tailed study of a Lifshitz transition.

We are indebted to R. J. Baughman for growing the single crystal ingots from which the samples were cut, to R. L. White for technical assistance, and to A. Narath for valuable comments on the manuscript.

---

†Work supported by the U. S. Atomic Energy Commission.

[1]I. M. Lifshitz, Zh. Eksp. Teor. Fiz. 38, 1569 (1960) [Sov. Phys. JETP 11, 1130 (1960)].

[2]L. D. Jennings and C. A. Swenson, Phys. Rev. 112, 31 (1958).

[3]C. W. Chu, T. F. Smith, and W. E. Gardner, Phys. Rev. B 1, 214 (1970).

[4]V. I. Makarov and I. Ya. Volynskii, Zh. Eksp. Teor. Fiz., Pis'ma Red. 4, 369 (1966) [JETP Lett. 4, 249 (1966)].

[5]V. I. Makarov and V. G. Bar'yakhtar, Zh. Eksp. Teor. Fiz. 48, 1717 (1965) [Sov. Phys. JETP 21, 1151 (1965)].

[6]E. S. Itskevich and A. N. Voronovskii, Zh. Eksp. Teor. Fiz., Pis'ma Red. 4, 226 (1966) [JETP Lett. 4, 154 (1966)].

[7]E. S. Itskevich and L. M. Fisher, Zh. Eksp. Teor. Fiz., Pis'ma Red. 6, 748 (1967) [JETP Lett. 6, 219 (1967)].

[8]R. J. Higgins and H. D. Kaehn, Phys. Rev. 182, 649 (1969).

[9]M. G. Priestley, L. R. Windmiller, J. B. Ketterson, and Y. Eckstein, Phys. Rev. 154, 671 (1967).

[10]P. J. Lin and L. M. Falicov, Phys. Rev. 142, 441 (1966).

[11]D. Schiferl and C. S. Barrett, J. Appl. Crystallgr. 2, 30 (1969).

[12]N. G. Pace, G. A. Saunders, and Z. Sümengen, J. Phys. Chem. Solids 31, 1467 (1970).

[13]B. Morosin and J. E. Schirber, Phys. Lett. 30A, 512 (1969).

[14]M. Maltz and M. S. Dresselhaus, Phys. Rev. 182, 741 (1969).

[15]A. P. Jeavons and G. A. Saunders, Brit. J. Appl. Phys. 1, 869 (1968).

[16]R. W. Stark and L. R. Windmiller, Cryogenics 8, 272 (1968).

[17]I. M. Templeton, Proc. Roy. Soc., Ser. A 292, 413 (1966).

[18]J. E. Schirber and W. J. O'Sullivan, Phys. Rev. 184, 628 (1969).

[19]J. E. Schirber, Cryogenics 10, 418 (1970).

[20]N. B. Brandt, N. Ya. Minina, and Yu. A. Pospelov, Zh. Eksp. Teor. Fiz. 55, 1656 (1968) [Sov. Phys. JETP 28, 869 (1969)], and references therein.

[21]J. E. Schirber and W. J. O'Sullivan, to be published.

[22]I. M. Lifshitz and A. M. Kosevich, Zh. Eksp. Teor. Fiz. 29, 730 (1955) [Sov. Phys. JETP 2, 636 (1956)].

Magnetic-Field–Induced Resonance in Raman Scattering from CdS

T. C. Damen and Jagdeep Shah
Bell Telephone Laboratories, Holmdel, New Jersey 07733
(Received 28 December 1970)

We have observed magnetic-field–induced resonance in Raman-scattering cross sections. The resonance enhancement for 1LO scattering from CdS, obtained with 4765 Å, is a factor of ~30 at 90 kG. The results are interpreted in terms of resonance of the *incident* photon with the Landau levels of the Γ_7 conduction and valence bands. With a 4579-Å laser, we see a resonance of the Stokes 3LO process. We explain this as the resonance of the *scattered* photon.

We report the observation of a thirtyfold increase in the 1LO Raman-scattering cross section in CdS excited at 4765 Å, when a magnetic field (~90 kG) is applied to the sample. We have also observed that the cross section for the 3LO scattering from CdS, obtained with a 4579-Å laser, increases by about a factor of 2.5 as the magnetic field is increased from 0 to 100 kG. We show that these results can be explained in terms of the resonance of the incident or the scattered photon with the band gaps of CdS as modified by the applied magnetic field. Although resonant Raman scattering (RRS) has been investigated extensively in the last two years,[1] this is the first report of resonance caused by a magnetic field.

Experiments were carried out by a backscattering technique using an Ar^+ laser with ~100 mW in 4765- and 4579-Å laser lines. Spectra from several pure CdS platelets were measured at 2°K and 0 kG. Platelets in which the luminescence did not interfere strongly with the Stokes 1LO line (obtained with 4765-Å laser) were selected for the experiments with magnetic field.

The platelets, typically ~2 μm thick, were im-

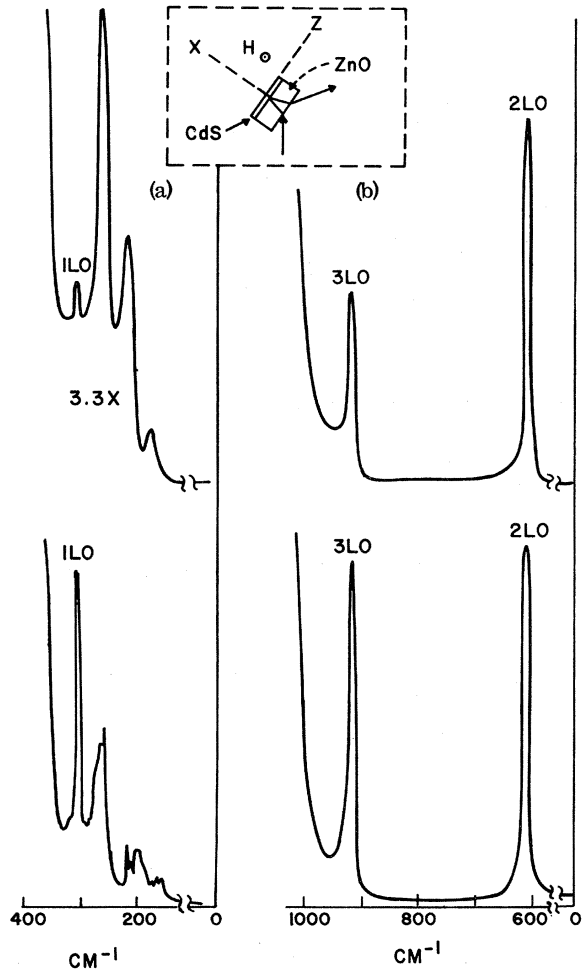

FIG. 1. (a) Stokes 1LO Raman scattering spectrum from CdS at 2°K for $H=20$ kG (85 kG) in upper (lower) traces. $H\perp C$ axis of crystal. $X(Z_Z^Y)\bar{X}$ geometry for both traces. Obtained with 4765-Å laser. (b) Raman spectra from CdS at 2°K obtained with 4579-Å laser. Upper (lower) trace corresponds to $H=0$ kG (95 kG). $X(Z_Z^Y)\bar{X}$ geometry for both traces. Other structure is due to luminescence. $H\perp C$.

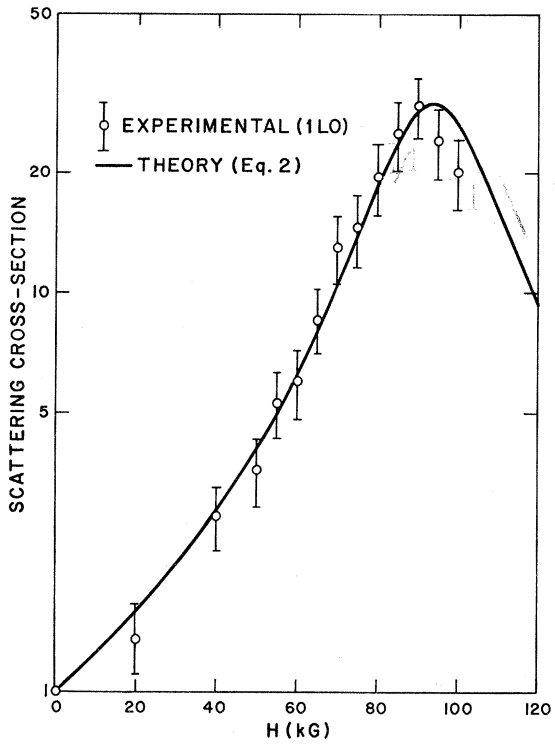

FIG. 2. Stokes 1LO scattering intensity (with 4765 Å) from CdS at 2°K. The points are experimental. The solid curve is calculated from Eq. (2) in the text. $\Gamma=0.6$ meV; $m_e^*=0.2m_0$, $m_\parallel=5.0m_0$, $m_\perp=0.7m_0$, where m_0 is the free-electron mass. We have assumed that the hole masses in the A and B bands are the same. Data normalized at $H=0$.

mersed in liquid He which was pumped to 2°K A 100-kG superconducting solenoid was used. The scattered spectra were analyzed using a double spectrometer. Scattering from the E_2 mode of ZnO, placed in front of CdS (see insert in Fig. 1), was used to monitor the collection efficiency of the system.

In Fig. 1(a), we show the spectra obtained with the 4765-Å laser for two different magnetic fields. One immediately notices a large increase in the scattering intensity of 1LO phonons with increase in the magnetic fields. In Fig. 2 we have plotted the 1LO Stokes-scattered intensity as a function of magnetic field. We notice that the intensity increases by approximately a factor of ~30 between 0 and 90 kG. As the field increases above 90 kG, the intensity begins to decrease.

This large increase in scattering intensity of 1LO phonons could be due to (i) an increase in the scattering cross section and/or (ii) a reduction in the absorption coefficient at 4765 Å caused by the applied magnetic field. In order to evaluate the relative importance of these two factors, we have measured the absorption of these CdS platelets as a function of photon energy and magnetic field. These results will be presented in detail elsewhere.[2] From these absorption measurements, we have calculated that the reduction in absorption caused by the applied magnetic field (up to 100 kG) will increase the scattering intensity by less than 20% of its zero-field value. This is clearly negligible compared to the observed increase of a factor of ~30. We conclude therefore that the observed effect is due to an increase in the scattering cross section for the 1LO process. In what follows, we show this increase is almost entirely due to the resonance

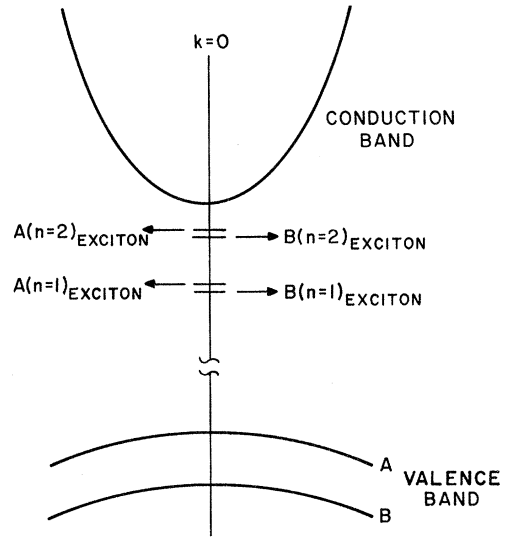

FIG. 3. Relevant energy-level diagram of CdS.

of the incident photon with the B band gap (Γ_7-Γ_7) of CdS, as modified by the magnetic field.

The cross section σ for Raman scattering from phonons has the form[3]

$$\sigma \sim \left| \sum_{I_1,I_2} \frac{A_{I_1,I_2}}{(\omega_i-\omega_{I_1})(\omega_s-\omega_{I_2})} \right|^2, \quad (1)$$

where ω_i (ω_s) is the incident (scattered) frequency and $\hbar\omega_{I_1}$ ($\hbar\omega_{I_2}$) is the energy of the intermediate state I_1 (I_2). A_{I_1,I_2} depends on the strength of the electron-photon and electron-phonon interactions and constants characteristic of the solid under consideration. We assume that the applied magnetic field does not change any of these significantly so that change in A_{I_1,I_2} can be neglected. Then the change in the cross section must come from the resonant denominator through change in ω_I.

We will now simplify the problem by assuming that only the following intermediate states need be considered: (i) the $n=1$ and $n=2$ A exciton, (ii) the $n=1$ and $n=2$ B exciton, (iii) the A (Γ_9-Γ_7) band gap, and (iv) the B (Γ_7-Γ_7) band gap. The relevant energy-level diagram is shown in Fig. 3. The associated energies and energy denominators are given in Table I. Let us first consider how the magnetic field affects these intermediate states. The shifts and the splittings of the A and the B excitons, as determined by Hopfield and Thomas,[4] are small compared to ω_i-ω_I and ω_s-ω_I. Therefore the excitons can be neglected in our case.[5] The valence and conduction bands are expected to be split into a series of Landau levels[6] $E(B)=E(0)+(n+\frac{1}{2})\hbar\omega_c$ for $k=0$ where $\omega_c=eB/m^*$ and m^* is the carrier effective mass in the band. The changes in the intermediate-state energies produced by the magnetic field [i.e., $|\omega_I(0)-\omega_I(B)|$] are small compared to the difference between the scattered frequency ($\omega_s=20\,675.5$ cm^{-1}) and $\omega_I(0)$. Hence the scattered-photon pole can be neglected in this case. Thus the observed increase in scattering cross section must be due to the resonance of the incident photon with A and B transitions.

We can distinguish between the contributions from A and B transitions by using the known selection rules[4] for optical transitions. Both the A and B transitions will contribute to σ in the $X(Y_Z{}^Y)\bar{X}$ geometry but only the B transition will contribute to $X(Z_Z{}^Y)\bar{X}$ geometry. Experimentally we find that the enhancement in the scattering cross section up to 100 kG is nearly the same for both geometries. This indicates that even in

Table I. Energy denominators for various intermediate states. Values are taken from Hopfield and Thomas (Ref. 4) and Segall and Marple (Ref. 9). $\hbar\omega_\mu$ is defined in the text. We have assumed identical effective masses for holes in A and B valence bands. In the last column, $\omega_s=\omega_i-3\omega_{LO}$ and ω_i corresponds to 4579 Å.

Intermediate state, ω_I	$(\omega_I-\omega_i)^{-2}$ for ω_i $=20\,980.5$ cm^{-1} (10^{-6} cm^2)	$(\omega_I-\omega_s)^{-2}$ for ω_s $=20\,917.7$ cm^{-1} (10^{-6} cm^2)
$A(n=1)$ exciton 2.5537 eV 20 594.3 cm^{-1}	6.7	9.6
$A(n=2)$ exciton 2.5758 eV 20 772.6 cm^{-1}	23.2	47.5
$A(\Gamma_9$-$\Gamma_7)$ band gap $E_A=2.5831$ eV 20 830.6 cm^{-1}	44.5	132
$B(n=1)$ exciton 2.5686 eV 20 714.5 cm^{-1}	14.1	24.4
$B(n=2)$ exciton 2.5908 eV 20 893.6 cm^{-1}	132	1730
$B(\Gamma_7$-$\Gamma_7)$ band gap $E_B=2.5986$ eV 20 956.5 cm^{-1}	1730	666
E_B(100 kG; $n=0$) $=E_B(0)+\frac{1}{2}\hbar\omega_\mu$ $=20\,982.1$ cm^{-1}	390 000	240
E_A(100 kG; $n=0$) $=E_A(0)+\frac{1}{2}\hbar\omega_\mu$ $=20\,856.2$ cm^{-1}	65	265

$X(Y_Z{}^{Y})\bar{X}$ geometry, the contribution of the A transition is negligible. Thus the increase in cross section is due to magnetic-field–induced resonance of the incident photon with the B band gap.

In order to make a quantitative estimate of the expected resonance enhancement, we will make the assumption[7] that only the transition between the lowest Landau levels of the Γ_7 conduction and valence bands at $k=0$ is important, i.e.,

$$\hbar\omega_I(B) = E_B(0) + \tfrac{1}{2}\hbar eB[1/m_e{}^* + 1/(m_\parallel m_\perp)^{1/2}]$$
$$= E_B(0) + \tfrac{1}{2}\hbar\omega_\mu$$

for $B \perp C$, where $m_e{}^*$ is the electron effective mass and $m_\parallel$ ($m_\perp$) is the hole effective mass parallel (perpendicular) to the crystal C axis. Then

$$\frac{\sigma(B)}{\sigma(0)} = \frac{[\hbar\omega_i - E_B(0)]^2 + \Gamma^2}{[\hbar\omega_i - E_B(0) - \tfrac{1}{2}\hbar\omega_\mu]^2 + \Gamma^2}. \quad (2)$$

In this equation we have introduced a phenomenological damping constant $\Gamma = \hbar/\tau$, where τ is the effective carrier scattering time. The magnetic field at which the maximum in $\sigma(B)/\sigma(0)$ occurs is independent of Γ. From Eq. (2) and Table I, the maximum is expected at ~93 kG. This is in agreement with our experimental observation. The magnitude of $\sigma(B)/\sigma(0)$ does depend on Γ. The best agreement with the experimental data is obtained for $\Gamma = 0.6$ meV. The calculated curve for $\Gamma = 0.6$ meV is shown in Fig. 2 along with the experimental points. One can see that there is a good agreement between the two. The value of Γ (=0.6 meV) obtained in this manner is also in agreement with our magnetoabsorption data in CdS.[2] This provides a strong support for our interpretation.

It should be emphasized that the calculation is not exact. But a rigorous theory, starting from the first principles and including such effects as electron-hole Coulomb interaction, actual shape of the density of states in the presence of magnetic field, etc., is clearly outside the scope of this communication. The intent here is to present a simple physical model which can be quantitatively compared with the experimental results.

Finally, we briefly discuss the 3LO and 2LO scattering processes obtained with a 4579-Å laser. Figure 1(b) shows two traces at two different magnetic fields. We find that the 2LO process remains constant between 0 and 100 kG. In the same range, the 3LO process monotonically increases by a factor of ~2.5. Thus, in this case, the magnetic field has brought about a resonance with the scattered photon.[8]

In conclusion, we have observed the first magnetic-field–induced resonance in Raman scattering and provided a simple model to explain it.

We would like to thank R. M. Martin and Van-Tran Nguyen for helpful discussions, J. P. Gordon and C. K. N. Patel for comments on the manuscript, and A. E. DiGiovanni for skillful technical assistance. We have also benefitted from discussions with R. C. C. Leite in the early stages of these experiments.

---

[1]See, for example, J. F. Scott, R. C. C. Leite, and T. C. Damen, Phys. Rev. 188, 1285 (1969), and references cited therein.

[2]J. Shah and T. C. Damen, to be published.

[3]R. Loudon, Advan. Phys. 13, 423 (1965).

[4]J. J. Hopfield and D. G. Thomas, Phys. Rev. 122, 35 (1961).

[5]The assumption implicit in this treatment is that at zero magnetic field the band-gap contribution to the scattering cross section is comparable to or larger than the exciton contribution, for the 4765-Å laser. This is quite reasonable in view of the differences in the energy denominators (Table I) and our unpublished data.

[6]See, for example, R. A. Smith, in *Semiconductors*, edited by N. B. Hannay (Cambridge Univ., Cambridge, England, 1959).

[7]The resonance with $n \geq 1$ Landau levels is expected to occur for $H \leq 30$ kG. Our magnetoabsorption data (Ref. 2) indicate that Landau levels are not resolved for $H \leq 30$ kG. This implies that strong singularities in the density of states are not present at such low fields, probably because of damping. A smoothly varying density of states does not give a large resonance enhancement. [R. Loudon, J. Phys. (Paris) 26, 677 (1965)]. Therefore, the assumption of neglecting Landau levels $n \geq 1$ is reasonable in our case.

[8]The observed resonant enhancement is a result of many opposing effects: The A band gap moves towards ω_s, whereas the B band gap moves away from it. The B ($n=2$) exciton, being very close to ω_s, may also play an important role.

[9]B. Segall and D. T. F. Marple, in *Physics and Chemistry of II-VI Compounds*, edited by M. Aven and J. S. Prener (Wiley, New York, 1967).