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Photon Induced Resonant Raman Scattering in CdS

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Abstract

A novel aspect of Resonant Raman Scattering is observed in CdS by means of the ratio of Stokes to anti-Stokes intensities. With increasing temperature, as the forbidden band energy approaches a value that is twice the incident photon energy, (from a Nd-Yag-laser) a large enhancement of the above ratio is observed for both the LO and the 2LO phonon Raman intensities. The results indicate a resonance with the scattered photon. Resonance is only observed for high incident photon intensities. A possible explanation for the above observations is that flooding of the crystal with photons of energy hν induces states of energy hν displaced from the electronic bands by mixing of electronic and photon states.

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Enhanced Raman Scattering cross-section in solids was first observed in CdS(1) where resonance was related to real electronic(2) (or excitonic(3)) states. The abundant subsequent experimental and theoretical study(4) have been restricted until now to systems in which resonance were due to real states. The result to be presented here indicates that resonance can be obtained with states induced by the presence of a large photon field.

CdS was selected for this experiment because its room temperature energy gap is slightly larger than twice the photon energy from a Nd: Yag laser. During the laser pulse power densities of the order of 10^8 Watts/cm^2 are obtainable in the crystal bulk. Rupture of the crystal occurred at such power densities. Convenient laser paths in the crsystal had to be selected on a trial and error basis. The small Raman cross sections and poor detection efficiency near 1.06μ precluded observation at powers significantly smaller than these for which rupture occurred. Experiments were performed in the typical 90° geometry and the sample was inside a temperature controlled resistence furnace.

The Nd: Yag laser operating at 1.06μ had a maximum power of 40 Watts when operation in the CW mode and 15 KW in pulsed operation ($1.5\mu s$ pulse duration a 1000 pulses per second). A 75 cm double monochromator and an S-1 photomultiplier followed by a box car integrator system were used for the present experiment.

Due to small scattering efficiencies and small collecting angles (because of the furnace geometry) it was impossible to introduce a reference material as employed in the case of CdS excited with an Argon laser line(5). Therefore an indication of resonance was searched for by means of the ratio of the Stokes to anti-Stokes intensities. As the incident radiation $h\nu_i$ approaches the energy of an electronic transition E_a both Stokes and anti-Stokes intensities are enhanced, however the anti-Stokes and Stokes cross sections should attain their maximum value at different incident frequencies according to the energy denominators of the deminant terms near resonance, i.e;

$(E_a - h\nu_i)\ [E_a - (h\nu_i - h\omega_0)]$ and $(E_a - h\nu_i)\ [E_a - (h\nu_i + h\omega_0)]$

respectively, where $h\omega_0$ is the phonon energy.

Scanning of the resonance region was obtained by increasing the temperature of the sample and thus continuously reducing its energy gap. At such high power densities, most probably, the local temperature at the laser beam path is higher than the furnace temperature.

Fig. I give the ratio of anti-Stokes to Stokes intensities (A/S) as a function of the furnace temperature for both the one and two longitudinal optical phonon processes. Similar results were obtained in several different runs. Persuasive evidence exists for a resonance point followed by an anti-resonance point near mid band energies for both LO and 2LO lines. Indeed theoretically one should expect for the A/S ratio a cusp near $E_a - h\nu_i + h\omega_0$ followed by a depression near $E_a - h\nu_i - h\omega_0$. We may therefore conclude that a characteristic energy E_a exists near the mid band gap. Furthermore the above anomalies completely disappears in a C. W. experiment for which the beam intensity is ~ 10^3 times smaller. This indicates that E_a is induced by the high density of photons in the pulsed experiment. Another positive aspect of results in Fig. I is that the cusp of A/S for the 2LO scattering occurs at lower temperatures as expected. The depression however is not very well defined. The main drawback thought of the present experiment is the inherent lack of control of the local temperature at the beam path. Since this is the significant temperature, no quantitative values can be inferred from this parameter in Fig. I.

A possible explanation for the emergence of E_a is though mixing of conduction (or valence) electronic band states inducing a band displaced by the photon energy $h\nu_i$ from the original electronic (or excitonic) states. Such a density of states would emerge if the crystal were flooded by photons of a energy $h\nu_i$ and resonance obtained with these displaced states. The appropriate manner to execute this experiment would use a high power laser with frequency $h\nu_a$ to induce the states E_a and a tunable laser of frequency $h\nu_i$ to scan the region $E_g \quad h\nu_a$ (E_g is the semiconductor energy gap). The above results, preliminary though, are forwarded here in the hope of stimulating experimental and theoretical work that might bring about some light on this interesting effect which may become a simple tool to study mixing of electronic and photon states.

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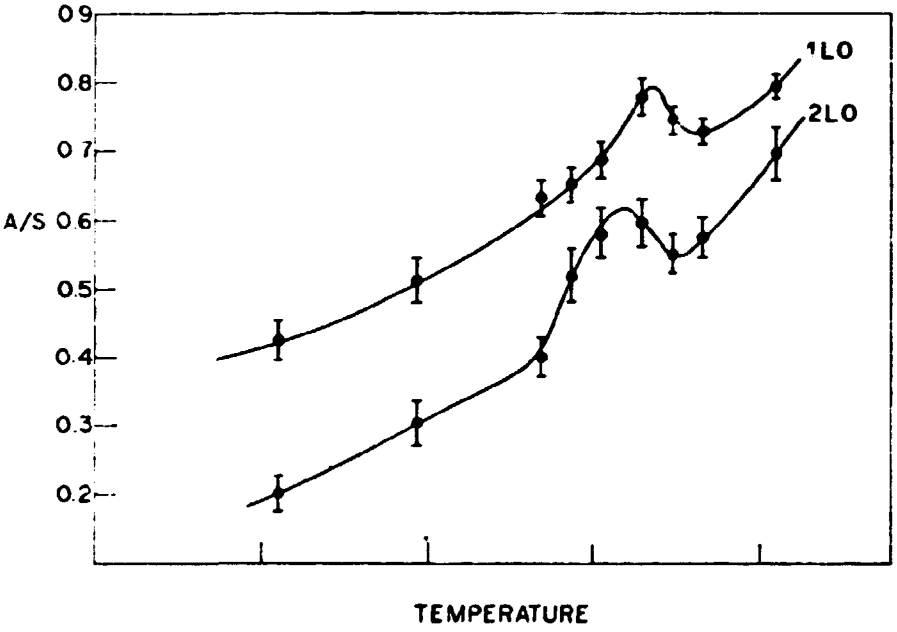

Fig.: 1 Ratio of anti-Stokes to Stokes intensities as a function of the furnace temperature for LO and 2LO phonons.