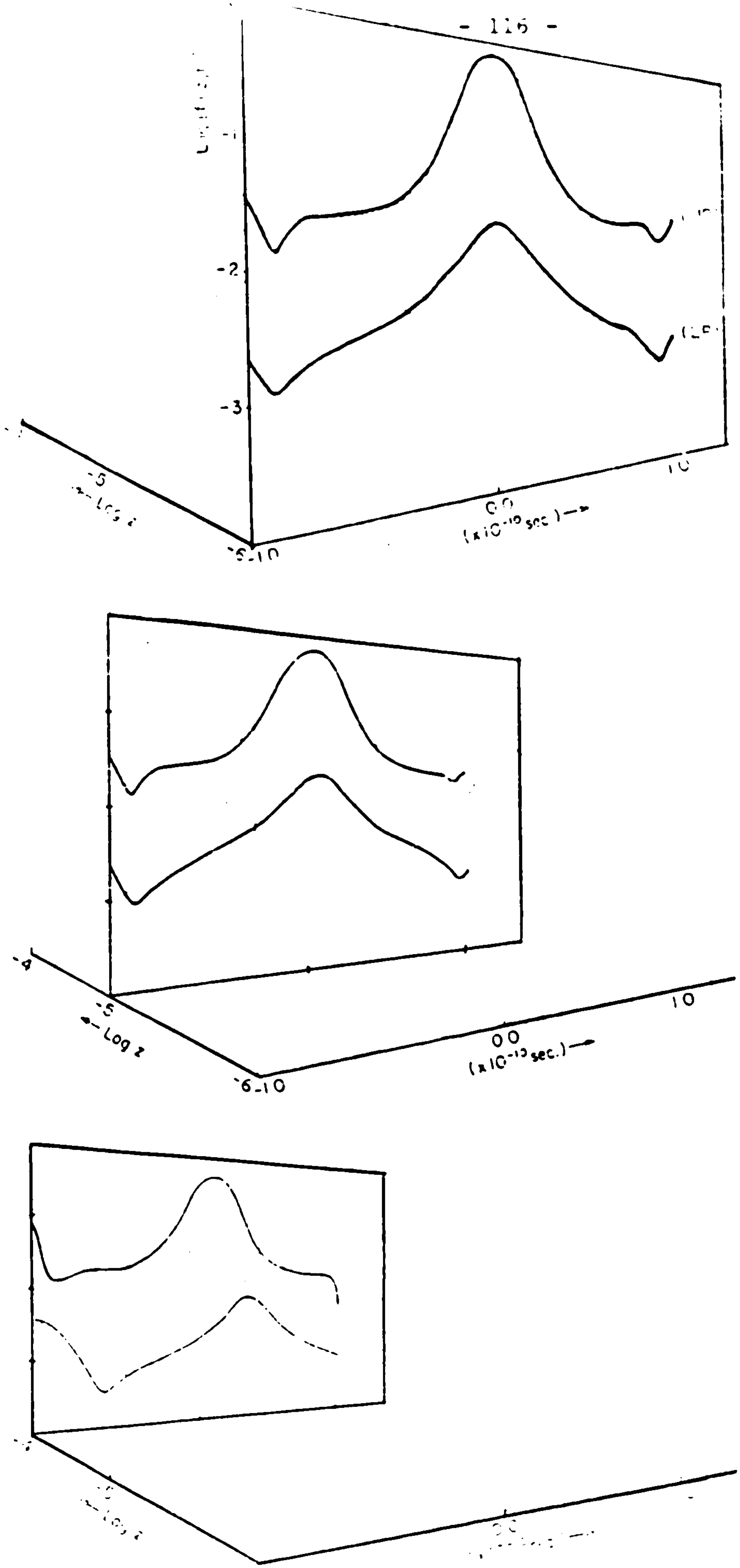


Fig. 8

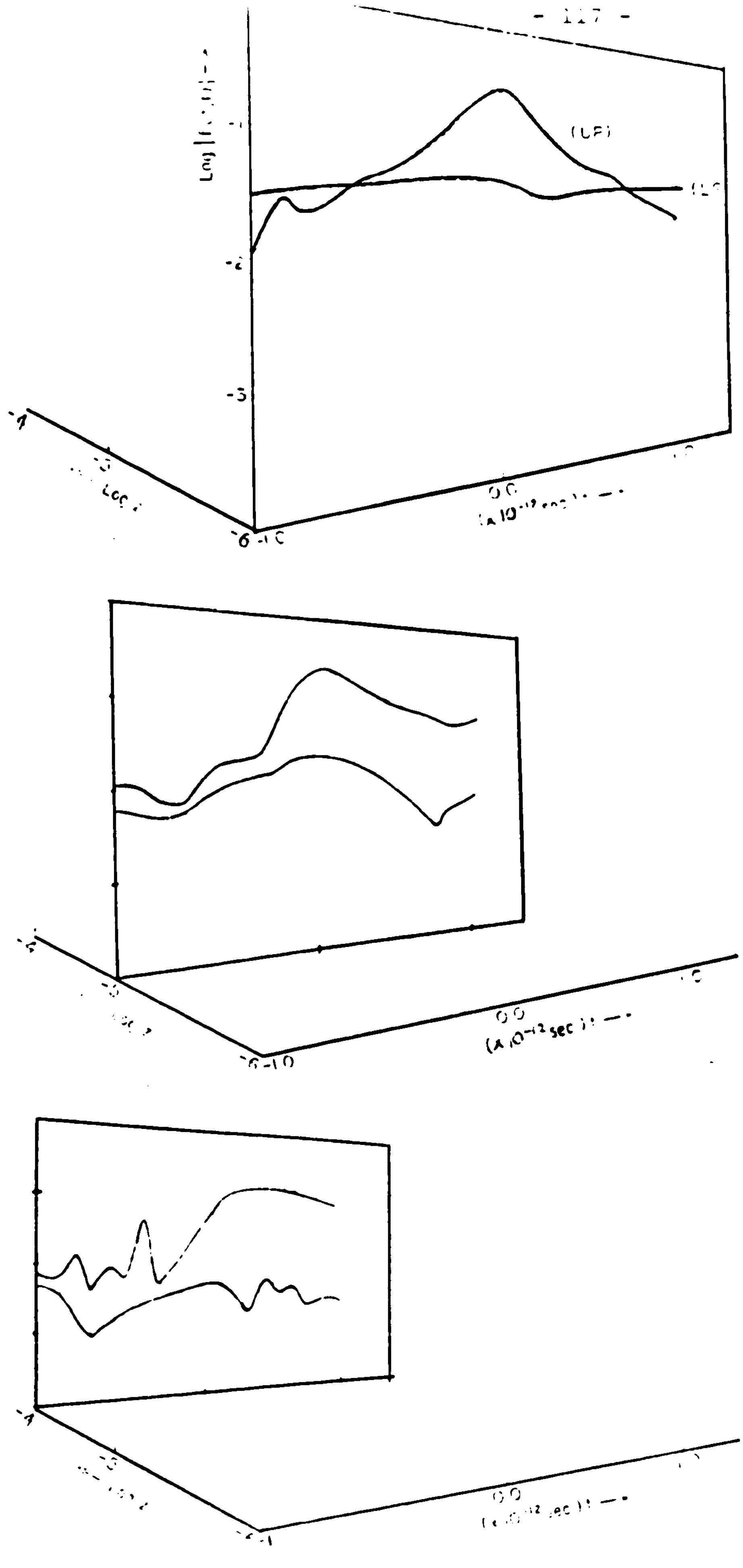


Fig. 9

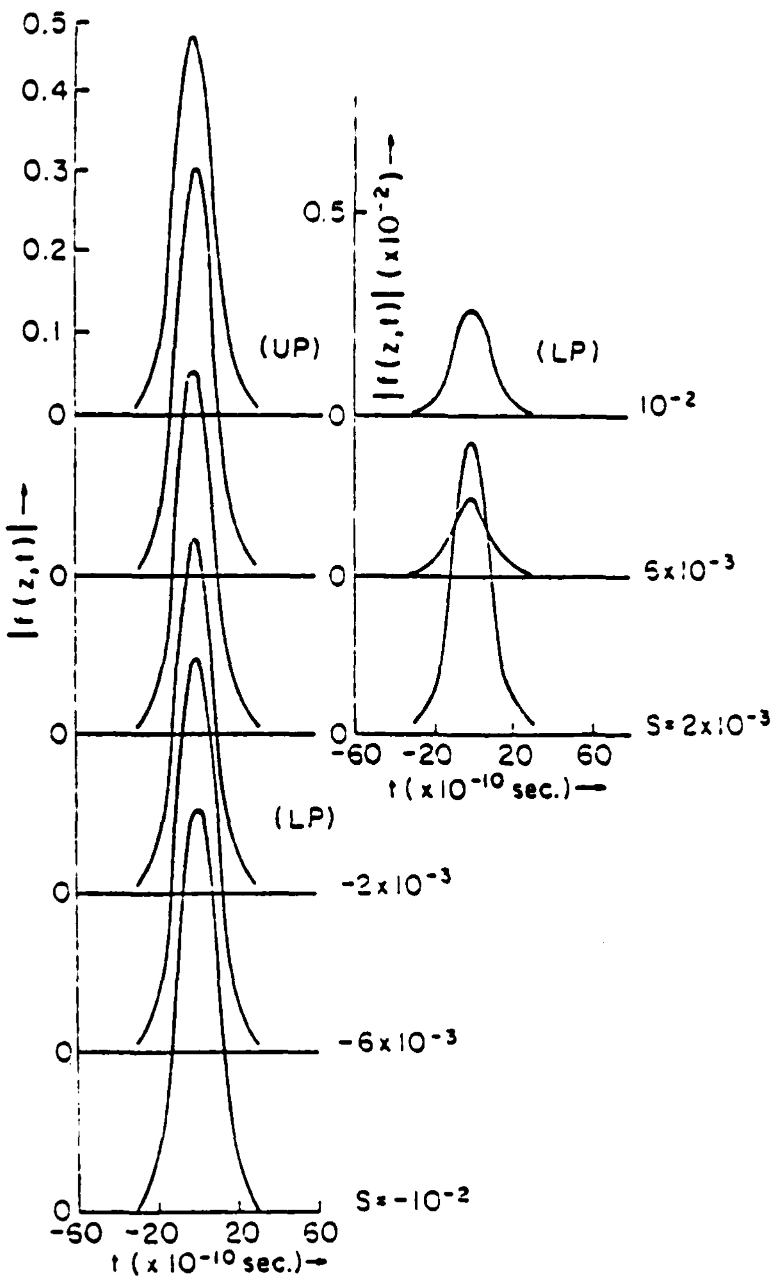

0.5
0.4
0.3
0.2
0.1
0
(UP)
(LP)
|f(z,t)|
-2x10^-3
-6x10^-3
S=-10^-2
-60 -20 20 60
t (x10^-10 sec.)
|f(z,t)| (x10^-2)
0.5
(LP)
10^-2
5x10^-3
S=2x10^-3
-60 -20 20 60
t (x10^-10 sec.)

Fig. 10

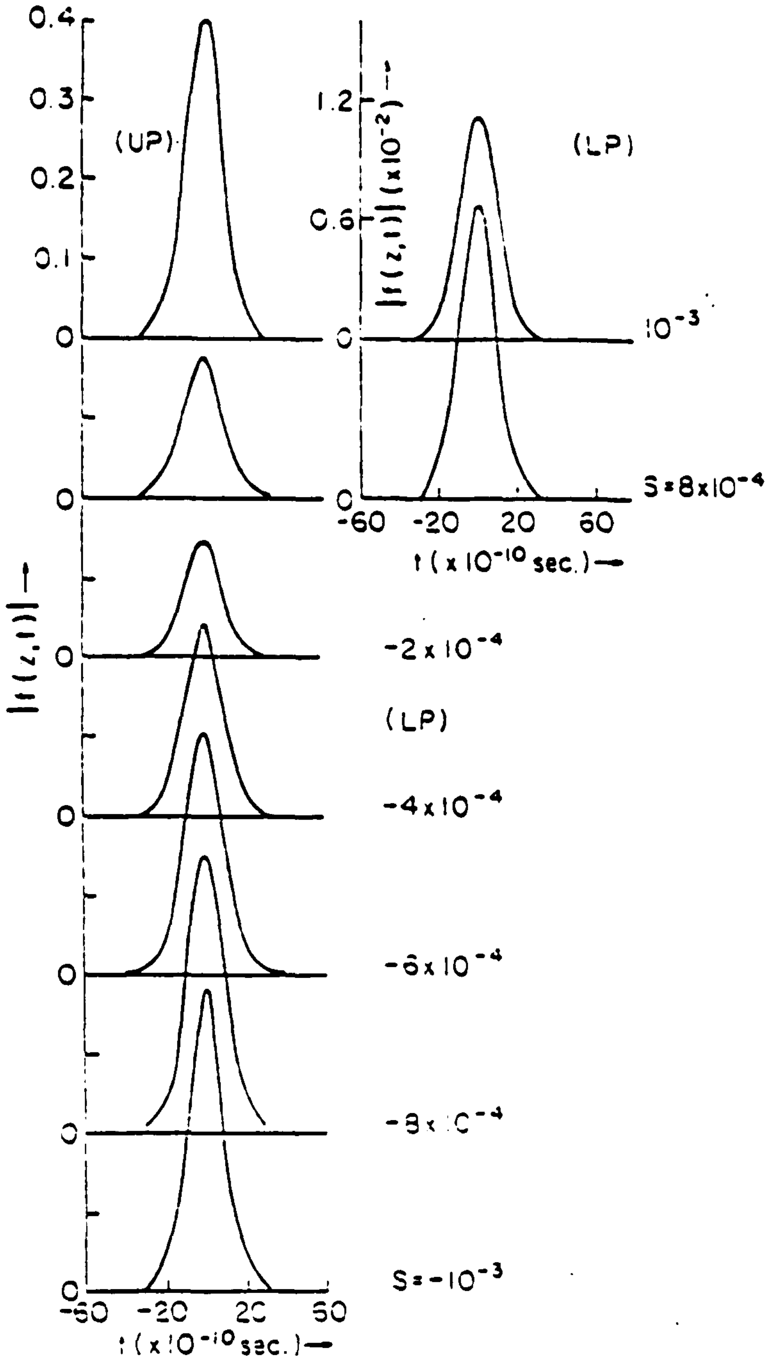

(UP)
(LP)
(LP)
0.4
0.3
0.2
0.1
0
1.2
0.6
$|f(z,t)|$
$|f(z,t)|\ (\times 10^{-2})$
10^{-3}
$S = 8\times 10^{-4}$
-2×10^{-4}
-4×10^{-4}
-6×10^{-4}
-8×10^{-4}
$S = -10^{-3}$
-60 -20 20 60
$t\ (\times 10^{-10}$ sec.)

Fig. 11

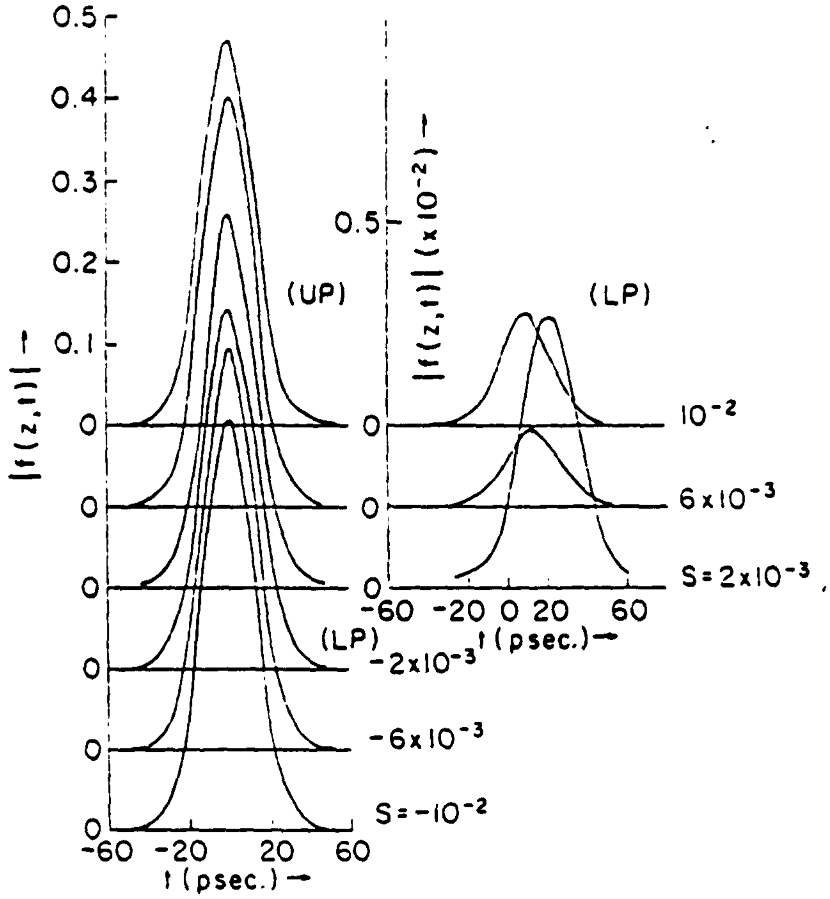

0.5
0.4
0.3
0.2
0.1
0
|f(z,t)| →
(UP)
(LP)
-60 -20 20 60
t (psec.) →
-2x10⁻³
-6x10⁻³
S=-10⁻²
0.5
|f(z,t)| (x10⁻²) →
(LP)
10⁻²
6x10⁻³
S=2x10⁻³
-60 -20 0 20 60
t (psec.) →

Fig. 12

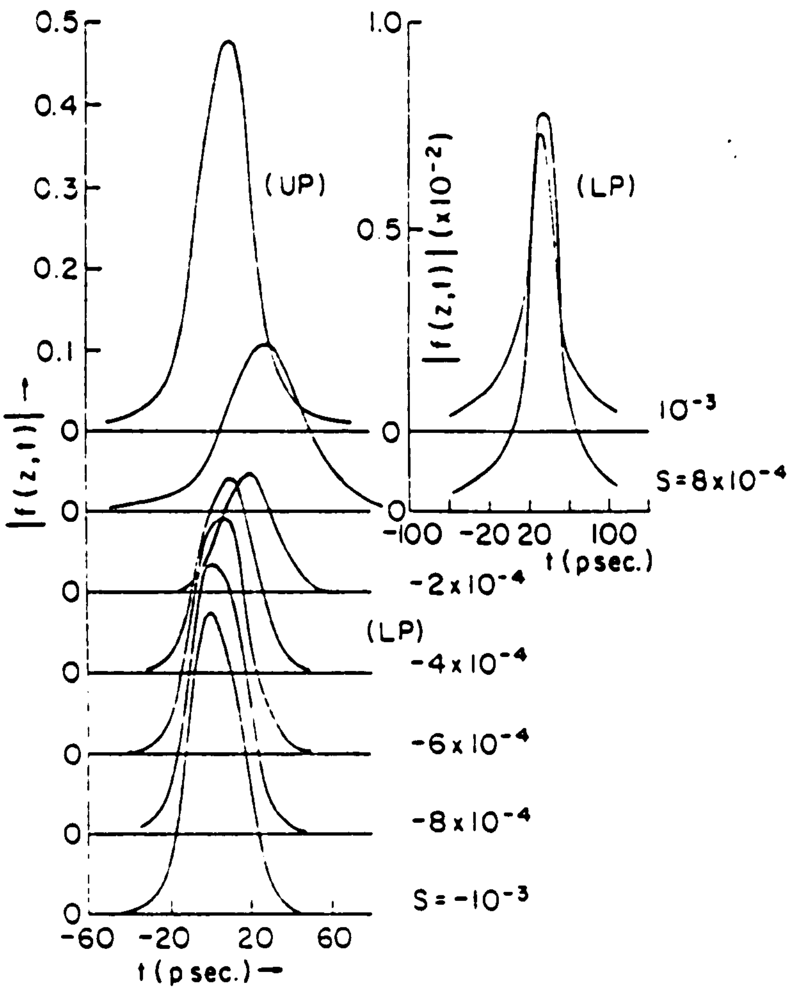

0.5
0.4
0.3
0.2
0.1
0
(UP)
|f(z,t)| →
1.0
0.5
(LP)
|f(z,t)| (x10⁻²)
10⁻³
S=8x10⁻⁴
-100 -20 20 100
t (psec.)
-2x10⁻⁴
(LP)
-4x10⁻⁴
-6x10⁻⁴
-8x10⁻⁴
S=-10⁻³
-60 -20 20 60
t (psec.) →

Fig. 13

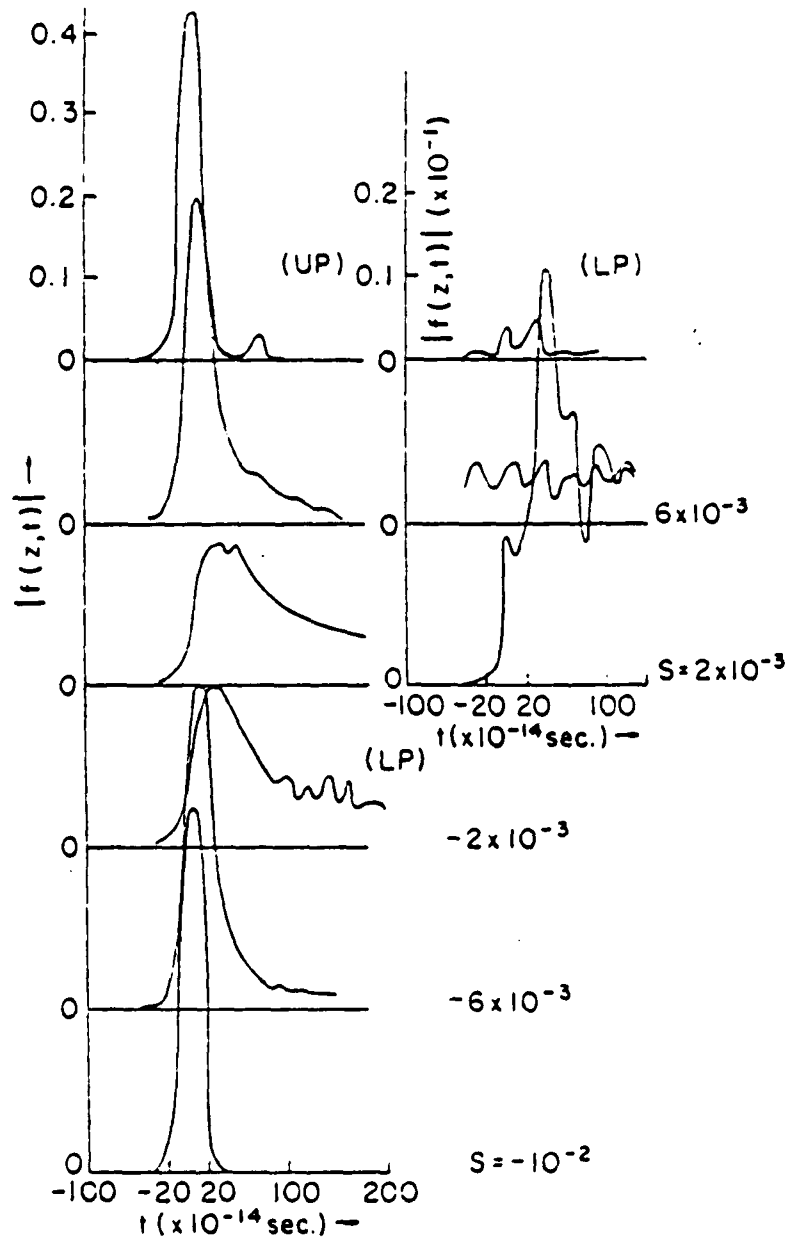

0.4
0.3
0.2
0.1
0
(UP)
|f(z,t)| →
(LP)
0.2
0.1
|f(z,t)| (x10⁻¹)
(LP)
6x10⁻³
S=2x10⁻³
-100 -20 20 100
t(x10⁻¹⁴sec.) →
-2x10⁻³
-6x10⁻³
S=-10⁻²
-100 -20 20 100 200
t(x10⁻¹⁴sec.) →

Fig. 14

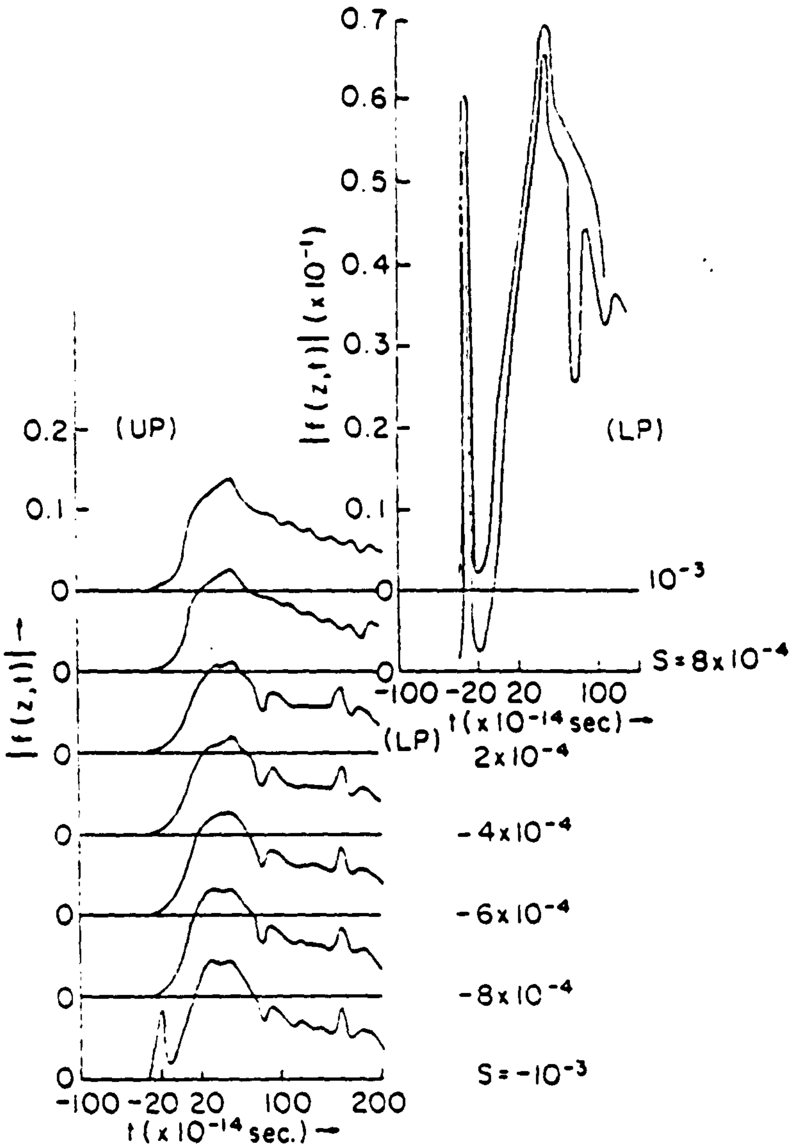
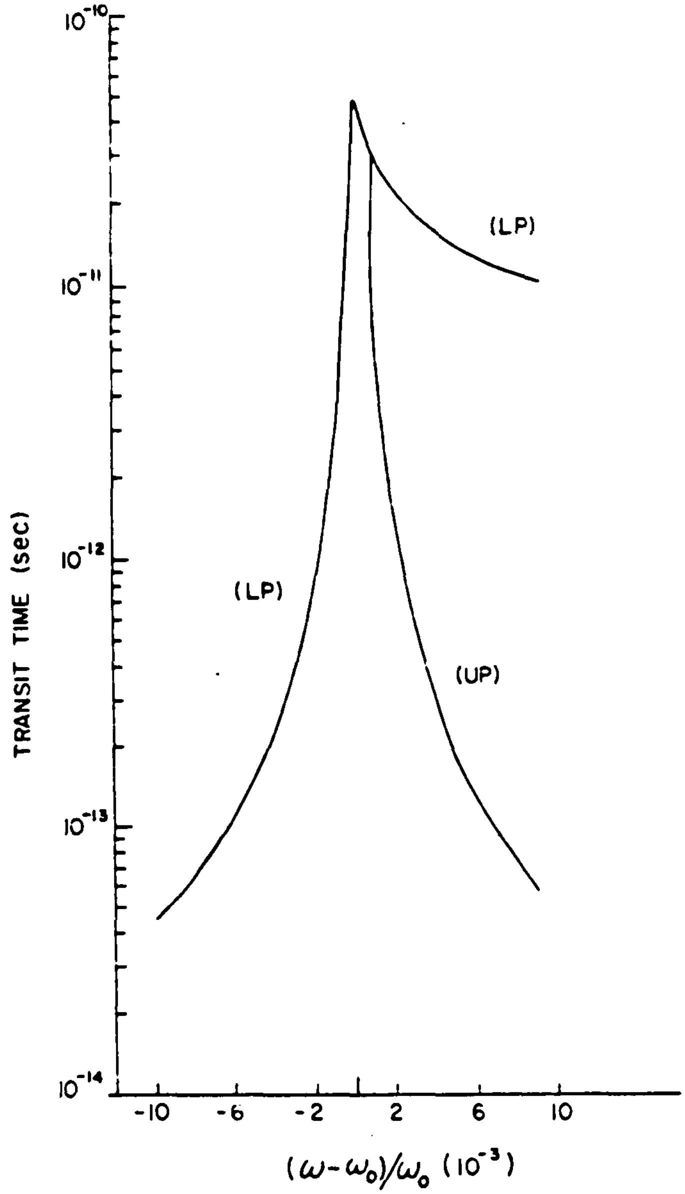

0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
|f(z,t)| (x10^{-1})
(LP)
10^{-3}
S = 8x10^{-4}
-100 -20 20 100
t (x10^{-14} sec) →
(UP)
0.2
0.1
|f(z,t)| →
(LP)
2x10^{-4}
-4x10^{-4}
-6x10^{-4}
-8x10^{-4}
S = -10^{-3}
-100 -20 20 100 200
t (x10^{-14} sec.) →

Fig. 15



10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
TRANSIT TIME (sec)
(LP)
(LP)
(UP)
-10
-6
-2
2
6
10
$(\omega-\omega_0)/\omega_0$ (10^{-3})

Fig. 16

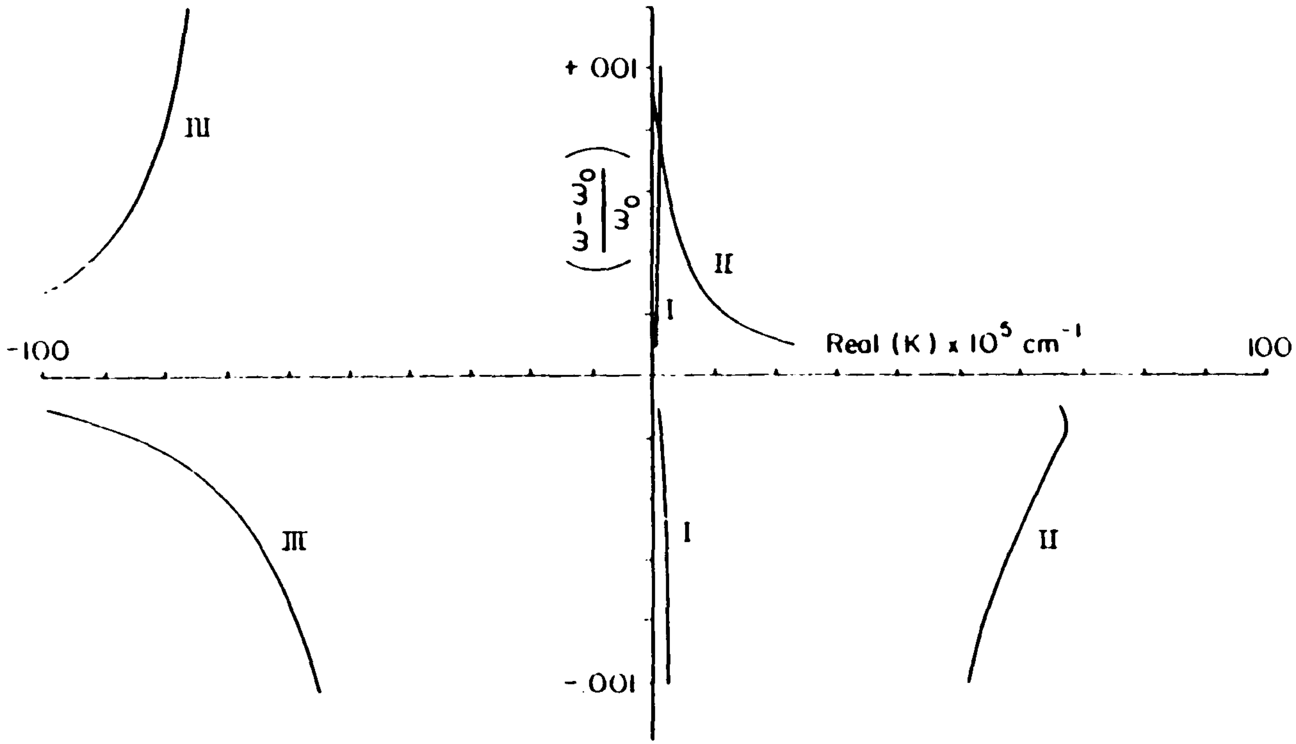

+ .001
- .001
-100
100
Real (K) x 10^5 cm^-1
$\left(\frac{\omega-\omega_0}{\omega_0}\right)$
III
II
I

Fig. 17

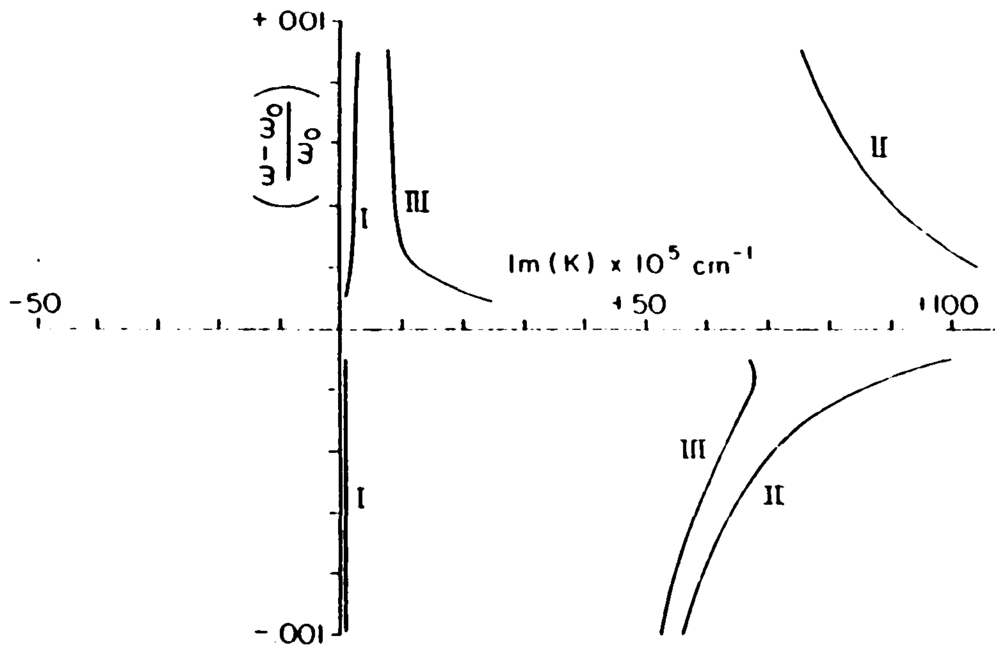

+ 001
- 001
$\left(\frac{\omega-\omega_0}{\omega_0}\right)$
Im (K) x 10^5 cm^{-1}
-50
+50
+100
I
III
II
I
III
II

Fig. 18

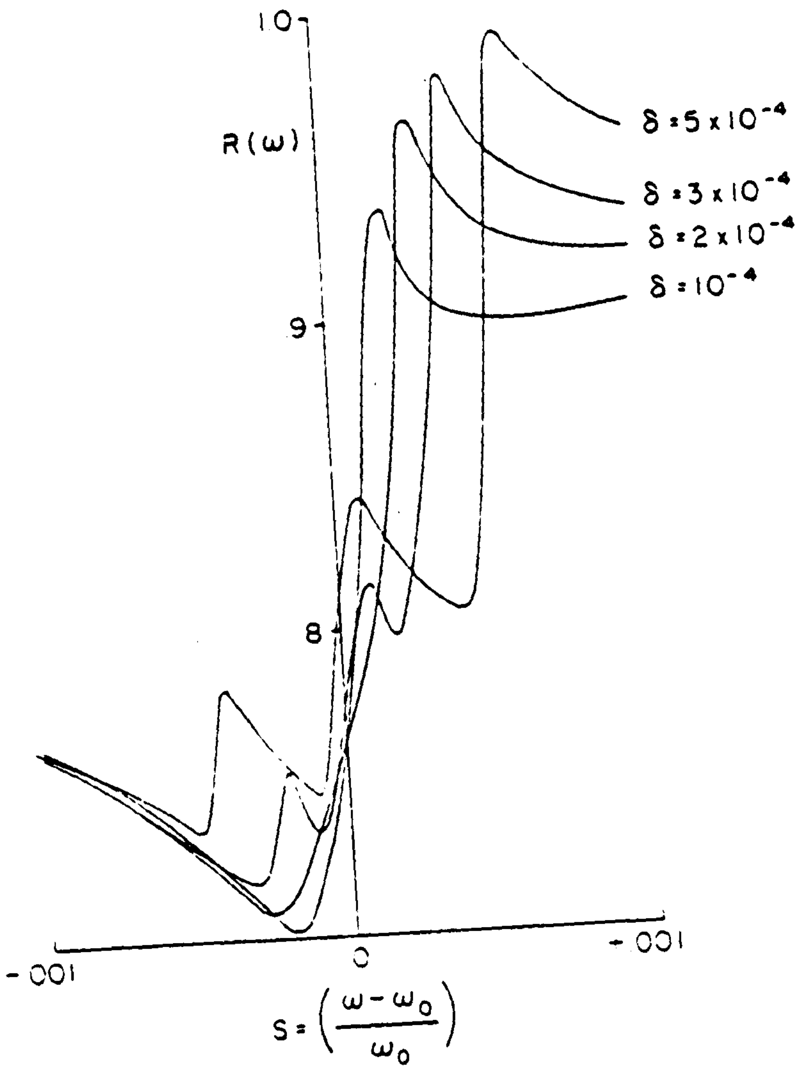


Fig. 19

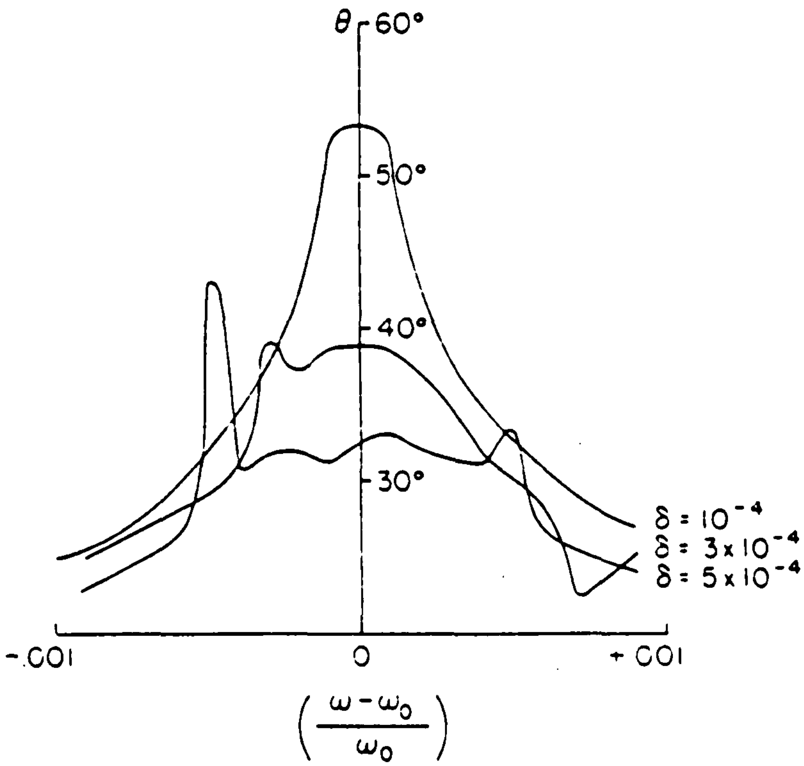

θ
60°
50°
40°
30°
δ = 10⁻⁴
δ = 3 x 10⁻⁴
δ = 5 x 10⁻⁴
-.001
0
+.001
$\left(\frac{\omega-\omega_0}{\omega_0}\right)$

Fig. 20

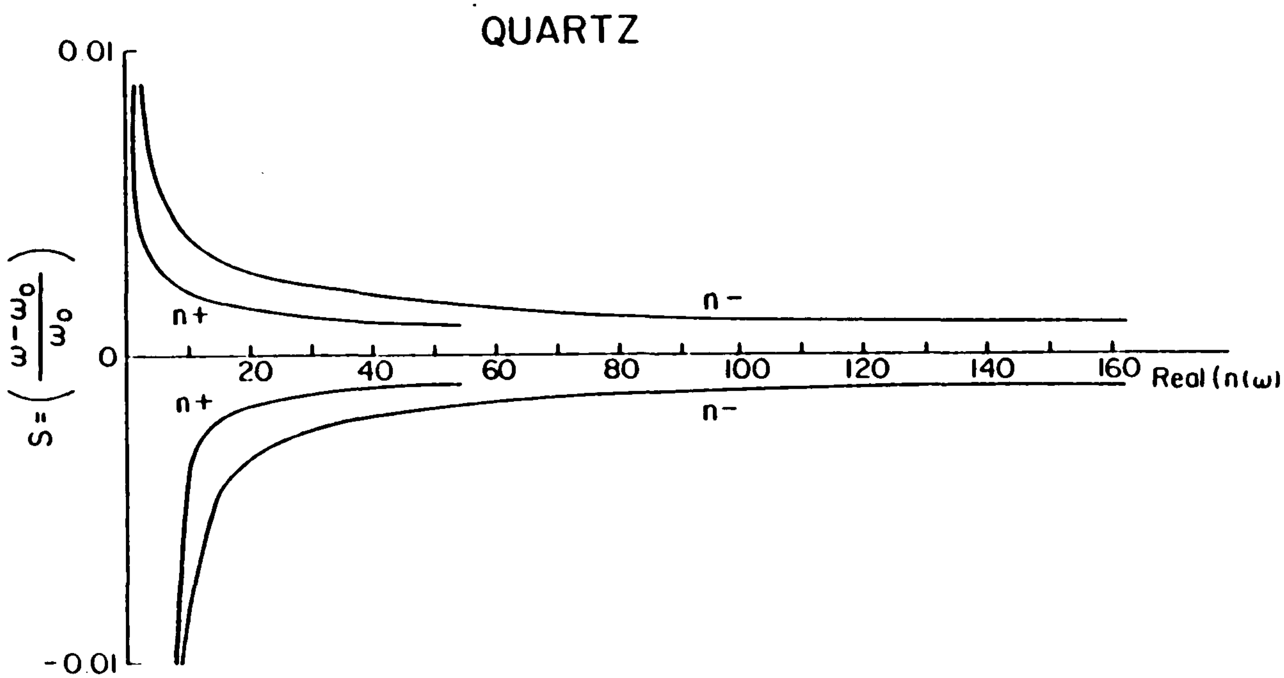

QUARTZ
0.01
0
-0.01
$S = \left(\frac{\omega - \omega_0}{\omega_0}\right)$
n+
n−
n+
n−
20
40
60
80
100
120
140
160
Real (n(ω))

Fig. 21

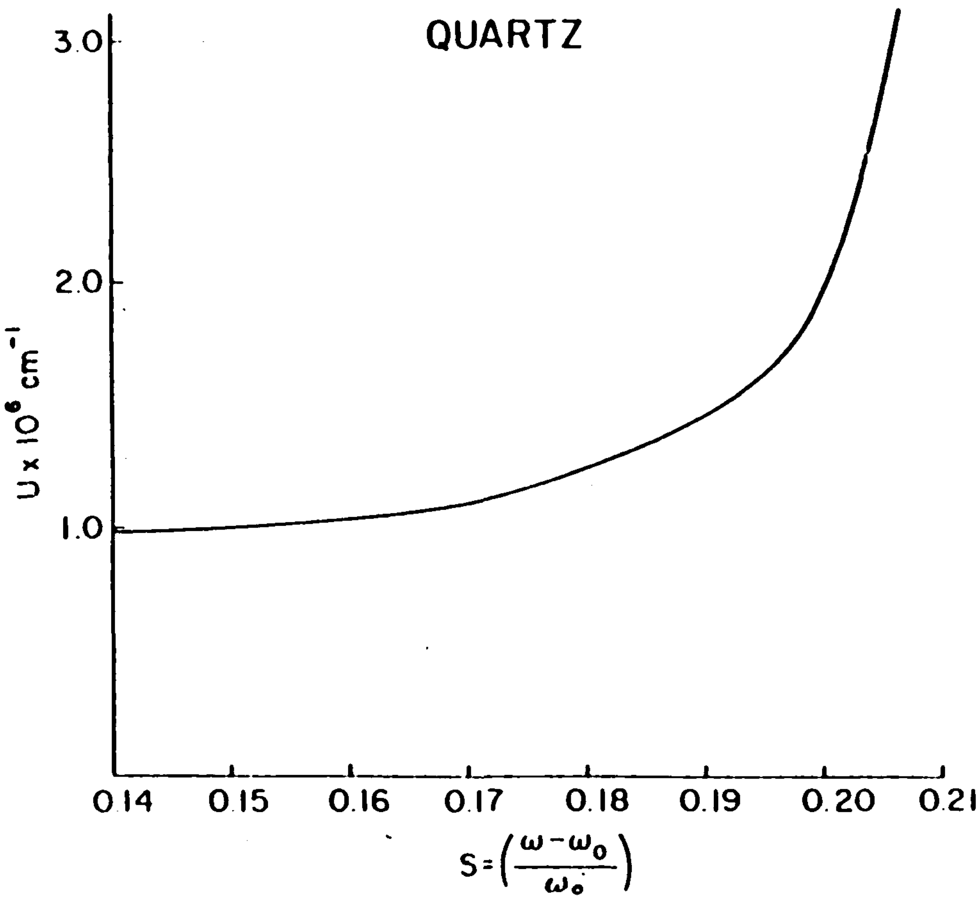

QUARTZ
3.0
2.0
1.0
U x 10⁶ cm⁻¹
0.14
0.15
0.16
0.17
0.18
0.19
0.20
0.21
S = ((ω − ω₀)/ω₀)

Fig. 22

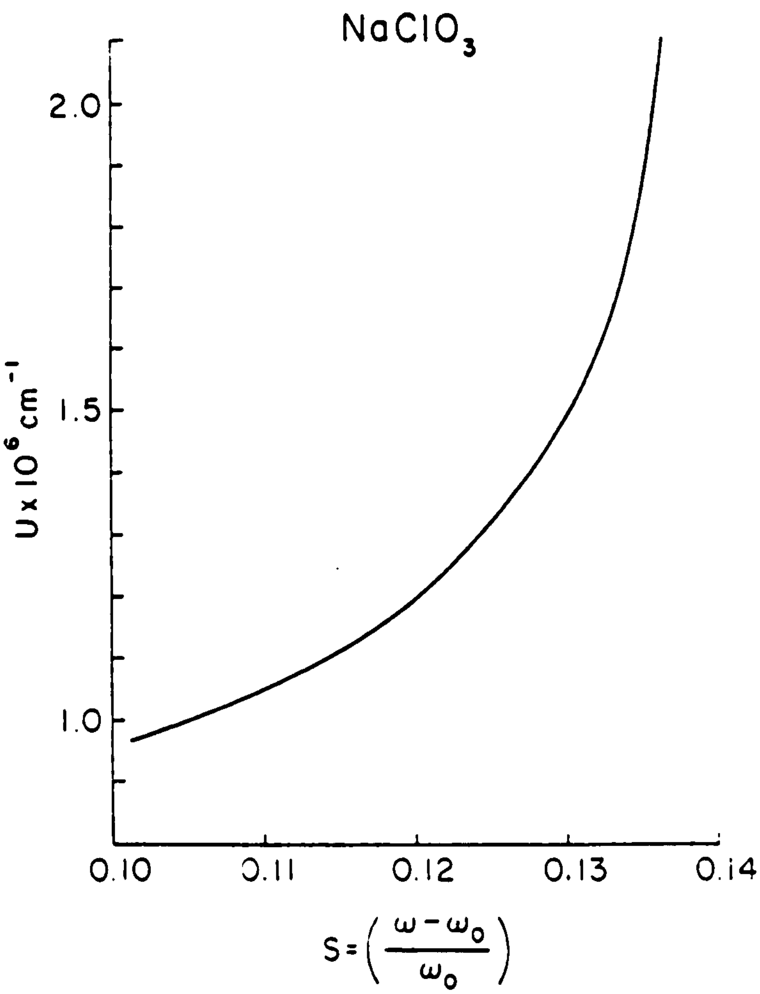

NaClO$_3$
2.0
1.5
1.0
U x 10^6 cm^{-1}
0.10
0.11
0.12
0.13
0.14
S = $\left(\frac{\omega-\omega_0}{\omega_0}\right)$

Fig. 23

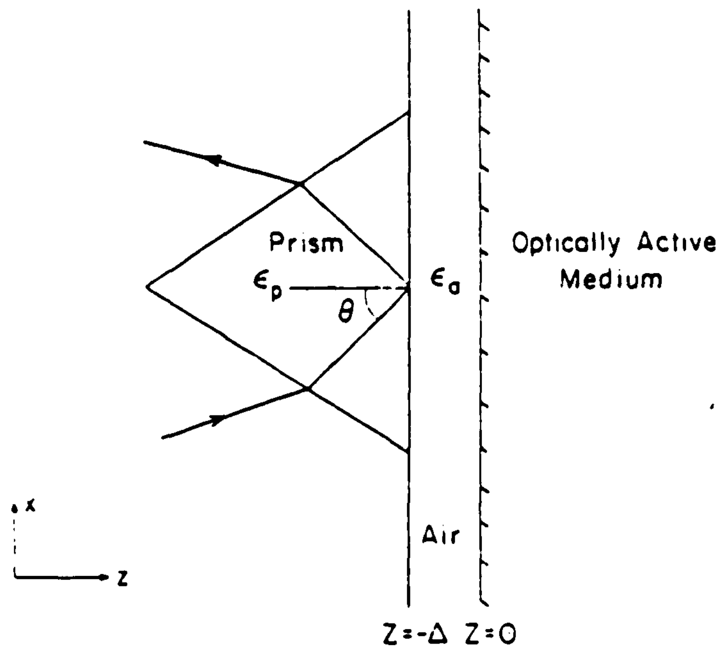

Prism
ϵ_p
θ
ϵ_a
Optically Active Medium
Air
x
z
Z=-Δ Z=0

Fig. 24

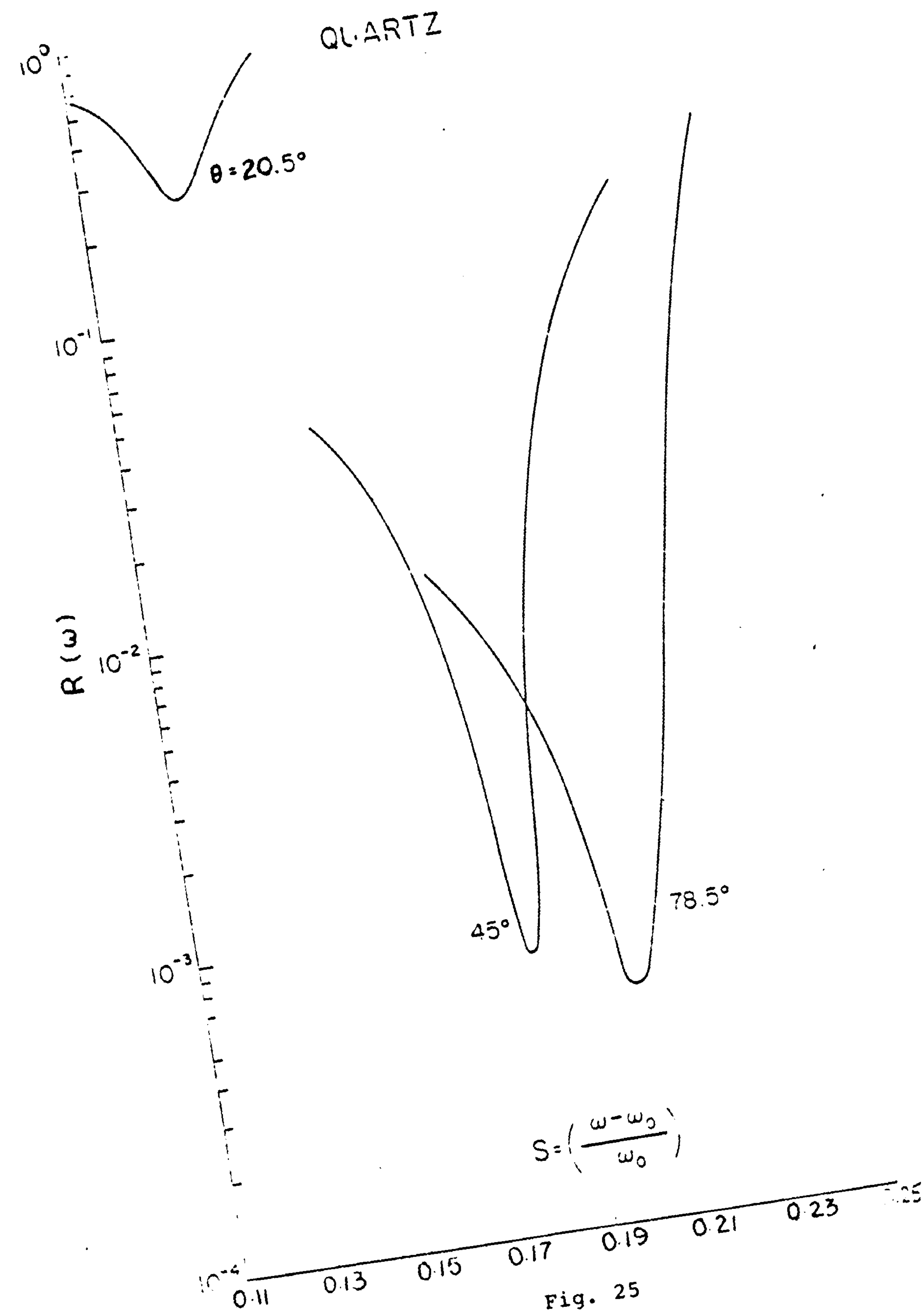

QUARTZ
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
R (ω)
θ = 20.5°
45°
78.5°
$S = \left(\frac{\omega - \omega_0}{\omega_0}\right)$
0.11
0.13
0.15
0.17
0.19
0.21
0.23
0.25

Fig. 25

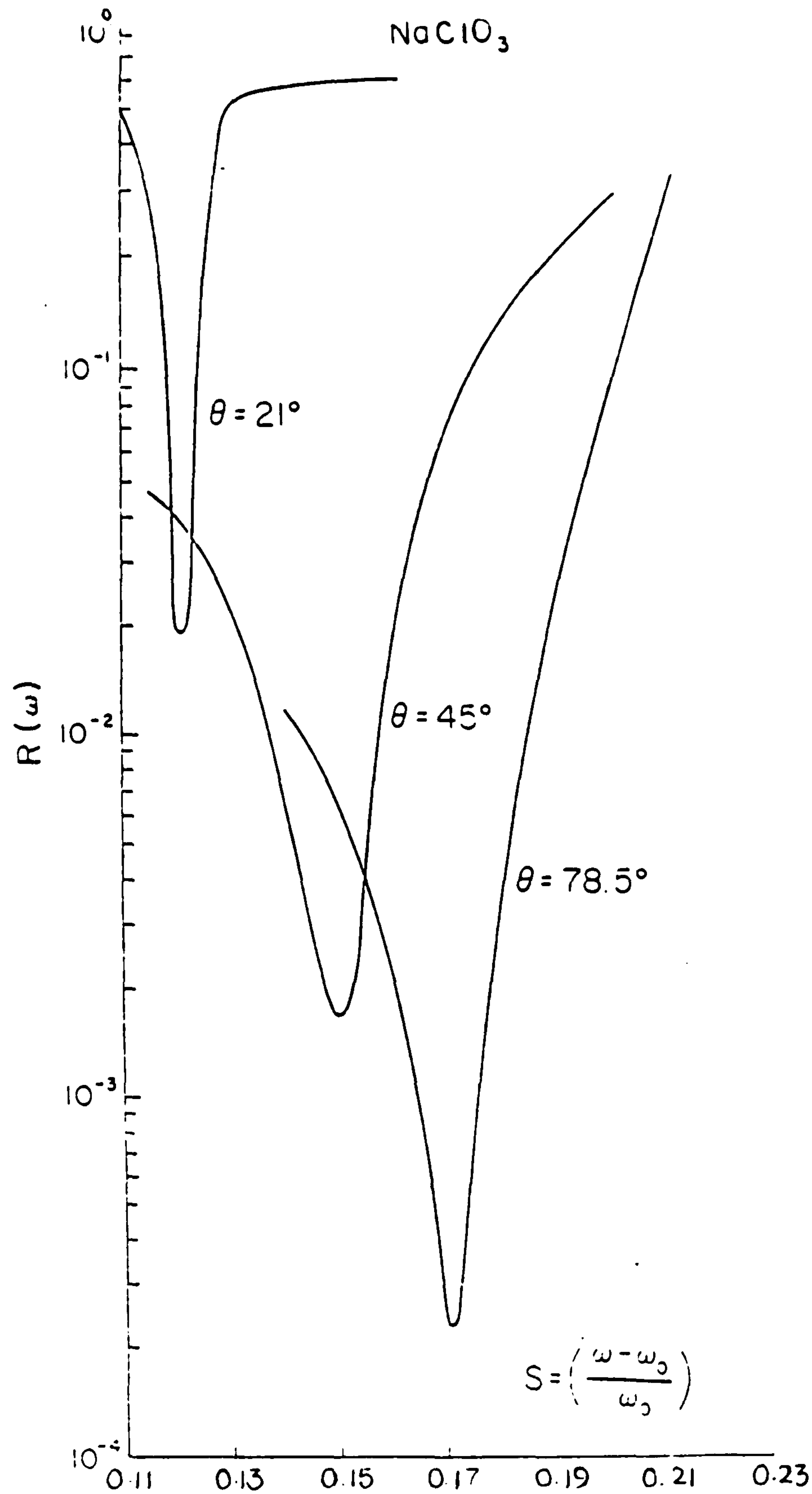

$NaClO_3$
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
R (ω)
θ = 21°
θ = 45°
θ = 78.5°
$S = \left(\frac{\omega - \omega_0}{\omega_0}\right)$
0.11
0.13
0.15
0.17
0.19
0.21
0.23

Fig. 26