Coupling of Electromagnetic Waves in CdS

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A theoretical and experimental study is made of the coupling of the ordinary and extraordinary electromagnetic waves in CdS. The coupling, which may be induced by an external magnetic field or stress, occurs only at energies near the accidental crossing of the index-of-refraction curves $n_0(E)$ and $n_e(E)$. This effect was pointed out by J. J. Hopfield and D. G. Thomas, who showed that this condition, when combined with the anisotropic optical absorption present in the crystal, leads to a discrete absorption line. The oscillatory behavior of the coupled energy and the magnitude of these absorption effects are quantitatively explained. These effects can be used to produce a unique type of optical filter which may be either band-pass or band-reject. Finally, the Faraday rotation in CdS normal to the optic axis is measured at the isotropic point.

I. INTRODUCTION

Recently Hopfield and Thomas[1] have observed and interpreted a new type of optical absorption phenomena in the hexagonal semiconductors ZnO and CdS. The geometry used in these experiments (and in the experiments to be presented) is shown in Fig. 1. In these crystals, at energies below the band gap, light polarized along the y direction (mode y) is strongly absorbed, while light polarized in the z direction (mode z) is more readily transmitted. Hopfield and Thomas found several weak absorption lines for light polarized in the transmitting direction. They named these absorptions polariton absorption lines. The polariton absorption lines are not due to transitions between two discrete electronic energy levels, but result from a coupling of energy from the transmitting mode z to the absorbing mode y. This coupling would ordinarily not take place in an anisotropic crystal. However, in ZnO and CdS there are several accidental crossings of the index of refraction curves $n_y(E)$ and $n_z(E)$. At these crossings the anisotropic crystal becomes optically isotropic and mode y and mode z become coherent and easily coupled. Since this coupling only takes place at energies near the isotropic point, where the refractive index curves cross, it gives rise to discrete absorption lines. Hopfield and Thomas confirmed this explanation by applying a magnetic field in the direction of light propagation which caused a Faraday rotation at the isotropic points, greatly enhancing the polariton absorption lines. They did not explain, however, the reason for the mode coupling in the absence of a magnetic field.

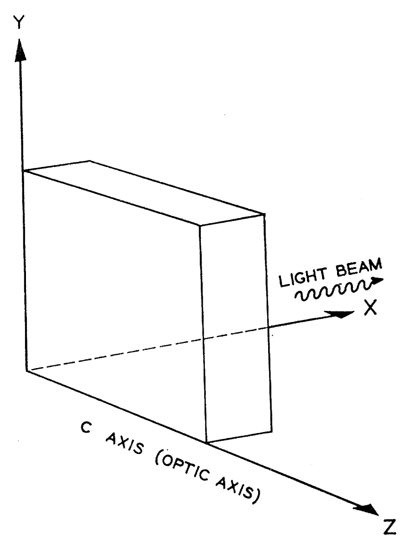

Fig. 1. Experimental geometry.

In this paper, a detailed study of mode coupling effects will be presented. We have produced mode coupling by means of an applied stress as well as by a magnetic field. We will show that these two perturbations can be treated in a unified manner and lead to the same observed effects.

There are great similarities between the mode coupling discussed in this paper and the coupling of modes at different frequencies produced in the field of nonlinear optics, particularly in regard to the effect of wave vector mismatch.[2]

The problem of coupling two electromagnetic waves discussed in this paper is identical to the familiar problem in mechanics of the coupling of two pendula by a weak spring. This is shown in Fig. 2. Suppose pendulum 1 is initially at rest, while pendulum 2 initially has some energy. Assume a negligible energy is stored in the spring and that damping is small. If $\omega_1=\omega_2$, the energy will pass from pendulum 2 to pendulum 1 until finally pendulum 2 is at rest and all the energy that has not been dissipated will belong to pendulum 1. The energy will then flow slowly back to pendulum 2, etc. If $\omega_1\neq\omega_2$, and if $\omega_1-\omega_2$ is large compared to the frequency at which the energy is transferred between identical pendula, then long before pendulum 2 has transferred all of its energy to pendulum 1, the relative phase of the two pendula will change and the energy will begin to flow back to pendulum 2. Thus, the frequency of energy transfer will increase and the energy transfer will be far from complete. The equation describing the transferred energy as a function of time t in the case

[1] J. J. Hopfield and D. G. Thomas, Phys. Rev. Letters **15**, 22 (1965).

[2] See, for example, N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965), Chap. 4.

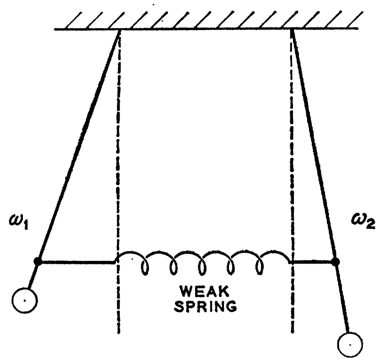

FIG. 2. The mechanical analog of two coupled electromagnetic waves.

of two pendula with frequencies ω_1 and ω_2 is the same as the equation for the transferred energy as a function of distance x through the crystal between two electromagnetic waves with wave vectors k_y and k_z. In the the former case, the transferred energy is a function of $(\omega_1-\omega_2)t$ while, in the latter case, it is a function of $(k_y-k_z)x$.

The theory of mode coupling will be developed in Sec. II, the details of this calculation being presented in the Appendix. In Sec. III the theory is checked by producing mode coupling with a magnetic field and with a uniaxial stress. The coupling was found to be very sensitive to applied stress; a stress of 1 kg/mm^2, which was easily produced, was found to be equal to the coupling produced by 350 kG. The large coupling parameters produced by stress allowed a complete check of the theoretical formulas. The great sensitivity to stress also showed that the polariton absorption lines found in zero magnetic field are due to internal stresses in the sample. The possibility of using the mode coupling effects to produce a unique type of optical filter is also discussed in Sec. III. The results are summarized in Sec. IV.

II. THEORY

We wish to consider the waves polarized in the y and z directions and propagating through the crystal along the x direction as shown in Fig. 1. The wave equation can be written as

$$\frac{\partial^2}{\partial x^2}E_y+\frac{\omega^2}{c^2}E_y=-4\pi\frac{\omega^2}{c^2}P_y\,,$$
$$\frac{\partial^2}{\partial x^2}E_z+\frac{\omega^2}{c^2}E_z=-4\pi\frac{\omega^2}{c^2}P_z\,. \qquad (1)$$

In the absence of perturbations which lower the symmetry of the crystal, the electric polarization $\mathbf{P}$ is related to the electric field $\mathbf{E}$ by a diagonal susceptibility tensor χ of the form

$$\begin{pmatrix}P_y\\P_z\end{pmatrix}=\begin{pmatrix}\chi_{yy}&0\\0&\chi_{zz}\end{pmatrix}\begin{pmatrix}E_y\\E_z\end{pmatrix}. \qquad (2)$$

This leads to the uncoupled wave equations

$$\frac{\partial^2}{\partial x^2}E_y+k_y^2E_y=0\,,$$
$$\frac{\partial^2}{\partial x^2}E_z+k_z^2E_z=0\,, \qquad (3)$$

where

$$k_y^2=\frac{\omega^2}{c^2}(1+4\pi\chi_{yy})=\frac{\omega^2}{c^2}n_y^2\,,$$
$$k_z^2\ \frac{\omega^2}{c^2}(1+4\pi\chi_{zz})=\frac{\omega^2}{c^2}n_z^2\,. \qquad (4)$$

Consider the effect of two perturbations which lower the symmetry of the crystal, a magnetic $\mathbf{H}$ field in the x direction, and a uniaxial stress applied at about 45° with respect to the c axis in the xy plane. These perturbations will cause an additional polarization $\Delta\mathbf{P}$ given by

$$\begin{pmatrix}\Delta P_y\\\Delta P_z\end{pmatrix}=\begin{pmatrix}\Delta\chi_{yy}&\chi_{yz}\\\chi_{zy}&\Delta\chi_{zz}\end{pmatrix}\begin{pmatrix}E_y\\E_z\end{pmatrix}. \qquad (5)$$

The changes in the diagonal elements of χ will only slightly alter k_y and k_z with no important experimental effect. These effects will not be discussed any further.

The off-diagonal elements are responsible for mode coupling. In the first approximation, these elements will be linear in stress and magnetic field. If both stress and magnetic field are present, the change in χ will be

$$\begin{pmatrix}\Delta P_y\\\Delta P_z\end{pmatrix}=\begin{pmatrix}0&\chi\\\chi^*&0\end{pmatrix}\begin{pmatrix}E_y\\E_z\end{pmatrix}$$
$$\chi=\chi_S+i\chi_H\,, \qquad (6)$$

where χ_S and χ_H are real and proportional to the applied stress and to the applied magnetic field. The form of χ follows from general considerations discussed by Landau and Lifschitz.[3] The effect of the off-diagonal elements is to couple wave equations (4);

$$\frac{\partial^2}{\partial x^2}E_y+k_y^2E_y=-\frac{4\pi\chi\omega^2}{c^2}E_z\,,$$
$$\frac{\partial^2}{\partial x^2}E_z+k_z^2E_z=-\frac{4\pi\chi^*\omega^2}{c^2}E_y\,. \qquad (7)$$

These equations are solved in the Appendix. If an energy I_0 is incident upon the crystal in mode y or in mode z, the energy, in the absence of coupling, will merely be attenuated to $I_y(x)$ or $I_z(x)$ after transversing

[3] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley Publishing Company, Reading, Massachusetts, 1958), Sec. 124.

a distance x through the crystal. It is shown in the Appendix that if there is coupling, an energy I_T will be transferred from the incident mode to the other mode, where

$$I_T(x,\beta)=[I_y(x)I_z(x)]^{1/2}|\beta|^2\frac{\sin^2\{[|\beta|^2+(\Delta k/2)^2]^{1/2}x\}}{|\beta|^2+(\Delta k/2)^2}. \tag{8}$$

Here β is a coupling constant porportional to χ (that is, proportional to the applied stress or to the applied magnetic field) and $\Delta k=k_y-k_z$ is the wave vector mismatch between the y and z modes. At the isotropic point, $\Delta k=0$ and

$$I_T(x,\beta)=[I_y(x)I_z(x)]^{1/2}\sin^2(|\beta|x). \tag{9}$$

The energy that is not transferred but remains in the incident mode (I_{NT}) is more difficult to express. At the isotropic point the expression for I_{NT} simplifies to

$$I_{NT}=[I_y(x)I_z(x)]^{1/2}\cos^2|\beta|x. \tag{10}$$

For energies where Δk is large compared to $|\beta|$, I_{NT} approaches the energy in the incident mode for zero coupling, $I_y(x)$ or $I_z(x)$. Thus the total energy I transmitted through the crystal at the isotropic point is given by

$$I(x,\beta)=I_{NT}(x,\beta)+I_T(x,\beta)=[I_y(x)I_z(x)]^{1/2}. \tag{11}$$

III. RESULTS

A. Experimental

To test the theory of Sec. II and the Appendix, measurements were made on polished plates of vapor-grown CdS, ½ mm thick, and with the c axis lying in the plane of the plate. Two perturbations were applied to these crystals: a magnetic field of up to 20 kG applied normal to the plate and a uniaxial stress of up to 2.14 kg/mm² applied in the plane of the plate at 45° with respect to the c axis. I_T was measured by placing the crystal between crossed polaroids and I_{NT} was measured by placing the crystal between parallel polaroids. All measurements were carried out at room temperature except those discussed in Sec. III.D. In these measurements light from a tungsten lamp was passed through the sample and into a 2-m-focal-length Bausch and Lomb spectrograph. The transmission spectrum was recorded with a 1P28 photomultiplier tube.

Figure 3 shows plots of k_y and k_z as a function of

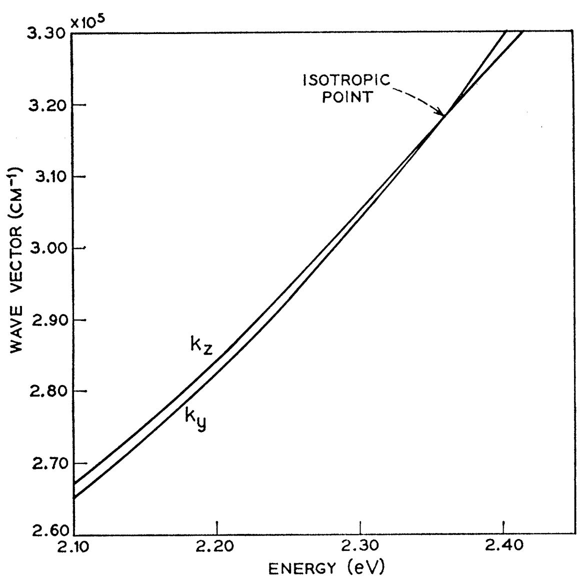

FIG. 3. Wave vectors for mode y, the ordinary ray, and for mode z, the extraordinary ray, in CdS at 300°K.

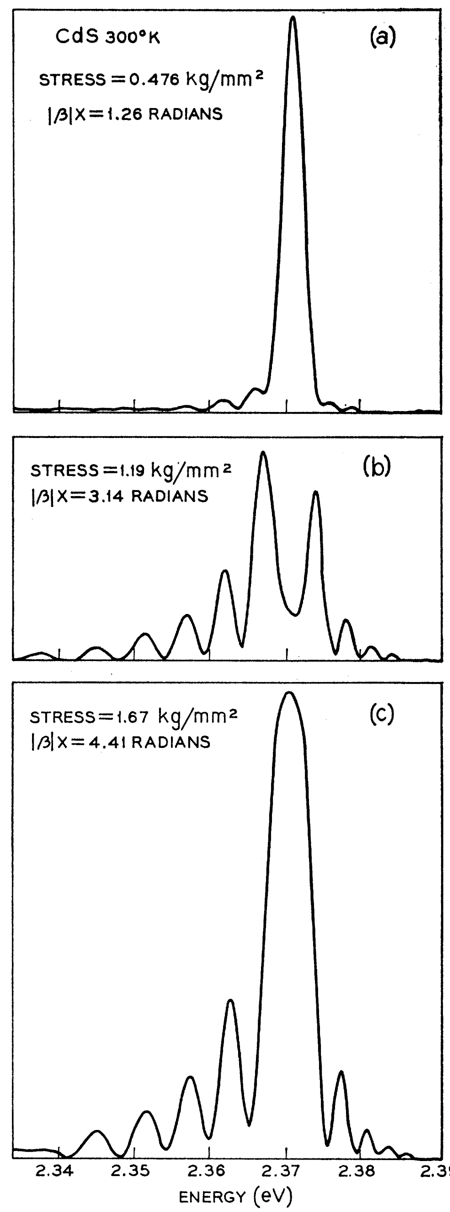

FIG. 4. The transferred energy $I_T(x,\beta)$ for $|\beta|x$ approximately $\pi/2$ in Fig. 4(a), π in Fig. 4(b), and $\frac{3}{2}\pi$ in Fig. 4(c).

energy made from the refractive index measurements of Gobrecht and Bartschat.[4] Near the isotropic point, Δk is almost linear in energy, so that recordings of I_T and I_{NT} versus energy are equivalent to plots of I_T and I_{NT} versus Δk.

B. Mode Coupling Measurements

Typical measurements of I_T produced by a stress are shown in Fig. 4 and a measurement of I_T produced by a magnetic field is shown in Fig. 5. The structure in these spectra is described by Eq. (8). $I_T(x,\beta)$ appears the same when produced by an applied stress or by a magnetic field (for the same value of $|\beta|$). This is predicted by theory. The two effects differ in that in the magnetic case β is pure imaginary, causing Faraday rotation, while in the stress case β is real, causing the incident plane polarized light to become elliptically polarized. $I_T(x,\beta)$ depends only upon $|\beta|$, however, so that it appears the same in both cases. As mentioned in the Introduction, the mode coupling was found to be much more sensitive to stress than to the magnetic field. A moderate stress of 1 kg/mm² produced a $|\beta|x$ of 2.64 rad in a sample 0.5 mm thick. If produced magnetically, the same degree of coupling would have required a field of 350 kG. Because of the large stress couplings available, the stress measurements were ideal to study Eq. (8) for $I_T(x,\beta)$.

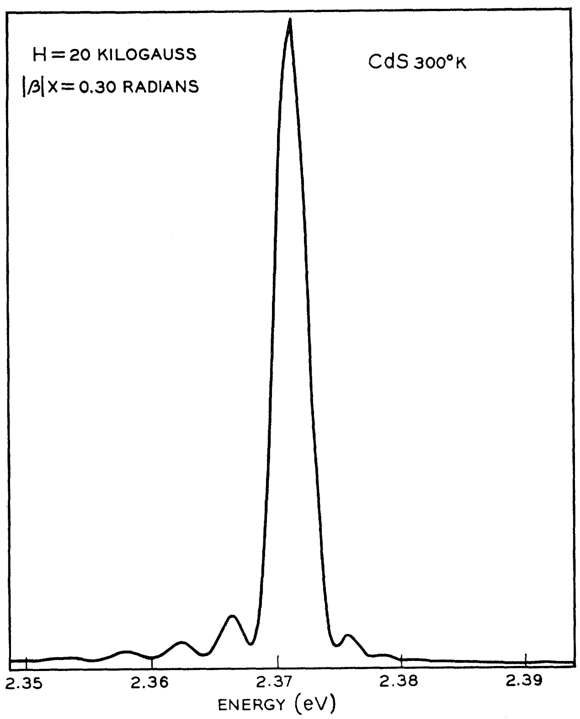

FIG. 5. The transferred energy $I_T(x,\beta)$ in the magnetic case.

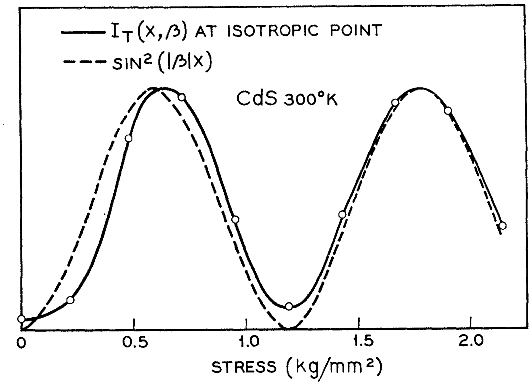

FIG. 6. The transferred energy $I_T(x,\beta)$ measured at the isotropic point as a function of stress.

Equation (9) states that the amplitude of $I_T(x,\beta)$ at the isotropic point is proportional to $\sin^2|\beta|x$. A plot of $I_T(x,\beta)$ at the isotropic point versus stress is shown in Fig. 6. The theoretical fit determined the value of $|\beta|x$ quoted above. The samples used in the stress experiments were only 1 mm×1 mm in area. Because of the small sample size, some stress inhomogeneities were unavoidable. It is believed that these inhomogeneities were the principle cause of discrepancies between theory and experiment in all of the stress measurements.

Figure 4(a) shows a plot of $I_T(x,\beta)$ for small stress. At either side of the isotropic point there is a series of nodes given by

$$[(|\beta|x)^2+(\Delta kx/2)^2]^{1/2}=\pm n\pi,\quad n=1,2,\cdots \tag{12}$$

and a series of maxima which occur at

$$[(|\beta|x)^2+(\Delta kx/2)^2]^{1/2}\approx\pm[(n+\tfrac{1}{2})\pi-1/(n+\tfrac{1}{2})\pi]. \tag{13}$$

As $|\beta|$ increases, the first minima move together and finally join at $|\beta|x=\pi$ as shown in Fig. 4(b); then the first maxima move together and join at $|\beta|x=(\frac{1}{2}3\pi-0.21)$, as shown in Fig. 4(c), etc. Figure 7 shows the measured positions of the first two minima and the

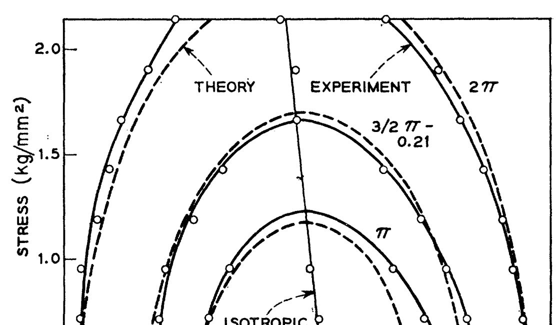

FIG. 7. Energy positions of the first and second minima and the first maximum as a function of applied stress.

[4] H. Gobrecht and A. Bartschat, Z. Physik 156, 131 (1959).

first maxima versus stress and energy. The dashed curves are the positions predicted by theory from Eqs. (12) and (13) assuming that Δk is linear in energy and independent of stress. The horizontal scale of the theoretical curves was fixed by requiring that the two lowest points on the second minima curve coincide with the experimental points. Deviations between theory and experiment are mainly due to Δk being somewhat nonlinear in energy and the isotropic point shifting slowly as the stress is applied.

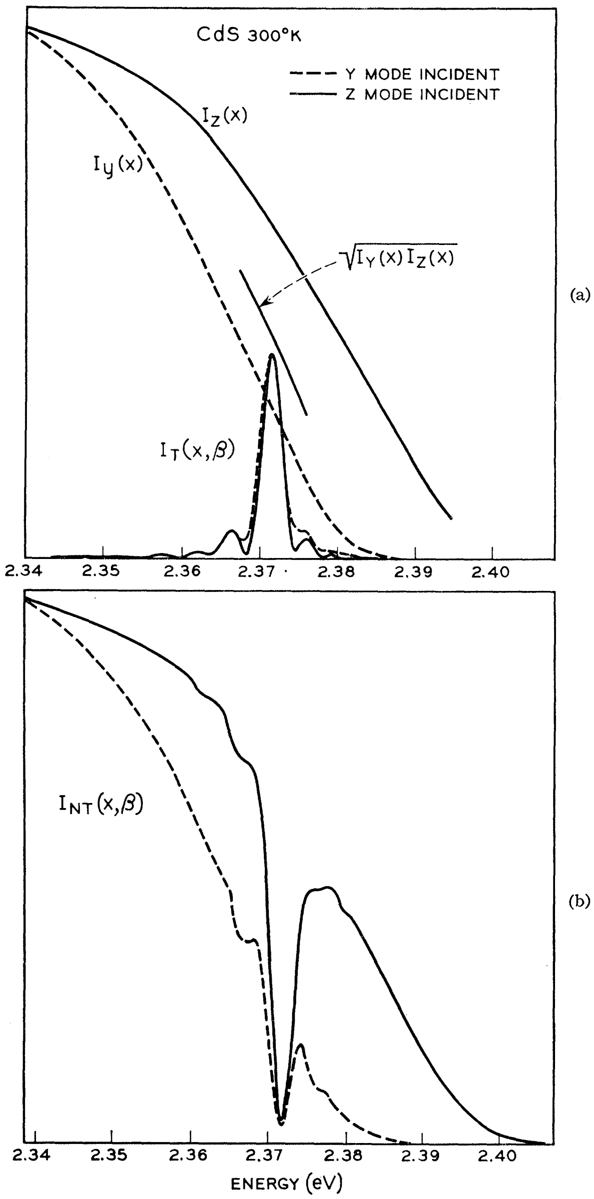

FIG. 8. The energy transferred $I_T(x,\beta)$ and the energy not transferred $I_{NT}(x,\beta)$ at maximum coupling. As predicted, the curves $I_T(x,\beta)$ are identical and the curves $I_{NT}(x,\beta)$ come together at the isotropic point. At the isotropic point, due to stress inhomogeneities, $I_T<(I_yI_z)^{1/2}$ and $I_{NT}>0$. However, the relation $I_T+I_{NT}=(I_yI_z)^{1/2}$ is obeyed.

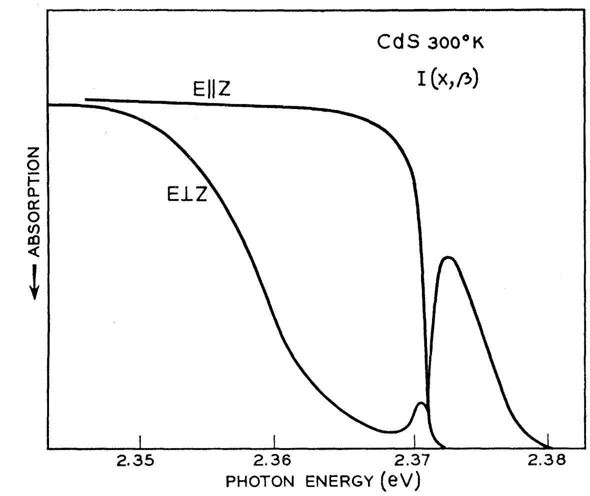

FIG. 9. Data of J. J. Hopfield and D. G. Thomas (Ref. 1) showing the two curves $I(x,\beta)$ touching at the isotropic point.

C. Polariton Absorption Lines

Theory predicts that the transferred energy $I_T(x,\beta)$ will be the same regardless of whether the incident energy is in mode y or in mode z. It is also predicted that at the isotropic point, $I_T(x,\beta)=[I_y(x)I_z(x)]^{1/2}\times\sin^2|\beta|x$. Both predictions are confirmed in Fig. 8(a). $I_{NT}(x,\beta)$, the energy not transferred, is predicted to approach $I_y(x)$ or $I_z(x)$ away from the isotropic point depending upon whether the energy is incident in mode y or in mode z. At the isotropic point, both curves for I_{NT} should approach the common value $I_{NT}(x,\beta)=[I_y(x)I_z(x)]^{1/2}\cos^2|\beta|x$. The experimental curves [Fig. 8(b)] verify this. The curves $I(x,\beta)=I_{NT}(x,\beta)+I_T(x,\beta)$ will approach $I_y(x)$ or $I_z(x)$ away from the isotropic point. At the isotropic point, the two curves for $I(x,\beta)$ will come together and touch at the common value given by the geometric mean of $I_y(x)$ and $I_z(x)$. This is confirmed strikingly by the data of Hopfield and Thomas[1] which we reproduce in Fig. 9.

It should be noted that there will be polariton absorption lines even if unpolarized light is incident on the crystal. For unpolarized light, the transmission curve will be $\frac{1}{2}[I_y(x)+I_z(x)]$ away from the isotropic point. At the isotropic point, the transmission will dip down to $[I_y(x)I_z(x)]^{1/2}$ which is always less than

$$\tfrac{1}{2}[I_y(x)+I_z(x)].$$

D. Optical Filter

Since a moderate stress can cause a coupling of virtually all the energy from one polarization mode to another polarization mode, and since this coupling only takes place near the isotropic point, these effects may be used to produce a unique type of light filter. The device will be a band-pass filter when used with crossed polarizers and a band-reject filter when used with parallel polarizers. The filter energy may be altered by

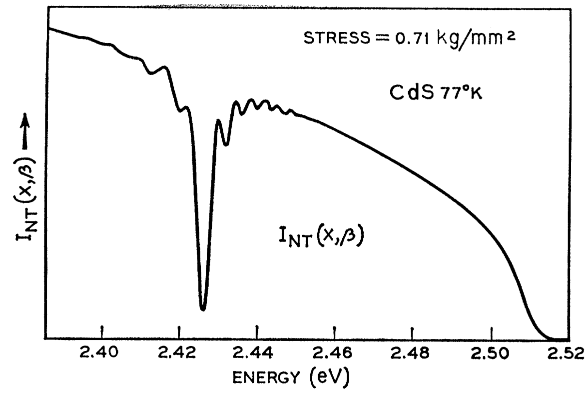

FIG. 10. The energy not transferred, $I_{NT}(x,\beta)$ at 77°K.

varying the temperature. Figure 10 shows $I_{NT}(x,\beta)$ at 77°K.

E. Faraday Rotation at the Isotropic Point

At the isotropic point, one can make a direct measurement of the Faraday rotation with the light and magnetic field directed normal to the c axis. The results are shown in Fig. 11.

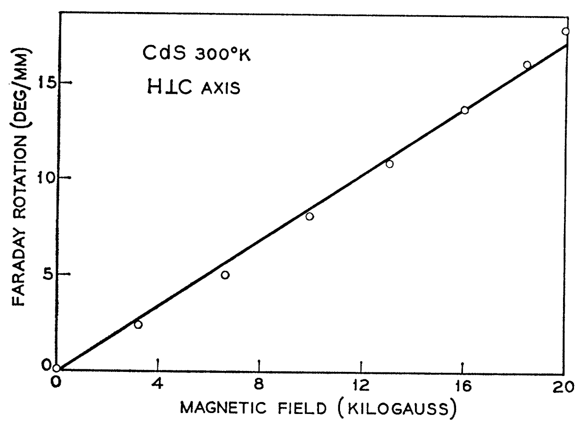

FIG. 11. Faraday rotation measured at isotropic point with $H\perp c$ axis (optic axis).

IV. SUMMARY

We have derived simple analytic expressions describing the coupling of energy between two electromagnetic modes at the same frequency but with different wave vectors. These expressions have been shown to be in accord with experiment in all essential details. This problem greatly parallels a problem encountered in nonlinear optics involving the coupling of energy between electromagnetic modes of differing frequency and wave vector.

The principal results obtained were: (a) The effects of an applied magnetic field and an applied stress may be treated in a unified manner. (b) The great sensitivity of the mode coupling to applied stress allowed the coupling parameter to be varied over a wide range. It also explained the polariton absorption effects observed by Hopfield and Thomas in the absence of a magnetic field. It is felt that such effects were due to internal stress in the samples they used. (c) The touching of the two transmission curves observed by Hopfield and Thomas at the isotropic point was explained. (d) A unique type of light filter could be developed using the mode coupling effects. (e) Faraday rotation in CdS for light directed normal to the c axis was measured at the isotropic point.

ACKNOWLEDGMENTS

The author is greatly indebted to D. G. Thomas whose experiments stimulated this work and with whom the author carried out his first mode coupling measurements. The author had helpful conversations with J. J. Hopfield, J. A. Giordmaine, and D. A. Kleinman about this work. He is grateful to S. Bortas for cutting and polishing samples of CdS.

APPENDIX: SOLUTION OF THE COUPLED WAVE EQUATIONS

We wish to solve Eq. (7) given by

$$\frac{\partial^2}{\partial x^2}E_y+k_y^2E_y=-\frac{4\pi\omega^2}{c^2}\chi E_z\,,$$

$$\frac{\partial^2}{\partial x^2}E_z+k_z^2E_z=-\frac{4\pi\omega^2}{c^2}\chi^* E_y\,,$$

for waves $E_y(x)$ and $E_z(x)$ traveling in the x direction with boundary conditions

$$E_y(0)=0\,,\qquad E_z(0)=E_0\,. \tag{A1}$$

Because the coupling is weak, we expect solutions of the form

$$E_y(x)=A_y(x)e^{ik_yx}\,,\qquad E_z(x)=A_z(x)e^{ik_zx}\,, \tag{A2}$$

where $A_y(x)$ and $A_z(x)$ are slowly varying compared to e^{ik_yx} and e^{ik_zx}. Substitution of Eqs. (A2) into Eq. (7) and neglecting $(\partial^2/\partial x^2)A_y$ and $(\partial^2/\partial x^2)A_z$ leads to two coupled first-order equations

$$\frac{\partial A_y}{\partial x}=i\beta e^{-i\Delta kx}A_z\,,\qquad \frac{\partial A_z}{\partial x}=i\beta^* e^{i\Delta kx}A_y\,. \tag{A3}$$

Δk is the wave vector mismatch given by

$$\Delta k=k_y-k_z\,. \tag{A4}$$

The coupling constant β is given by

$$\beta=\frac{2\pi\chi\omega^2}{c^2k_z}\approx\frac{2\pi\chi\omega^2}{c^2k_y} \tag{A5}$$

in the region of interest where $\Delta k\ll k_y$. Differentiation of Eqs. (A3) leads to the second-order uncoupled differential equations

$$\begin{aligned}&\frac{\partial^2}{\partial x^2}A_y-i\Delta k\frac{\partial A_y}{\partial x}+|\beta|^2A_y=0,\\&\frac{\partial^2}{\partial x^2}A_z+i\Delta k\frac{\partial A_z}{\partial x}+|\beta|^2A_z=0,\end{aligned} \tag{A6}$$

with boundary conditions

$$\begin{aligned}&A_y(0)=0,\quad \frac{\partial A_y}{\partial x}(0)=i\beta E_0,\\&A_z(0)=E_0,\quad \frac{\partial A_z}{\partial x}(0)=0,\end{aligned} \tag{A7}$$

determined by Eqs. (A1) and (A3). Solving Eq. (A6) and using Eq. (A2) gives

$$E_y(x,\beta)=E_0\exp\tfrac{1}{2}i(k_y+k_z)x(i\beta)\frac{\sin\{[|\beta|^2+(\Delta k/2)^2]^{1/2}x\}}{[|\beta|^2+(\Delta k/2)^2]^{1/2}} \tag{A8}$$

and

$$E_z(x,\beta)=E_0\exp\tfrac{1}{2}i(k_y+k_z)x\Bigg[\cos\{[|\beta|^2+(\Delta k/2)^2]^{1/2}x\} -i\sin\{[|\beta|^2+(\Delta k/2)^2]^{1/2}x\}\frac{(\Delta k/2)}{[|\beta|^2+(\Delta k/2)^2]^{1/2}}\Bigg]. \tag{A9}$$

Let I_0 be the incident energy, I_T the transferred energy observed by placing the crystal between crossed polaroids, and I_{NT} the energy remaining in the incident mode that has not been transferred or lost through absorption. Then

$$I_0\sim|E_0|^2,\quad I_T\sim|E_y|^2,\quad I_{NT}\sim|E_z|^2. \tag{A10}$$

In the absence of dissipation, we find

$$\begin{aligned}&I_T(x,\beta)=\frac{I_0|\beta|^2\sin^2\{[|\beta|^2+(\Delta k/2)^2]^{1/2}x\}}{[|\beta|^2+(\Delta k/2)^2]},\\&I_{NT}=I_0-I_T(x,\beta).\end{aligned} \tag{A11}$$

The effects of absorption may be taken into account by making the wave vector complex

$$\begin{aligned}&k_y=k_y'+ik_y'',\\&k_z=k_z'+ik_z''.\end{aligned} \tag{A12}$$

In the absence of coupling, an incident energy I_0 would be attenuated to energy $I_y(x)$ in mode y and to energy $I_z(x)$ in mode z after traveling a distance x through the crystal:

$$\begin{aligned}&I_y(x)=I_0e^{-2k_y''x},\\&I_z(x)=I_0e^{-2k_z''x}.\end{aligned} \tag{A13}$$

Using this notation, the coupled energy can be expressed as

$$I_T(x,\beta)=[I_y(x)I_z(x)]^{1/2}|\beta|^2\times\left|\frac{\sin\{[|\beta|^2+(\Delta k/2)^2]^{1/2}x\}}{[|\beta|^2+(\Delta k/2)^2]}\right|. \tag{8'}$$

In the experiments described in this paper, the imaginary part of the wave mismatch $i\Delta k''$ makes only a negligible contribution to the phase $[|\beta|^2+(\Delta k/2)^2]^{1/2}x$ and will be neglected in Eq. (A14). The general expression for $I_{NT}(x,\beta)$ cannot be written down so simply. For large Δk, $I_{NT}\approx I_y(x)$. At the isotropic point where $\Delta k=0$,

$$I_T(x,\beta)=[I_y(x)I_z(x)]^{1/2}\sin^2|\beta|x, \tag{9'}$$

$$I_{NT}(x,\beta)=[I_y(x)I_z(x)]^{1/2}\cos^2|\beta|x, \tag{10'}$$

$$I(x,\beta)=I_T(x,\beta)+I_{NT}(x,\beta)=[I_y(x)I_z(x)]^{1/2}. \tag{11'}$$