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PHYSICAL REVIEW B VOLUME 4, NUMBER 12 15 DECEMBER 1971

Spectral Dependence and Polarization Anisotropy of Optical-Transmission Modulation by Acoustoelectric Domains in CdS†

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(Received 2 August 1971)

The modulation by acoustoelectric domains of monochromatic light transmitted through platelets of CdS has been studied with emphasis on the spectral variation and polarization dependence of the effect near the band edge. Samples of several crystallographic orientations were used. It was found that for light propagating along the *c* axis of the crystal, the domain-induced modulation was strongest with the light polarized along the drift-field direction, in agreement with previously reported work on GaSb. The spectral variation of the transmission modulation observed with polarized light propagating perpendicular to the *c* axis was consistent with the double band edge of CdS. The modulation resembles a shift of the absorption spectrum to longer wavelengths. Recently published theoretical and experimental studies of the Franz-Keldysh effect, together with the observed spectral, polarizational, and orientational dependences of the optical modulation lead us to conclude that the modulation is produced by the Franz-Keldysh effect. Further, it is shown that the fluctuating high-frequency electric fields in the domain, rather than the "steady" field across the domain, produce the effect. Previously measured properties of the domain, such as current noise, induced birefringence, and induced light emission, corroborate this interpretation. Other proposed mechanisms are shown to be invalid.

I. INTRODUCTION

Several techniques have been used for probing acoustoelectric domains in piezoelectric semiconductors. These include direct electrical probing, microwave probing, and optical probing. One optical probing technique involves observing the transient change in optical transmission of the semiconductor sample as the domain traverses the optical probe. This technique was used by Kumar, Sliva, and Bray[1] in GaSb, and by Spears and Bray[2] in GaAs. It was demonstrated that the change in the optical-absorption coefficient correlated well with the domain strength as measured by other techniques, and that the modulation was strongest for wavelengths approaching the band edge of the semiconductor. More recently, Kumar and Hutchinson[3] and Yamamoto *et al.*[4] applied the technique to CdS and observed the characteristics of domain propagation in that material. The work being reported here is primarily concerned with the spectral variation of the transmission modulation produced by acoustoelectric domains in CdS at wavelengths near the band edge, and with the relationship between the orientational dependence of this spectral variation, including polarization anisotropy, and the band structure of CdS. These topics have been covered only briefly in the available literature.

The results of the present work viewed in the light of recently published experimental and theoretical studies of the Franz-Keldysh effect, lead us to conclude that this effect is responsible for the observed modulation. However, it is the strong fluctuating electric fields in the domain, associated with the intense high-frequency phonon flux, rather than the "steady" field across the domain, which produce the effect. The presence of such strong fluctuating fields is adduced also from earlier studies of the acoustoelectric domains in CdS, namely, the high levels of current noise at current saturation, induced birefringence, and induced light emission.

II. DESCRIPTION OF EXPERIMENT

The system used for measuring transmission modulation in the presence of acoustoelectric domains is shown in Fig. 1. A Perkin-Elmer model 83 monochromator containing a quartz prism was used in conjunction with a pulsed xenon lamp. The peak pulsed-light intensity near the band edge of CdS was over 1000 times greater than that obtainable from CW arc lamps, and this high intensity was essential to the experiment. Furthermore, the xenon spectrum was relatively flat, a desirable characteristic for spectral measurements. The detector was an RCA 6217 photomultiplier which,

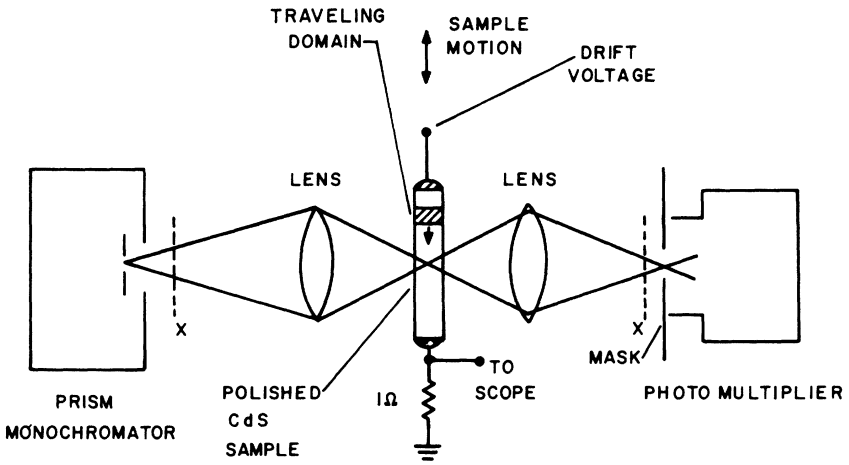


FIG. 1. Diagram of the experimental apparatus used for the measurement of transmission modulation. For some of the measurements, polarizers and color filters were placed at the positions marked "X."

when connected to the oscilloscope, had a measured risetime of 20 nsec. The probe had a spatial resolution of 50 μ. The CdS samples were pulsed with a specially built vacuum tube pulser capable of providing 20 A at 1.5 kV. The lamp and the pulser were triggered by a Tektronix 546/1A1 oscilloscope and were timed so that the structure in the optical transmission occurred at the peak light intensity.

The samples were cut from Eagle-Picher UHP single-crystal CdS of resistivity between 10 and 50 Ωcm. The samples were 5–10 mm long in the direction of current flow, 1.25 mm wide and 0.5–1 mm along the optical path. Contacts were made with an In-Ga eutectic. All measurements were made at room temperature. A typical domain signal is shown in Fig. 2(a). Here the shape of the light pulse is seen, as well as a domain signal of almost 50% modulation. Figure 2(b) shows a modulation signal 60 nsec long corresponding to a domain 100 μ wide.

III. SPECTRAL AND POLARIZATIONAL DEPENDENCE OF MODULATION

The dependence of the modulation on the polarization of the incident light is particularly interesting in CdS since CdS is a uniaxial crystal whose optical band gap is dependent upon the polarization of the light. Several sample orientations were used in this work, as shown in Fig. 3. In orientations "*a*" and "*b*" in this figure, the drift field is perpendicular to the *c* axis. The domains obtained are known to be narrow and are squared up with the sample.[5] In Fig. 3(a) the light propagates along the *c* axis, while in Fig. 3(b) the light propagates in a direction perpendicular to the *c* axis. Thus, in the absence of a domain, in orientation *a* the crystal appears optically isotropic, while in orientation *b* it is birefringent and exhibits the two optical band edges.[6] Another difference is that elastooptic coupling exists between domain stresses and the light in *b* but not in *a*. A possible consequence of the elastooptic coupling in *b* is photoelastic scattering. Indeed, *b* is the orientation primarily used by investigators interested in studying domain-induced birefringence,[5] or Brillouin scattering.[7] Some of the samples used in this work were polished on all sides so that the same samples could be used in orientations *a* and *b*. While this had certain advantages, it was preferable to have the samples thin along the optical path so that modulation of strongly absorbed light could be observed. Thin samples could be used in only one orientation.

In Fig. 3(c) the sample is oriented with the drift field parallel to the *c* axis. This is the orientation for which "off-axis" domains are known to occur.[5] This orientation is similar to *b* in that natural birefringence exists as well as elastooptic coupling to the domain stresses.

Figure 3(d) describes a sample cut so that the drift field makes an angle of 30° with the *c* axis. Because the domains arising in samples of type "*c*" were known to contain shear waves propagating 30° off axis, it was suspected that, in "*d*," domains aligned with the dimensions of the sample might occur. Such samples were studied independently by Moore[8] using photoelastic stroboscopic photography, and this was shown to be the case. For the transmission modulation work done here, there was no reason to use polarized light for orientation *d* since the principal axes of the crystal were neither parallel nor perpendicular to the drift field, and light initially polarized in one of these directions would have been rapidly depolarized by the natural birefringence of the crystal. However, initially unpolarized light, which was partially absorbed by the band edge, became effectively polarized along the *c* axis because of the stronger absorption of light polarized perpendicular to the *c* axis of CdS.

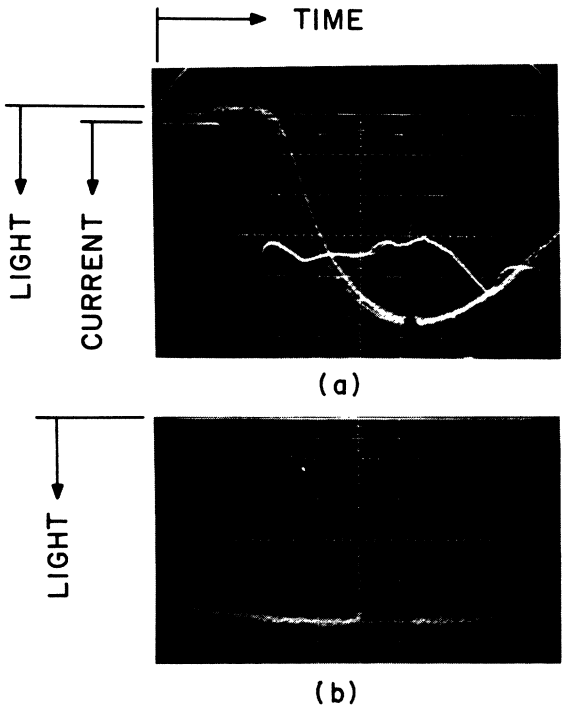


FIG. 2. Transmission modulation observed with the pulsed lamp. (a) Traces of sample current (0.3 A/div) and optical signal (relative magnitude) as a function of time (0.5 μsec/div). (b) Expanded view of a similar, but stronger, optical signal (0.1 μsec/div). The drift field was perpendicular to the c axis; the light propagated along the c axis and was polarized along the drift-field direction, as in Fig. 3(a).

A. Results with Samples Oriented Perpendicular to c Axis

When studying the polarization dependence of the transmission modulation in samples of this orientation, it is important to make a distinction between the cases sketched in Figs. 3(a) and 3(b). In *a*, where the optical electric field is in the basal plane of the crystal, any polarization dependence of the modulation is a consequence of the symmetry reduction caused by the acoustoelectric domain itself. In *b*, where the light travels in a direction perpendicular to the c axis, this is not so since one polarization is perpendicular to the c axis while the other is parallel to it. Whatever features the curve of modulation vs photon energy has in the first case may be expected to occur here too, but in addition, we might expect to find the curve for light polarized parallel to the c axis shifted towards higher energies by 18 meV, the difference between the optical band gaps for the two principal polarizations.

It is seen in Fig. 4 [orientation of Fig. 3(a)] that the modulation signal for light polarized parallel to the drift field is larger than for light polarized perpendicular to it. Although the samples were not quiescently birefringent or dichroic for light travelling in this direction, this result was observed in all CdS samples of this orientation. Kumar *et al.*[1] reported that, in GaSb, the modulation of light polarized parallel to the drift field was 17% larger than the modulation of light polarized perpendicular to it. Since GaSb is a cubic crystal, it is optically isotropic, and is in that respect similar to CdS of this orientation. Since the domain stresses in these two isotropic situations are also of the same orientation with respect to the applied field, the qualitative agreement between GaSb and CdS samples of type *a* is certainly meaningful. Also plotted in Fig. 4 is the measured relative transmission of the sample as a function of photon energy. It is clearly seen that the modulation becomes large where the band-gap absorption becomes strong, establishing the modulation as a band-gap process.

The results obtained from a similar measurement on a sample of the type sketched in Fig. 3(b) are shown in Fig. 5. The optical transmission of the sample for the two polarizations in the absence of domains is also shown. Here, the difference in the modulation for the two polarizations appears larger than in the previous case because of the combined effects of (i) the difference between the absorption edges for the two polarizations and (ii) the larger modulation for light polarized paral-

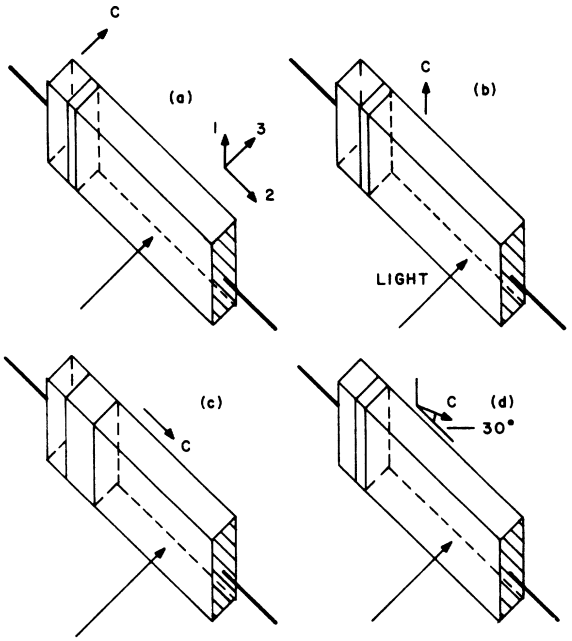


FIG. 3. Crystal orientations used in the experiments. The heavy solid lines in each case indicate the direction of the drift current and the arrows indicate the direction of light propagation.

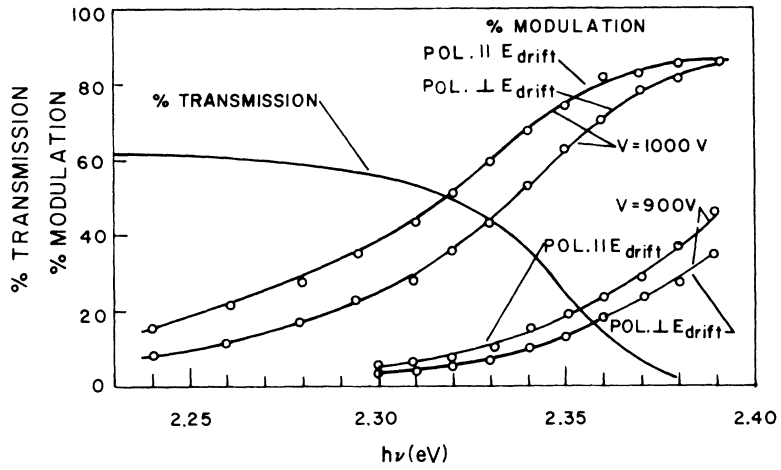


FIG. 4. Spectral dependence of the transmission modulation for a sample with the drift field perpendicular to the c axis and the light propagating along the c axis [Fig. 3(a)]. The sample was 1 mm thick in the direction of the optical path and 7 mm long in the direction of current flow. The figure shows data for two domain strengths and for both polarizations. Also shown is the measured transmission of the sample in the absence of domains.

lel to the drift field.

A few of the samples studied had square cross sections and were polished on all sides, thus permitting the same domains to be probed in both orientation a and b. A valid comparison is obtained by using only light polarized parallel to the drift field (hence, perpendicular to the c axis). Light polarized perpendicular to the drift field would be parallel to the c axis in one case, b, and perpendicular to the c axis in the other case, a. For each sample, the difference in modulation between the two orientations was small and could have been in either direction. The data taken for different samples and for different points in the same sample differed in detail, but yielded the same general results pointed out above. The modulation always increased sharply at the absorption edge and, in the isotropic direction, was larger for light polarized parallel to the drift field. The presence of the double band edge in the orientation in Fig. 3(b) always showed up as a shift between the curves for the two polarizations.

B. Results with Samples Oriented Parallel to c Axis

The samples oriented with their drift field along the c axis were very difficult to work with. The modulation caused by the domains was relatively weak and had fine structure that changed along the sample length. Since it was known that shear waves travelling at angles of about 30° from the c axis were present in these domains,[5] the sample holder was rotated 30° in each direction from its normal position in the hope that, with the monochromator slit aligned with the domain structure, a more definitive domain signal would be observed. However, the attempt was largely unsuccessful. Most of the samples showed very little difference in the type of signal seen as the sample holder was rotated. One sample, however, seemed to show a double dip in transmission at many points along its length when aligned perpendicular to the slit image, but showed a single dip over a large part of its length when the sample holder was rotated about 30° in either direction. Since this was observed in only one sample, it is not certain that the observation is meaningful. The behavior of domains in samples oriented parallel to the c axis appears to be very complicated and may very well depend upon the sample and electrode shapes as well as its bulk properties. One sample, when probed near the anode, gave reasonably clean and repeatable modulation signals. The spectral dependence of the modulation for this sample is shown in Fig. 6. The difference between the modulation signals for the two polarizations is much smaller than it was for any other orientation. This is

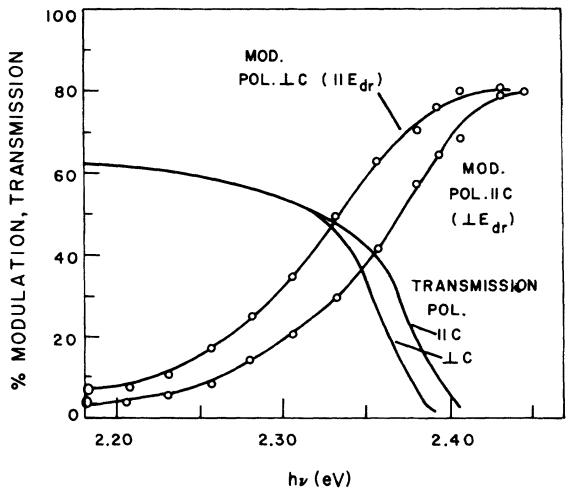


FIG. 5. Spectral dependence of the transmission modulation for a sample with the drift-field and the light-propagation directions both perpendicular to the c axis [as in Fig. 3(b)]. The unmodulated transmission is also shown for both polarizations. The sample was 0.5 mm thick and 6 mm long.

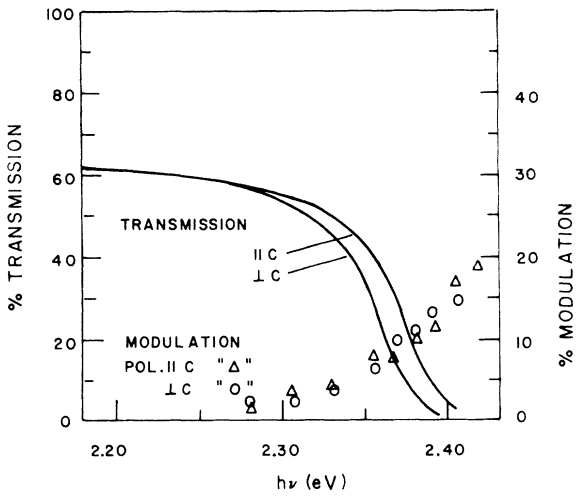


FIG. 6. Spectral dependence of the transmission modulation for a sample with the drift field along the c axis. Both polarizations are shown for both the modulation and the unmodulated transmission. The sample was 0.5 mm thick and 7 mm long.

probably because the effect of the different band edges for the two polarizations and the effect of the modulation being larger when the light is polarized parallel to the drift field tended to cancel each other for this orientation.

C. Results with Samples Oriented 30° from c Axis

The samples with the configuration shown in Fig. 3(d) produced domain modulation signals whose spectral dependence was similar to that in Fig. 6, but whose magnitude was somewhat larger. Still, it was not possible to look deeply enough into the absorption edge to see the modulation saturate. The modulation signals were still increasing with photon energy for strongly absorbed light. Rotating the sample holder from its normal position did not increase the modulation, suggesting that the domain was aligned along the same axes.

IV. CALCULATION OF THE CHANGE IN ABSORPTION COEFFICIENT

In order to facilitate understanding of the mechanism responsible for the observed transmission modulation, it is desirable to have the data in terms of the optical-absorption coefficient α of the material. The standard relation between the transmission T of a slice of material and the absorption coefficient α of the material is

$$T = (1-R)^2 e^{-\alpha d}/(1-R^2 e^{-2\alpha d}) , \tag{1}$$

where R is the reflection coefficient at each surface and d is the thickness of the slice. For all but very thin samples, the denominator may be taken equal to unity. Using this approximation, it follows that an increase in absorption coefficient $\Delta\alpha$ results in a new transmission T' given by

$$T'/T = e^{-d\Delta\alpha} = 1 - m \tag{2}$$

or

$$\Delta\alpha = -(1/d)\ln(1-m) . \tag{3}$$

The quantity m is the modulation fraction. For m small compared to unity, Eq. (3) becomes

$$\Delta\alpha \approx m/d , \quad m \ll 1 . \tag{4}$$

We see from Eq. (2) that if the transient change in the absorption coefficient $\Delta\alpha$ increases continuously with photon energy, the modulation m also increases monotonically with energy, and can saturate only at 100%. Indeed, Kumar *et al.*[1] and Spears and Bray[2] reported that in similar experiments with III-V semiconductors, the observed modulation always did approach 100% with increasing photon energy. Since our results showed the modulation to saturate at a value less than 100%, the experiment deserves closer examination. The modulation caused by weak domains was measured over the same photon energy range in which the modulation caused by strong domains appeared to saturate at a value less than 100%. It is seen in Fig. 4 that the modulation caused by the weak domain (and thus the change in absorption $\Delta\alpha$) continued to increase with photon energy where the modulation due to the strong domain saturated. Unless we are willing to believe that there is a maximum value that the change in the absorption coefficient α can have regardless of the original absorption coefficient α or the domain strength, this result appears anomalous. The idea of a maximum $\Delta\alpha$ not only appears unlikely, but is in disagreement with the results of Kumar *et al.*[1] and Spears and Bray.[2] We therefore believe that the anomaly results from a limitation of our experiment, namely, the speed of response of the optical system.

Consider the effect of a finite photodetector bandwidth on the observed modulation signal. A light pulse whose half-width (≈50 nsec) is only a few times the half-width of the impulse response of the optical system (≈20 nsec) will be slightly modified in shape. It will be slightly wider and smaller in amplitude. However, since the optical system is a linear system, the height of the pulse, seen on the oscilloscope as a function of photon energy for a given domain, is directly proportional to the true height of the light pulse. Since the maximum true height of the light pulse is 100% of the unmodulated light, and since it is expected that the true modulation goes to 100% as the photon energy is increased, the true change in the absorption coefficient can be found from the expression

$$\Delta\alpha = -(1/d)\ln(1 - m/m_{\max}) . \tag{5}$$

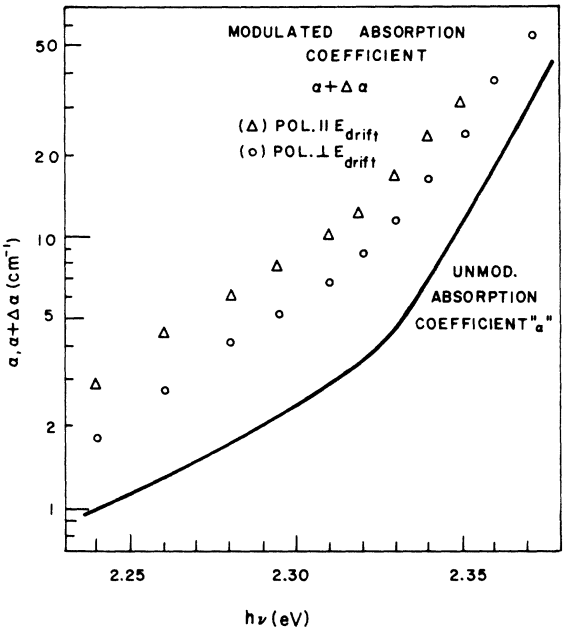


FIG. 7. The modulated absorption edges for the two polarizations as compared with the unmodulated absorption edge for the data on the stronger domain in Fig. 4.

Thus the saturation which is believed to lie at 100% provides a reference point for scaling the curve. For small modulation signals,

$$\Delta\alpha = m/m_{\max} d \ . \tag{6}$$

It is also seen in Fig. 4 that while the change in the absorption coefficient for light polarized parallel to the drift field is larger than for the other polarization, the two curves appear to saturate at the same modulation. This is consistent with the interpretation of the saturation at a value less than 100% as a limited-bandwidth effect. While it is possible to increase the system bandwidth making $m_{\max}$ higher, there is little to be gained since it would still be necessary to scale the resultant modulation curve up to 100% by Eq. (5), and the noise on the signal would increase with bandwidth while the signal would increase only slightly. Speed of response was not a problem for Kumar *et al.*[1] and Spears and Bray[2] since their domains were an order of magnitude wider than those in CdS.

The change in the absorption coefficient caused by the stronger domain in Fig. 4 was calculated for both polarizations using Eq. (5). The corresponding modulated absorption coefficients $(\alpha + \Delta\alpha)$ are plotted in Fig. 7 and compared with the unmodulated absorption coefficient α. Similar curves were calculated and plotted from the data taken with samples of the other orientations. These display the same features seen in the curves of percent modulation, namely, the effect of the double band edge and the polarization anisotropy.

It is of interest to consider what the spectral dependence of the modulation would be like if the effect of the modulation were equivalent to a pure shift in the band edge. Dutton[6] found that the spectral and temperature variation of the absorption coefficient α could be fitted by the relation

$$\alpha = \alpha_0 \exp[-(\beta/kT)(E_0 - h\nu)] \ , \tag{7}$$

where α_0, β, and E_0 are fitting parameters with E_0 depending upon polarization. This expression is valid for absorption coefficients as high as 10^4/cm, a value much higher than the greatest absorption coefficient encountered in this experiment. If the edge were shifted by the amount ΔE, the absorption coefficient would change by

$$\begin{aligned}\Delta\alpha &= \alpha_0 \exp[-(\beta/kT)(E_0 - h\nu - \Delta E)] - \alpha \\ &= \alpha\{\exp[(\beta/kT)\Delta E] - 1\} \ . \end{aligned} \tag{8}$$

Since the term in curly brackets in the final expression depends only upon the shift and not on the incident photon energy, we find that the change in the absorption coefficient is directly proportional to the absorption coefficient itself, and the latter increases without limit over a tremendous range. From Eq. (2), it is clear that a domain producing solely a shift of the absorption curve would cause the modulation to approach 100% for strongly absorbed light. Even if not a pure shift, this last statement is valid for any change in the absorption curve remotely resembling a shift in the band edge.

V. OBSERVATION OF LIGHT SCATTERING

As a check for scattering, the modulation was observed with parallel polarizers on either side of the sample. Since the sample was optically isotropic for the orientation in Fig. 3(a), and since the polarizers were parallel to a principal axis of the sample for each of the other orientations in Fig. 3(b), the presence of a second polarizer did not significantly change the amount of unmodulated light reaching the photodetector. It is known that when light is scattered by shear waves, the plane of polarization of the light is rotated by 90°. Therefore, light scattered by the domain through angles sufficiently small that the photodetector can still collect it would be blocked by the second polarizer. The modulation observed in this manner would be larger than without the second polarizer if scattering were present. While the absence of a change in modulation when the second polarizer is inserted would not rule out the contribution of large-angle scattering to the observed modulation, it would make it appear less likely since the $f/4$ optical system may be expected to collect a measureable fraction of the predominantly forward-scattered light.

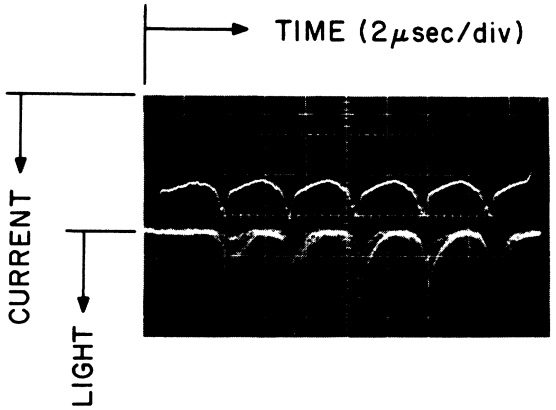


FIG. 8. Domain-related light emission from the anode region. The current (0.3 A/div) and the emitted light (relative magnitude) are compared as a function of time (2 μsec/div). The drive pulse was long enough to permit several domain transits.

For samples with the light travelling along the *c* axis, the insertion of the second polarizer was found to increase the modulation by about 2% of the unmodulated signal. When the polarizers were crossed, there was no light transmitted in the absence of a domain, but the passage of a domain created an optical signal consistent with the additional modulation in the previous argument. The contribution of scattering to the modulation was found to be almost independent of photon energy. It is not surprising that the scattering is very small for this orientation since the elasto-optic coupling is theoretically zero. However, the light beam has a cone angle of about 10°, and the shear waves themselves are known to have a spread of several degrees, accounting for the small amount of scattering observed.

For samples in which the light was propagated in a direction not along the *c* axis, the scattering was larger, ranging from about 5 to 10% of the unmodulated signal, with considerable variation between samples. However, for a given sample, the modulation was still almost independent of photon energy. Since, for the orientations in Fig. 3(b), the modulation due to scattering was variable and, over part of the range of interest, was larger than the modulation caused by the change in the absorption coefficient, the study of the band edge in the presence of domains is best accomplished with samples of the type in Fig. 3(a). Another determination of whether scattering is present can be made from the spectral dependence of the modulation. This is discussed later.

VI. DOMAIN-CONTROLLED LIGHT EMISSION

During the course of the experiment, it was found that many of the samples emitted light. The magnitude of this emission varied widely from sample to sample, and from pulse to pulse in the same sample. The most significant feature of the light emission is that it always came from the anode end of the sample, and that it occurred just as the domain struck the anode. It can be seen in Fig. 8 that the spikes of light occur just when the current returns to its Ohmic value with very little light between the spikes. It can also be seen in Fig. 8 that within a burst of light pulses arising from a drift pulse long enough to permit several domain transits, the emitted light pulses exhibit a tendency to increase. However, this can be seen only when several bursts are simultaneously viewed, as in Fig. 8. If a single burst were viewed, the amplitudes of the light pulses would appear almost random. This randomness made it very difficult to obtain any quantitative information. The light was examined with color filters, and it was qualitatively determined that the emission was in the red, not at the band edge, and that it was not polarized.

The phenomenon of domain-controlled light emission has been previously observed by several workers although relatively little discussion of it has appeared in the literature. It was reported by Yee[9] in CdS at 77 °K where the emission was at the band edge. This difference is likely to be related to the difference in temperature, since band-edge emission from CdS is not normally seen at room temperature. While the light emission seen by Yee had the periodicity of the acoustoelectric oscillations, his samples were too short to establish domain arrival times. The phenomenon was also reported by Kumar *et al.*[1] in GaSb, where two decay times were observed. The light first decayed rapidly after the domain struck the anode, but a small part of it persisted for many μsec longer. This was not seen here in CdS.

VII. DISCUSSION

Several mechanisms have been suggested[1,2] to explain the optical modulation near the absorption edge. These include the Franz-Keldysh effect,[10] deformation-potential modulation of the gap,[11] self-energy of the electron in the intense high-frequency phonon field,[12] uncertainty broadening of the absorption,[13] and photoelastic scattering.[14] Recently published theoretical and experimental results[15,16] on the Franz-Keldysh effect, taken together with the observed properties of the optical modulation, as well as other properties of the acoustoelectric domain, lead us to conclude that this effect produces the modulation. However, the *strongly fluctuating* electric fields inside the domain, associated with the intense flux of partially incoherent high-frequency phonons generated by the acoustoelectric interaction rather than the steady field across the domain, appear to be the source of the effect.

A. Franz-Keldysh Effect

The Franz-Keldysh effect[10] is the increase in the optical-absorption coefficient near the band edge of a semiconductor in the presence of a strong electric field. In materials whose optical-absorption edge takes an exponential form, the effect of the electric field is to shift the band edge toward smaller photon energies by an amount proportional to the square of the electric field. The dependence of the constant of proportionality on the polarization of the light with respect to the strong electric field is of particular interest to us since an asymmetry of this type appeared in our modulation experiments, as well as in previous ones. The theoretical literature up to 1968 had been singularly unenlightening, and even misleading, on this point. Nonetheless, Bagaev *et al.*[15] demonstrated that a strong electric-field-induced anisotropy does indeed exist in GaAs and CdTe, which are both cubic and, hence, isotropic semiconductors. For both materials, the change in the absorption coefficient was largest for light polarized along the applied electric field. In explanation of these results, Keldysh *et al.*[16] then explicitly exhibited the asymmetry theoretically for a direct-gap III-V semiconductor, ignoring the lack of inversion symmetry, i.e., taking the valence band as fourfold degenerate—a common approximation in theoretical studies of the III-V semiconductors where correct treatment of the inversion nonsymmetry generally leads to only insignificant corrections.

The experimental results of Bagaev *et al.*[15] and theoretical calculations of Keldysh *et al.*[16] are completely in line with our observations of the polarization anisotropy in the optical modulation induced by the acoustoelectric domains in CdS. In our measurements the modulation was strongest with the polarization along the drift field. The fluctuating high-frequency electric fields in the domain are also essentially along this direction since the high-frequency phonons generated by the drifting electrons are bunched tightly in angle in the forward drift direction (see below). Furthermore, the ratio of the change in the absorption coefficient for light polarized along the drift field to the change in the absorption coefficient for light polarized perpendicular to the drift field does increase with increasing photon energy deficit, as also seen by Bagaev *et al.*[15] and predicted theoretically by Keldysh *et al.*[16] This can be seen in Fig. 7 where the log of this ratio is the vertical distance between the two data curves.

Strictly speaking, the calculations of Keldysh *et al.*[16] refer to situations of cubic symmetry, whereas the CdS platelets used in our experiments are hexagonal. Nonetheless, the nature and magnitude of the anisotropy effects calculated by Keldysh *et al.*[16] will apply to hexagonal CdS because of the smallness of the splitting of the heavy- and light-hole valence bands at $\vec{k}=0$, namely, about 0.02 eV,[6,17] as compared to the band gap, 2.5 eV. From the structure of perturbation theory we would expect the magnitude of the anisotropy effects calculated for the cubic-symmetry case to be modified for the CdS case only by the multiplicative numerical factor: 1 – (energy of splitting)/(band-gap energy) $\simeq 0.99$. Further, the Franz-Keldysh effect was directly measured in CdS and CdSe by Gutsche and Lange[18] and they obtained a polarization anisotropy in the Franz-Keldysh coefficient $\gamma = -\Delta E/\mathcal{E}^2$ (with ΔE the shift in the absorption curve in eV, and $\mathcal{E}$ the applied electric field) of the same magnitude as that seen in our optical modulation measurements.

If the Franz-Keldysh effect is indeed the source of our observed optical modulation, with its polarization anisotropy, it remains to establish the presence of the requisite electric fields in the domain. The space-averaged steady electric field intensity in a typical acoustoelectric domain in our experiments was approximately 5×10^4 V/cm (the same value measured by Many and Balberg[19] in their studies of domains in CdS). This estimate is based on an excess voltage drop of 500 V across a 100-μ-wide domain. This field gives a Franz-Keldysh shift in the absorption edge of about 4 meV, which is only five times smaller than the observed shift. If our data were the only data available on optical modulation near the band edge by acoustoelectric domains in piezoelectric semiconductors, we would not rule out the steady electric field in the domain as the source of a Franz-Keldysh modulation. However observations by others make this highly unlikely. Kumar and Hutchinson[3] noted two properties of the optical modulation by domains in their samples of CdS which convincingly rule out the steady domain electric field as the effective agent: (i) The optical modulation steadily increases (in a particular case, by a factor of 5) *after* the current has reached saturation. Since the domain width does not change substantially after current saturation is reached, neither does the steady field across the domain, this being given by the overvoltage (i.e., excess voltage above threshold) divided by the domain width. On the other hand, the acoustic flux in the domain does continue to grow after current saturation is reached.[3,19] This spatial growth characteristic of the optical modulation requires a relatively homogeneous crystal for its observation. In our experiments this effect was masked by resistivity inhomogeneities which produced corresponding inhomogeneities in acoustic gain which dominated the generation of acoustic flux. (ii) The optical modulation was still observed many μsec after the applied voltage pulse

was turned off and there was no detectable current flowing through the sample, and concomitantly there was only a small average field across the domain. Here, as in (i), the optical modulation correlates with the acoustic flux in the domain, which decays much more slowly than the average field across the domain after voltage turnoff.[19] The situation is even more clear cut with the two piezoelectric semiconductors GaSb[1] and GaAs.[2] Domain-induced optical modulation with properties very similar to those seen in CdS are also seen in these two materials. However, the measured steady electric fields in the domains were as small as a few hundred V/cm—far too small to explain the observed modulation on the basis of the Franz-Keldysh effect. As a direct check, higher fields were applied to the same samples, under conditions inhibiting domain formation, and no modulation was observed, i.e., the Franz-Keldysh effect was unobservably small. Thus it is quite certain that the domain-induced optical modulation seen in GaSb and GaAs is not due to the steady electric field across the domain. It would be very surprising if the mechanism underlying the modulation were not the same for all three materials.

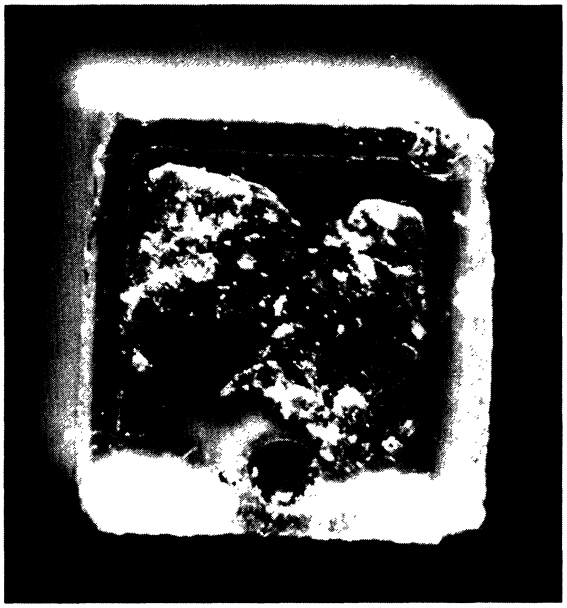
We propose here that the observed optical modulation is indeed due to the Franz-Keldysh effect, but that the effect is produced by strongly fluctuating electric fields inside the domain associated with the intense flux of high-frequency (0.1–1 GHz) phonons in the domain. There is considerable evidence for the presence of such fluctuating fields. Moore[20] measured the spectral distribution of the noise power in the current through a semiconducting CdS crystal in the range 2–800 MHz, both below and above current saturation. The fluctuations (noise power) increase enormously above saturation. At low frequencies the spectrum is flat and then falls off as $1/f^2$, the 3-dB down point occurring in the range 20–50 MHz. With the average current saturated, the noise power per unit bandwidth in the flat portion of the spectrum was typically 60 dB above Johnson noise in the equivalent resistor, 30–40 dB above shot noise for the measured average current and 50 dB above the generation-recombination noise associated with carrier trapping. Dependence of the noise spectrum on applied voltage, light level, temperature, and crystal length for both semiconducting and photoconducting CdS crystals was successfully reproduced by a model assuming strong electron bunching in the locally generated incoherent high-frequency phonon field. A single free parameter in the model, namely, the volume of the carrier bunch or microdomain, was determined to be one cubic wavelength, corresponding to a phonon frequency of about 500 MHz. A coherence time for a bunch of about five periods ($5/f$) was found. The strong electron bunching which is at the heart of Moore's model can be brought about only through the action of intense fluctuating electric fields, namely, fields much larger than the average steady field in the domain.

Although Moore's noise measurements establish unequivocally the presence of large fluctuating fields inside the domain, Moore's model is too schematic to yield the magnitude of the peak field. Here we turn to other measurements. The strain and the electric field are related by

$$S_4 = d_{24}\mathcal{E}_2 , \tag{9}$$

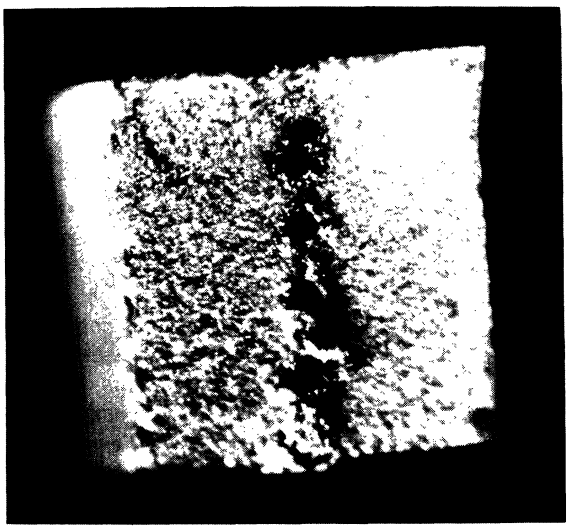
where S_4 is the shear strain and d_{24} is the relevant piezoelectric tensor element.[21] S_4 in a mature acoustoelectric domain in CdS was estimated by Kuzmany and Liederer,[22] from birefringence measurements, to be about 4×10^{-3}. That strains of this magnitude, or larger, exist inside a mature domain in CdS can be independently inferred from the common experience of those, including ourselves, who have studied such domains, namely, that the crystal is often subject to mechanical damage after repeated passage of a domain.[3,7,23] Photographs of such damage are reproduced in Fig. 9. The coefficient $d_{24}(=d_{15})$ was measured by Berlincourt *et al.*[24]: $d_{24}=1.4\times10^{-11}$ C/N. Inserting this value into (9), we find for the electric field $\mathcal{E}_2$ accompanying the strain S_4, $\mathcal{E}_2\approx3\times10^6$ V/cm. Although this is a crude estimate, because strain is not a smooth function of position inside the domain but rather has the incoherence features of Moore's electron-bunching model,[20] nonetheless it does establish that the fluctuating electric fields in the domain, associated with the fluctuating high-frequency strain, are indeed an order of magnitude (or more) stronger than the steady field in the domain. The fluctuating fields in mature domains in CdS are clearly large enough to produce Franz-Keldysh shifts of the observed magnitude. Further, since these fluctuating fields are associated with shear phonons traveling in a narrow beam in the drift-field direction [for the orientations in Figs. 3(a) and (b)], as shown by Brillouin-scattering measurements,[9] the fluctuating fields are also in the drift-field direction, and the polarizational dependence of the Franz-Keldysh effect is just as described above.

There is yet another class of experimental observations which make sense only if one accepts the presence of the very strong fluctuating electric fields in the domain, and that is the near-band-gap (red unpolarized) light emission associated with the domain at room temperature, seen by us and other workers.[3,7,9] Ordinarily this light emission is seen at the anode. However, Yee[9] fabricated a sample which had a massive anode contact so that the domain disappeared before reaching the contact; still, he saw light emission from the thin sec-



(a)

(b)



(c)

FIG. 9. Photographs of damage produced by acoustoelectric domains in single crystals of CdS. (a) End-on view of the anode showing hole at the bottom. Note deposited material over an extensive area. (b) Side view of same crystal as in (a). Note single long hole [side view of hole seen in (a)] and additional smaller holes and cracks. (c) End-on view of the anode (another crystal) showing row of holes.

tion where acoustoelectric domains had been. It is impossible to see how acoustic strain of *macroscopic* dimensions, namely, with a wavelength measured in μ can *directly* produce an electronic excitation of order 2 eV, which is necessary to account for the observed emission. On the other hand, the fluctuating electric fields induced by the large strains are strong enough to produce that amount of excitation. Fields in the range $(0.3-1.0)\times10^5$ V/cm are sufficient to produce field emission of holes from localized states several tenths of an eV above the valence band in photoconducting CdS.[25] Fields in the range $(1.0-2.5)\times10^6$ V/cm cause breakdown of a Schottky barrier on semiconducting *n*-type CdS.[26] The breakdown is due to field emission of electrons from surface states more than 1 eV below the conduction band. Finally, there is a vast literature reviewed by Henisch and Ivey[27] on electroluminescence (EL) in ZnS (both powders and single crystals) which has an even larger band gap, namely, 3.5 eV. Here again, excitations exceeding 2 eV, corresponding to visible light emission, are produced by electric fields in the range 10^5-10^6 V/cm. Finally, we mention that strong hot-carrier effects (impact avalanching across the band gap) have been seen in acoustoelectric studies of *n*-InSb.[28] The electric fields involved, ≈ 200 V/cm, are too small, however, to allow useful extrapolations to the CdS studies.

B. Deformation Potentials

It is well known that the application of a strain to a semiconductor changes the band structure. The principal effect is to shift the energy of each band maximum or minimum. In the III-V compounds, a shear strain can split the degeneracy at the valence-band maximum, resulting in a shift in the optical-absorption edge. However, in a hexagonal crystal such as CdS, the valence band is already completely nondegenerate (except for spin degeneracy), as already remarked in Sec. VII A). A strain consisting of an off-diagonal tensor element in the principal coordinate system cannot change the energy of the valence-band maximum or the conduction-band minimum *to first order*, and the strains encountered in the domains are of such an orientation. Since the deformation potentials associated with uniaxial strains in CdS are on the order of 2 eV,[17] a uniaxial strain of 4×10^{-3} would cause band motion of 8 meV. A shear strain of

this same magnitude which produces *only a second-order effect* may be expected to produce much smaller band motion, and it is not even clear that the net effect would be an increase in the absorption coefficient. Furthermore, band motion cannot account for the polarization asymmetry seen in the orientation in Fig. 3(a). Thus, band motion due to deformation potentials does not appear to be a likely mechanism here.

C. Self-Energy Effect and Uncertainty Broadening

The electrons in the conduction band and the holes in the valence band have self-energies due to interaction with the quantized lattice vibrations (phonons). It is well known that this self-energy accounts for the largest part of the temperature dependence of the band gap in most of the common semiconductors, and it has been suggested[1] that the additional self-energy due to interaction with the high-frequency domain phonons may be responsible for the modulation of the absorption edge. However, the nature of the interaction is such that phonons whose wavelengths are long compared with the mean free path of the carriers do not contribute significantly to the electron and hole self-energies.[12] These phonons interact with the carriers only through their deformation potentials. Since the energy spectrum of phonons in acoustoelectric domains in CdS is known to be in the frequency interval below 1000 MHz,[7] and this frequency corresponds to an acoustic wavelength of about 2 μ (two orders of magnitude longer than the mean free path of the carriers), the electron-phonon interaction would not be an effective mechanism by which the domains cause the band edge to change.

It has also been suggested[1] that the high density of phonons in an acoustoelectric domain causes the average collision time for the carriers to be shorter than in the normal material, and that this could result in an uncertainty broadening of the band edge. However, considerations involved in this mechanism are similar to those for the self-energy effect. The long-wavelength phonons are not effective at causing scattering, and it is reemphasized that the domain phonons have wavelengths two orders of magnitude longer than the original mean free path of the carriers. Viewed in another way, the mean time between collisions in the absence of domains is already orders of magnitude shorter than the period of the phonons in the domain. Thus, it is difficult to see how uncertainty broadening could contribute to the modulation. Furthermore, neither self-energy nor uncertainty broadening can account for the polarization asymmetry noted above.

D. Photoelastic Scattering

One of the experiments reported here involved measuring the change in the modulation caused by an acoustoelectric domain when a second polarizer was inserted into the optical path. The results of this experiment demonstrated that there was very little small-angle scattering and implied that there was very little total scattering. A second cogent reason for believing that photoelastic scattering is not responsible for the modulation is based upon the spectral dependence of the modulation. It is known[29] that the intensity I_s of photoelastically scattered light satisfies the relation

$$I_s = \text{const} \times n^6 p_{44}^2 |\lambda^2 , \tag{10}$$

where n is the index of refraction, p_{44} is the relevant elasto-optic coefficient, and λ is the optical wavelength. Since it was reported that certain elasto-optic coefficients of CdS increase drastically near the band edge,[30] the possibility that scattering could be responsible for modulation, which also increases near the band edge, merited investigation. An analysis of the behavior of the elasto-optic coefficients near the band edge of CdS was reported separately[31] and it was shown that while the elasto-optic coefficients associated with longitudinal strains do indeed increase drastically near the band edge, the coefficients associated with shear strains actually decrease according to the relation

$$p_{44} = \text{const}/n^4(\lambda) . \tag{11}$$

Using this expression, the anticipated spectral dependence of the scattered light becomes

$$I_s = \text{const}/n^2\lambda^2 . \tag{12}$$

This quantity is relatively constant in the vicinity of the band edge. While the simplifying assumptions that went into the derivation of Eq. (11) make Eq. (12) an approximation, the essential point of the argument is that the large positive dispersion seen by Tell *et al.*[30] is not present in the *shear* elasto-optic coefficient p_{44} and this is the coefficient relevant to the domains. Indeed, the small contribution of scattering to the modulation appeared to be nondispersive to the extent to which it could be accurately measured. Therefore, photoelastic scattering can be ruled out as a mechanism responsible for the modulation on the basis of the measured spectral variation of the modulation.

E. Other Observations

In comparing the results of this experiment with those previously obtained by the same technique for GaSb and GaAs, the differences appear to be primarily due to the much higher acoustic gain in CdS and the much narrower domains found in this material. The domain growth time in the III-V compounds was a large fraction of the domain transit time, making the study of domain growth possible. In CdS, domain growth takes place over very

short distances, with the result that the domain growth is easily confused with domain variation caused by the large variation in acoustoelectric gain over short distances. The widths of the domains in the III-V compounds were sufficient to permit the study of the domain shape, but in CdS this was very difficult. We observed domains traveling at about 1.75×10^5 cm/sec, the speed of shear waves, and their intensities varied with the applied pulse voltage in a reasonable manner. The domain widths were within the expected range and were observed to increase with drift voltage. Variation of domain strength with position was measured, and domain reflections from the anode were seen. These results are in accord with those published by Kumar and Hutchinson[3] and therefore we do not elaborate on them here.

VIII. CONCLUSION

The modulation of the optical-absorption edges of CdS in the presence of acoustoelectric domains has been studied with emphasis on the spectral and polarizational dependences of the modulation. Several interesting effects were noted:

(i) The change in the absorption coefficient caused by domains was observed to increase strongly for wavelengths approaching the band edge. This is consistent with the idea of the domain causing the band edge to shift toward longer wavelengths.

(ii) When the light was propagated along the optic axis of the crystal, the modulation was found to be largest with the polarization along the drift-field direction, in agreement with the results previously reported for GaSb.

(iii) When the light was propagated in a direction perpendicular to the optic axis, the polarizational dependence of the modulation was explainable as a superposition of the effects of the double absorption edge of CdS and of the angle between the polarization and the drift field as above.

All of the observed properties of the modulation are well explained by the Franz-Keldysh effect. However the electric field producing this effect is not the steady electric field across the domain. Several converging lines of evidence (obtained in studies of CdS and discussed in detail in Sec. VII A) indicate that the much larger fluctuating electric fields associated with the intense high-frequency phonon field inside the domain produce the observed Franz-Keldysh shifts. All other proposed mechanisms are either highly unlikely or completely impossible.

It would certainly be of great interest to study the strong fluctuating electric fields in other piezoelectric semiconductors, namely, GaSb, GaAs, and InSb, which have weaker coupling than CdS and thereby more controllable domains. A refinement of Moore's noise model, enabling the extraction of electric field information from current noise measurements, would also be very useful.

ACKNOWLEDGMENTS

The authors are pleased to acknowledge extensive assistance from A. R. Moore, of the RCA Laboratories, in planning the experiments. They would also like to thank P. J. Messineo, B. E. Tompkins, and H. D. Hanson of the RCA Laboratories for technical assistance, and R. W. Smith and I. Balberg of the RCA Laboratories and J. Dow of Princeton University for helpful conversations. In addition, they are indebted to R. W. Smith for supplying his striking photographs of crystal damage induced by acoustoelectric domains (Fig. 9).

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†Work based in part on a thesis submitted by E. S. Kohn to Princeton University in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

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conductors (Dunod, Paris, 1964), p. 129. These authors reported measurements on the Franz-Keldysh effect in CdS and CdSe platelets for four orientations of applied field and light polarization. However, since these platelets contain the c axis and the light must propagate normal to the platelets, the important additional case in which the applied field is in a direction perpendicular to c and the optical electric field is in a direction perpendicular to both the applied field and to c could not be measured. A value for the Franz-Keldysh coefficient γ for this case would enable a direct comparison with the polarization asymmetry in our data for the orientation in Fig. 3(a). Fortunately, the magnitude of γ for this case can be inferred from those measurements the authors did make. Their data show that with the drift field parallel to the optical field, the coefficient $\gamma = -\Delta E/\mathcal{E}^2$ was 1.58 or 1.62(10^{-12} eV cm^2/V^2) with the fields parallel to or perpendicular to the c axis, respectively. With the two electric fields perpendicular to each other, the coefficient was 1.08 or 1.23, depending on the orientation of the c axis. The last two numbers were even closer (1.84 and 1.88) for CdSe. Thus, it appears that while the orientations of the applied field and the optical field with respect to the c axis are relatively unimportant, the angle between the two fields is important, with the Franz-Keldysh effect about 40% larger when the fields are parallel. This agrees well with our measurements on acoustoelectric domains, where the shift with the fields parallel was about 30% larger.

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PHYSICAL REVIEW B VOLUME 4, NUMBER 12 15 DECEMBER 1971

Phonon Conductivity of InSb and GaAs in the Temperature Range 2-300 °K

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(Received 24 May 1971)

The recent modification of Holland's model of two-mode conduction, as proposed by us, has been applied to explain the phonon conductivity of InSb and GaAs. This model, known as the Sharma-Dubey-Verma model, makes use of Guthrie's classification of three-phonon scattering events. In this model, the exponent m of the temperature, i.e., $T^{m(T)}$ is a continuous function of temperature and approaches unity in the high-temperature region for both the longitudinal phonons as well as transverse phonons. The dispersion of acoustic branches is taken into account in replacing v_g/v_p^2 in the conductivity integrals and this forms the basis of the division of the conductivity integrals for the different polarization branches. The present model gives excellent agreement between the theoretical and experimental values of phonon conductivity except near the maximum where the scattering of phonons by point defects dominates over phonon-phonon scattering as well as the boundary scattering of phonons.

INTRODUCTION

Recently we have proposed a modification[1] of Holland's[2] model of two-mode conduction in semiconductors. The most significant feature of the present model [hereafter called the Sharma-Dubey-Verma (SDV) model] is the use of Guthrie's[3] classification on three-phonon scattering events. In class-I events the carrier phonon is annihilated by combination and in class II the annihilation takes place by splitting. Thus τ_{3ph}^{-1} is expressed as

$$\tau_{3ph}^{-1} = \tau_{3ph}^{-1}(\text{class I}) + \tau_{3ph}^{-1}(\text{class II}) .$$

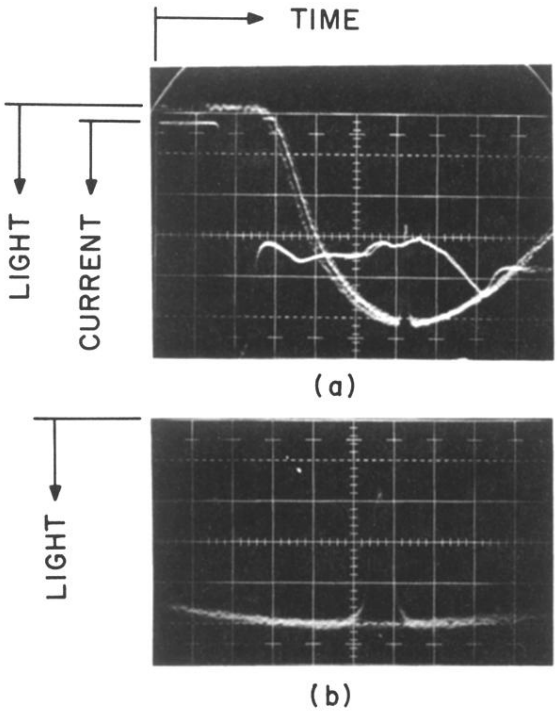


FIG. 2. Transmission modulation observed with the pulsed lamp. (a) Traces of sample current (0.3 A/div) and optical signal (relative magnitude) as a function of time (0.5 μsec/div). (b) Expanded view of a similar, but stronger, optical signal (0.1 μsec/div). The drift field was perpendicular to the c axis; the light propagated along the c axis and was polarized along the drift-field direction, as in Fig. 3(a).

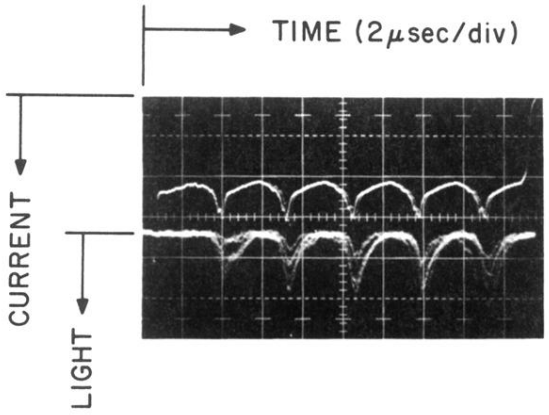
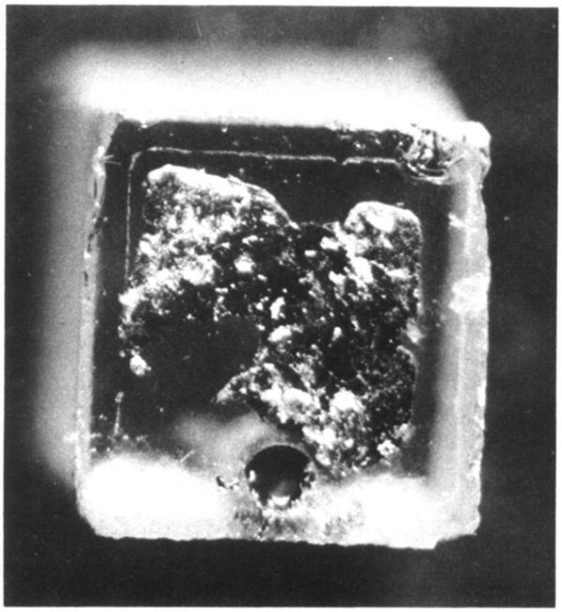
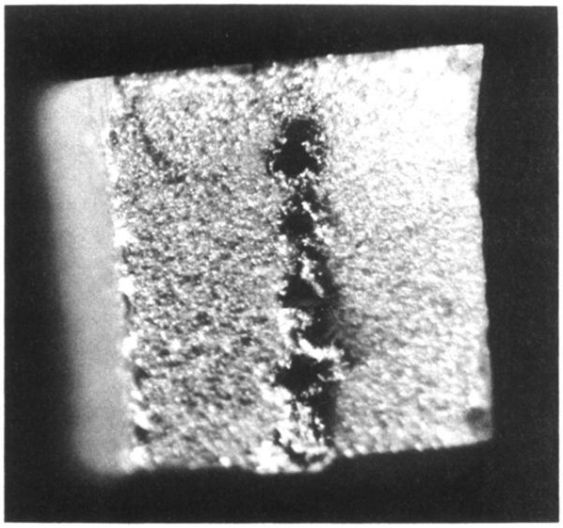


FIG. 8. Domain-related light emission from the anode region. The current (0.3 A/div) and the emitted light (relative magnitude) are compared as a function of time (2 μsec/div). The drive pulse was long enough to permit several domain transits.



(a)

(b)



(c)

FIG. 9. Photographs of damage produced by acousto-electric domains in single crystals of CdS. (a) End-on view of the anode showing hole at the bottom. Note deposited material over an extensive area. (b) Side view of same crystal as in (a). Note single long hole [side view of hole seen in (a)] and additional smaller holes and cracks. (c) End-on view of the anode (another crystal) showing row of holes.