Two-photon Raman scattering at high levels of excitation of CdS crystals

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(Submitted 31 August 1979)
Pis'ma Zh. Eksp. Teor. Fiz. **30**, No. 9, 603–608 (5 November 1979)

Dependence of the position, shape and intensity of two-photon Raman scattering in CdS crystals on the sample temperature, photon energy and power density of exciting radiation was investigated. The experimental results are explained in terms of the intersection of high-density electron-hole pairs in the system.

PACS numbers: 78.30.Gt

In this work we investigated emission from pure CdS samples placed in a cryostat with temperature regulation from 4 to 80 K, which were excited by a tunable pulsed laser operating on a 7D4TMK solution in dioxane (halfwidth of the emission line 0.25 meV, power ~ 10 kW, pulsewidth 4 nsec). The crystal C axis was vertical in the crystal surface plane. The wave vectors $\mathbf{k}_{\text{inc}}$ of the laser radiation and $\mathbf{k}_R$ of observed emission were in the horizontal plane and made angles α and β with the perpendicular to the sample surface (Fig. 1). At a pumping power $P \gtrsim 10^4$ W cm^{-2} two new lines R_L and R_T are observed alongside known M- and P_M-bands (Fig. 2), which are polarized E⊥C and whose spectral maxima position $\hbar\omega(R_L)$ and $\hbar\omega(R_T)$ were independent of the pumping but were dependent on the angle of incidence α and observation β, sample temperature $T°$ and laser photon energy $\hbar\omega_{\text{inc}}$. Thus, at $T° \approx 5$ K, $\alpha = 42°$ and $\beta = 90°$ the maxima position of these lines (circles in Fig. 1) satisfy the following relationships (solid straight lines in Fig. 1):

$$\hbar\omega(R_L) = 2\hbar\omega_{inc} - E_L(T°, \mathbf{k}_L), \tag{1}$$

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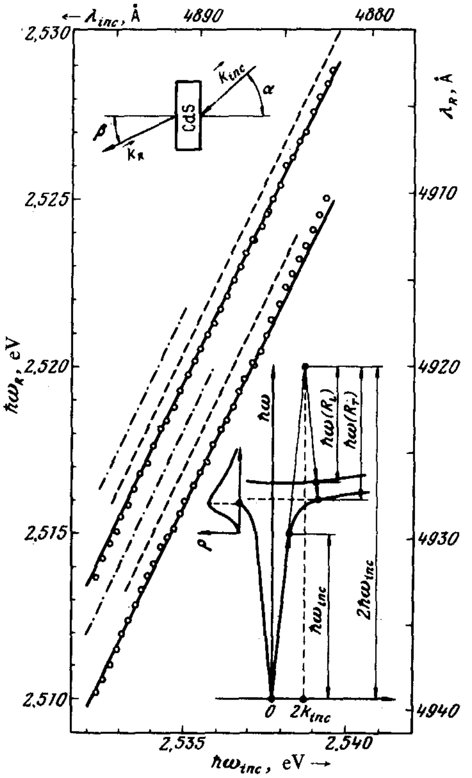

FIG. 1. Dependence of maxima position $\hbar\omega(R_L)$ and $\hbar\omega(R_T)$ of two-photon Raman scattering (RS) lines on photon energy $\hbar\omega_{inc}$ of pumping laser radiation at incidence $\alpha = 42°$ and observation $\beta = 90°$ angles and temperatures: ○—5, □—18, Δ—27 K. Right lower corner shows two-photon RS process scheme.

$$\hbar\omega(R_T) = 2\hbar\omega_{inc} - E_T(T°, \mathbf{k}_T), \qquad (2)$$

where $E_L(T°,\mathbf{k}_L) = 2.5549$ eV and $E_T(T°,\mathbf{k}_T) = 2.5516$ eV are the energies of a longitudinal exciton and transverse polariton of a lower branch at $T° = 5$ K and wave vectors $|\mathbf{k}_L|;|\mathbf{k}_T| \approx 0.4 \times 10^6$ cm^{-1}. As the temperature increased (at constant P and $\hbar\omega_{inc}$), the intensity of the R_L and R_T lines sharply dropped off to 0 at $T° \approx 25$ K; moreover, position of the line maxima shifted in the direction of large energies such that whereas equations (1) and (2) are satisfied as before, while now $E_L(T°,\mathbf{k}_L)$ and $E_T(T°,\mathbf{k}_T)$ are positions of the energy levels of a longitudinal exciton and transverse polariton at corresponding temperatures. Figure 1 shows approximations of the experimental functions (solid, dashed and dot-dash lines) $\hbar\omega_R = f(\hbar\omega_{inc})$ by Eqs. (1) and (2) for $T° = 5$, 18 and 27 K, respectively. At $|\beta - \alpha| < 90°$ the slope of $\hbar\omega_R = f(\hbar\omega_{inc})$ differs from 2 and depends heavily on $|\beta - \alpha|$.[1-3]

The aforementioned results are explained in terms of two-photon Raman scattering (RS), where two laser-emitted photons with energies $\hbar\omega_{inc}$ and wave vectors $\mathbf{k}_{inc}$ virtually excite a certain state of a high-density electron-hole (ED) pair system, after which they decay into a longitudinal exciton with energy $E_L(T°,\mathbf{k}_L)$ and $\mathbf{k}_L$ (or a transverse polariton of the lower branch with energy $E_T(T°,\mathbf{k}_T)$ and wave vector $\mathbf{k}_T$), and a photon with energy $\hbar\omega(R_L)$ and wave vector $\mathbf{k}(R_L)$ ($\hbar\omega(R_T)$ and $\mathbf{k}(R_T)$, respec-

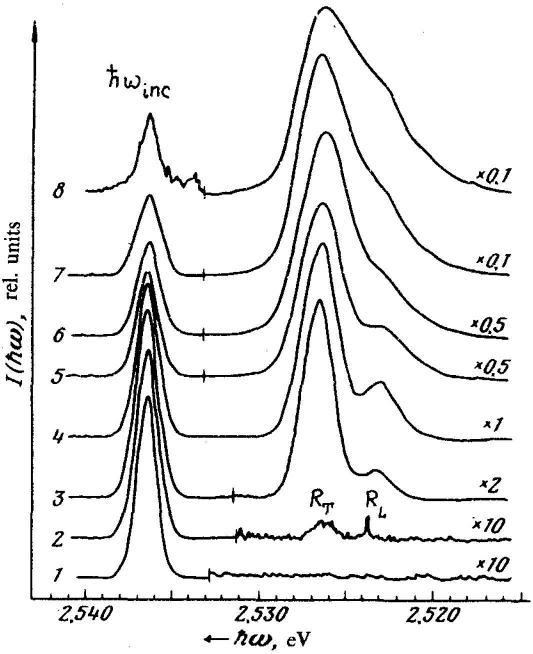

FIG. 2. Shape of two-photon RS lines at the temperature 5 K, $\alpha = 57°$, $\beta = 0°$, $\hbar\omega_{inc} = 2.5364$ eV, and different sample excitation levels. Curves 1–8 correspond to power densities of 1.2, 4.1, 5.3, 8.3, 11, 13 and 20 MW/cm^2, respectively.

tively, for the second RS line; the right corner in Fig. 1). From the laws of conservation of energy and momentum it follows

$$2\hbar\omega_{inc} = \hbar\omega(R_L) + E_L(T°, \mathbf{k}_L),$$

$$2\mathbf{k}_{inc} = \mathbf{k}(R_L) + \mathbf{k}_L.$$

The above relations also define the spectral positions (1) and (2), the angular and temperature dependence of the RS lines for different $\hbar\omega_{inc}$.

The halfwidth of the RS lines (at constant P and $\hbar\omega_{inc}$) depends weakly on the sample temperature and the angles α and β, increasing slightly with increasing $T°$ and decreasing $|\beta - \alpha|$. As the pumping increases the RS lines broaden significantly (Fig. 2), and at $P \sim 10^5$–10^7 W/cm^2 they take on the shape of a symmetrical bell and, subsequently, at $P > 10^7$ W/cm^2, an asymmetric shape with a mildly-sloping longwave tail, attaining a halfwidth of ~ 7 meV at $P \approx 2 \times 10^7$ W/cm^2.

At constant pumping and a 5 K temperature the intensity of the RS lines depends strongly on the laser radiation wavelength (λ_{inc}). At minimal excitation levels $P < 10^5$ W/cm^2 the intensity of RS line maxima increases in resonance when λ_{inc} and λ_{inc}^{res} draw toward a value of 4861.2 ± 0.3 Å.[4] As the pumping intensifies, the resonance position shifts in the direction of lower energies (Fig. 3) and the resonant curve $I_R(\lambda_{inc})$ broadens considerably; moreover, in the pumping range $P \sim 10^7$ W/cm^2 the position of the shortwave edge $\lambda_R^{\mu} \approx 4878 \pm 2$ Å of the curve $I_R(\lambda_R)$ remained virtually unchanged.

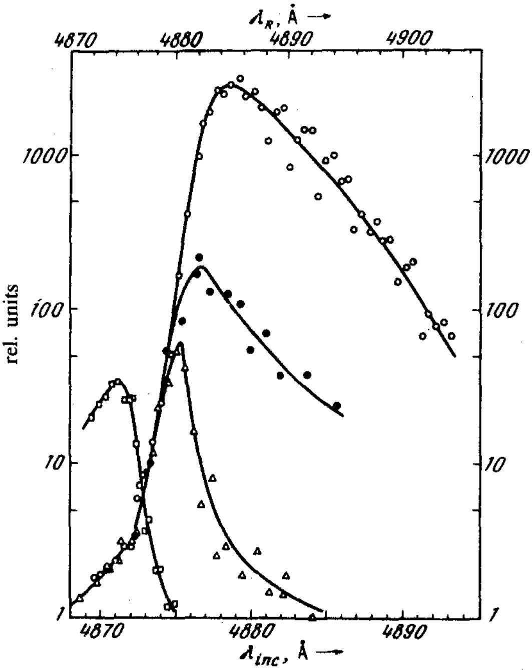

FIG. 3. Dependence of intensity I_R at line maxima of two-photon RS on the wavelength of exciting laser radiation $\lambda_{\rm inc}$ (lower scale) or on position of line maximum of two-photon RS λ_R (upper scale) at pump power densities in (MW/cm^2): □—1.5, Δ—8, ●—13; ○—22.

The results given are a function of interaction of high-density EH pairs in the system and may be explained as follows. Excitons and biexcitons at comparatively low excitation levels $\sim 10^4$ W/cm^2 constitute "good" quasi-particles with energies $E_L = 2.5549$ eV, $E_L - E_T \approx 2$ meV, $E_M = 5.1012$ eV.[3,4] The biexciton is the lower state that may be excited by two photons and, therefore, the intensity of two-photon RS resonantly increases when the laser photon energy converges on $\hbar\omega_{\rm inc}^{\rm res} = E_M/2$ ($\lambda_{\rm inc}^{\rm res} = 4861.2 \pm 0.3$ Å). It was shown [5] that with increasing excitation level, the absorption levels that correspond to the exciton bound states (and, consequently, also the biexciton) are broadened as a result of interaction, and convert at sufficiently high excitation levels into an asymmetric band with mildly-sloping shortwave and steeper longwave wings [the $\rho(\hbar\omega)$ curve in the right corner Fig. 1], which at 12–13 meV below the free exciton level changes into a broad asymmetrical band of "negative" absorption (i.e., amplification), with gain up to 10^3 cm^{-1}.[6] This kind of energy spectrum transformation also determines the behavior of the shape and intensity of RS lines with increasing excitation level. Actually, as a result of broadening of the exciton level (which serves as the final level in the act of two-photon RS) predominantly in the shortwave direction, the RS lines will broaden, in agreement with Eqs. (1) and (2), predominantly into the longwave direction (see, RS process scheme in the right corner of Fig. 1), as can be seen from Fig. 2.

The displacement of the resonance curve $I_R(\lambda_{\rm inc})$ is also associated not with a shift of the biexciton level with the pumping but with a change in the coefficients of absorption and amplification of the excited crystal regions through which the scattered radiation is propagating. As the pumping intensifies due to the broadening of the biexciton level, the minimum photon energy required for biexciton excitation is reduced; from the shortwave side of the resonance position $\lambda_{inc}^{res} = 4861.2$ Å due to the broadening of the exciton level, the absorption coefficient increases, and from the longwave side a region of "negative" absorption develops, which is associated with the formation of EH liquid.[6] As a result of these factors the resonant curve $I_R(\lambda_{inc})$ broadens with the pumping increase, is shifted in the direction of lower energies, and, finally, begins to a considerable degree to be determined by the spectral gain distribution[7] comprised in the EH liquid, since the scattered radiation in passing through the volume occupied by the EH liquid is attenuated in a region of those wavelengths where the absorption coefficient has increased, and amplified in a region $\lambda_R \gtrsim 4878$ Å that corresponds to large values of gain.

The authors thank L.V. Keldysh and E.I. Rashba for discussions of the work and the DFG for support of the work.

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