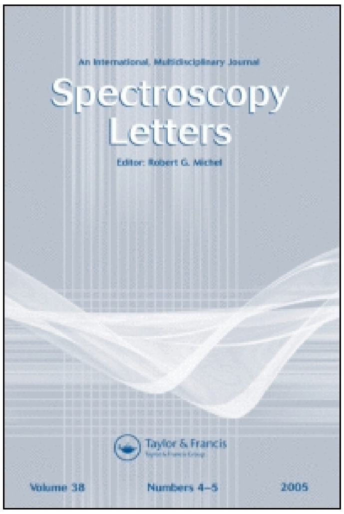

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ANTIRESONANCE OF RAMAN SCATTERING IN CdS_xSe_{1-x} MIXED CRYSTALS

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Resonant enhancement of the Raman scattering cross section in II - IV semiconductors has recently received much attention both theoretically and experimentally. All existing theories anticipate a monotonic increase in the scattering intensities when the scattering radiation energy approaches the direct-energy gap. Contrary to them in an early Raman study of CdS [1] a cancellation of scattering efficiencies for the two TO modes prior to the onset of the resonance was pointed out. In a latter work on pure CdS Damen et al.[2] found even a more pronounced "antiresonance" behavior of the nonpolar E_2 phonon at 41 cm^{-1}. Thus, this striking feature seems to be rather common for the Raman active modes in CdS for which no electrooptic contribution to the scattering amplitude exists. The experimental data were qualitatively explained by assuming a destructive addition between nonresonant and the weaker resonant terms in Loudon's expression for the first-order Raman tensor [3]. Consequently the cancellation energy difference $/E_G - \hbar\omega_L/$ depends on the ratio of the resonant term to the nonresonant terms.

In all these investigations the resonant effect has been achieved by varying the excitation wavelength. Another alternative would be the variation of the band gap in solid solutions at a fixed laser energy. The absorption edge of mixed crystals could be changed in a relatively wide range. Of course, such systems are more complicated because scattering intensities are related to the concentration of the participants.

In this work we report a Raman intensity antiresonance for the nonpolar E_2 phonon at 41 cm^{-1} in CdS_xSe_{1-x} mixed crystals. CdS_xSe_{1-x} crystals are quite convenient for resonance studies with the 6328 Å (1,96 eV) He-Ne laser line because their band gap energy $E_G(x)$ at room temperature varies from 2,41 eV to 1,70 eV for pure CdS and CdSe, respectively [4].

The experimental arrangement was similar to that reported in our previous work [5]. The following crystal compositions were used: 100%, 93.5%, 82%, 76%, 74%, 70%, 62%, 57% and 53% of CdS. It is known that CdS_xSe_{1-x} crystals demonstrate a two-mode behavior, i.e. for any concentration two sets of modes exist: CdS- and CdSe-like modes. The mode scattering intensities are nearly proportional to the content of the relevant components. To obtain integrated scattering intensities of CdS-like modes, we had to divide the measured intensities by the S concentrations.

The mode at 41 cm^{-1} is the dominant Raman band away from the resonance. Fig. 1 shows the scattering intensity change

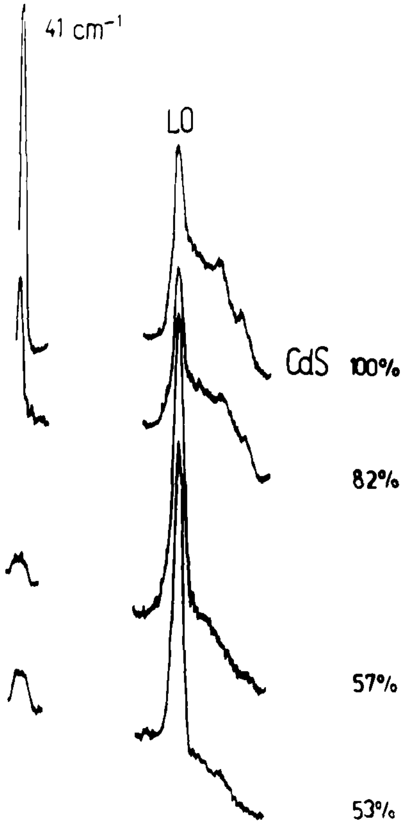

FIG. 1

Concentration Dependence of the 41 cm^{-1} and LO Raman Bands (CdS-like modes) of CdS_xSe_{1-x} Crystals Obtained with 6328 Å Excitation in $z(x^z_y)x$ Scattering Geometry

when the CdS mole percentage is decreased. The CdS-like LO mode (304 cm^{-1} in pure CdS) in the same compositions is shown for comparison. Its scattering as a function of gap-laser energy difference is shown in Fig. 2. Measurements of absolute intensities are difficult because samples with different compositions are not completely equivalent. Moreover absorption

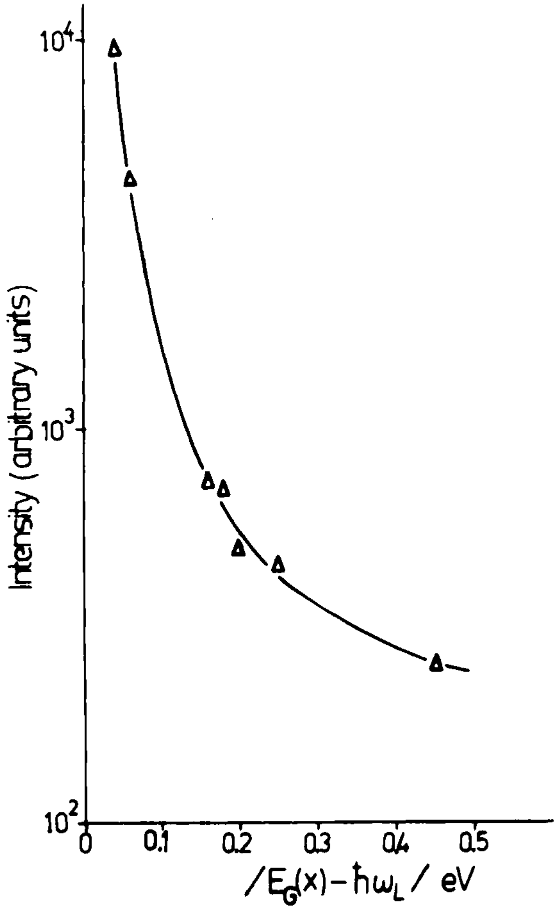

FIG. 2

Scattering Intensitiy of the CdS-like LO Mode as a Function of $/E_G(x) - \hbar\omega_L/$

corrections for samples with 57% and 53% CdS are necessary [6]. That is why, the accuracy of the scattering values plotted in Fig. 2 is not better than 40 per cent. It is seen that the LO scattering increases continuously as the resonance condition is approached.

Relative intensity measurements between two Raman frequencies in the same crystal remove all difficulties mentioned above. We have measured the integrated intensities of the mode at 41 cm^{-1} relative to those of the LO mode. The results are shown in Fig. 3. The experimental point at $/E_G(x) - \hbar\omega_L/=0.52$ eV was obtained by cooling the pure CdS

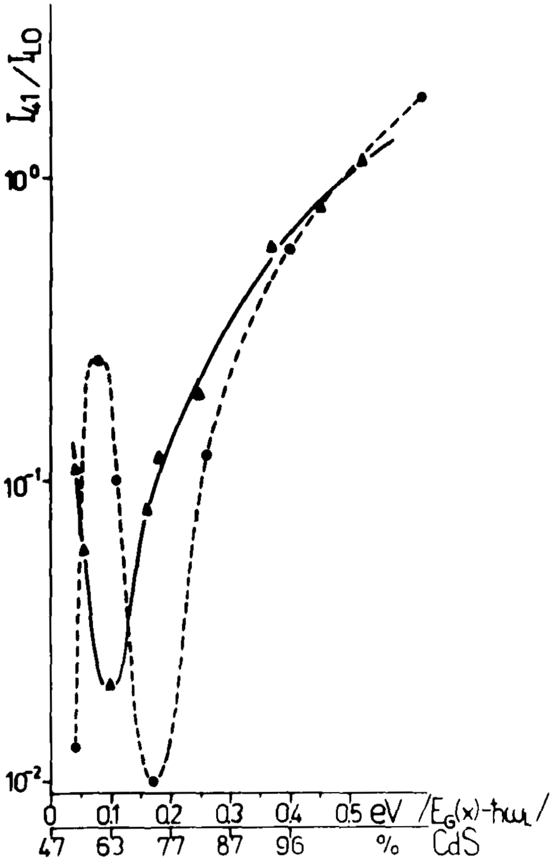

FIG. 3

Plots of I_{41}/I_{LO} versus $/E_G(x) - \hbar\omega_L/$. The solid curve presents our experimental results. The dotted curve is calculated from the experimental data of ref. [2].

crystal down to 200°K. At this temperature $E_G(CdS)=2.48$ eV [4]. Our data may be compared with the ratio of Raman cross sections I_{41}/I_{LO} calculated from reference [2] for the same modes in pure CdS. Cancellation of scattering amplitudes occurs in both cases but the cancellation energy in our case (0.07 eV) is somewhat lower than that in the case when the laser excitation was varied (0.20 eV).

A similar result was reported recently by Sussman et al. [7] studying the effect of alloying on resonance cancellation of the TO (A_1) mode in $CdS_{0.9}Se_{0.1}$ crystal. The antiresonance occured at an incident photon energy which is about 0.10 eV below the crystal band gap energy. The same phenomenon was observed in pure CdS at $/E_G - \hbar\omega_L/\approx 0.34$ eV [1]. Evidently the the existing resonance models considering the difference between crystal band gap and laser energy only are oversimplified. The importance of the antiresonance effects is clear from the fact that the CdS-like TO modes at 228 and 235 cm^{-1} (A_1 and E_1 symmetry) are not observable with 6328 Å excitation in all our samples whereas they are strong lines in the 5145 Å excited spectra. This can be explained only by such antiresonance intensity decrease. Calculations including data for mixed crystals could be useful in giving a correct mechanism responsible for the resonant Raman effect.

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