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Resonance Raman scattering in $CuGa_xAl_{1-x}S_2$ crystals

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Abstract
The change of polarization of the resonant Raman scattering lines is studied in $CuGa_xAl_{1-x}S_2$ crystals with x equal to 1, 0.95 and 0.9 assuming resonance conditions with 4880 and 4765 Å Ar laser lines. Linear and circular depolarization of the emission under the excitation by polarized light is observed. The depolarization of LO-phonon resonance Raman scattering (RRS) lines is found to occur practically in the same way for phonons with different symmetries. This observation is explained by the participation of polaritons in the processes of RRS as initial and final states.

Keywords: photoluminescence, chalcopyrites, exciton-mediated interactions, resonant Raman scattering, reflection spectra

1. Introduction

Resonance Raman scattering (RRS) is a versatile tool for studying the interaction of electrons and holes with elementary excitations in crystals and extracting information concerning the spin state of the excited electron–hole pairs. The spin orientation of carriers in semiconductors by means of optical pumping has been investigated experimentally and theoretically [1–3]. The excitation of excitons by polarized light in crystals leads to the change of radiation polarization [3–5]. The spin states of the excited carrier pairs remain always correlated due to their interaction in the exciton, while the energy dissipation by one or several longitudinal optical (LO) phonons occurs.

An excitonic luminescence band related to the recombination of excitons in the ground or excited states with momentum $\mathbf{k} = 0$ is observed in the region of ω_{exc} when the crystal is excited by light with a frequency ω_0 higher than the resonance exciton frequency ω_{exc}. At the same time, a series of narrow lines is observed at higher frequencies, shifted from the excitation line ω_0 by the frequency multiple to the frequency of the longitudinal optical phonon [4–6]. The emission line at $\omega_0 - 2\omega_{LO}$ was treated as resonance Raman scattering [7] or as the luminescence of hot excitons [8]. In the latter case, the exciton is formed as a result of indirect excitation with the participation of the LO phonon. The recombination takes place also with the emission of an LO phonon. As a result, the emission line, shifted by the frequency $(n + 1)\omega_{LO}$ from the excitation line, corresponds to n intermediate exciton states. It was shown [9] that the resonance scattering is equivalent to the hot luminescence if only the ground exciton state is considered as an intermediate state in the resonant scattering. In this case, the contour of the emission line is monotonic in the short-wavelength region, while it is sharp in the long-wavelength region [5, 6]. The more detailed analysis shows that the emission for the $\omega_0 - 2\omega_{LO}$ line has an additional component, which is comparable to the main one. This component cannot be explained by a simple kinetic treatment in the framework of hot luminescence theory [9].

In contrast to the II–VI compounds, the chalcopyrite crystals are characterized by the presence of several polar modes. The infrared (IR) and Raman active vibration modes have been determined in most of the chalcopyrite crystals [10–12].

Noncubic, anisotropic materials, in particular the chalcopyrite crystals with tetragonal lattice symmetry, have been scarcely investigated up to very recently. In the past few years, the resonant Raman scattering in $CuGaS_2$ has been intensively examined [13–15]. The polarization measurements in various resonant Raman scattering geometries under excitation by different wavelengths have been performed [13–19]. An argon laser (457.9, 465.6, 472.7, 476.5, 488.0, 496.5, 501.7 and 514.5 nm), a DCM-dye laser (650–720 nm) and a Ti:sapphire laser (720–840 nm) were used to achieve the

