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SOLID STATE SPECTROSCOPY

Spatial Dispersion in Polariton Spectra of CuGaS₂ Crystals

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Abstract—The unmodulated and wavelength-modulated reflectivity spectra of CuGaS₂ crystals for the polarization $\mathbf{E} \parallel c$, $\mathbf{k} \perp c$ at 77 and 8 K have been studied. The states n = 1, 2, and 3 of A excitons and n = 1 and 2 of B and C excitons are established. The luminescence spectra from the surface at $\mathbf{k} \parallel c$ and $\mathbf{k} \perp c$ are obtained. The fine structure of the reflectivity spectra of excitons are analyzed with due regard for the normal and oblique incidence of light onto the crystal surface. The main parameters of the A, B, and C excitonic series are determined such as the energies of the longitudinal and transverse excitons Γ_4 ($\mathbf{E} \parallel c$) for states n = 1 and 2, the longitudinal and the transverse mass of excitons in CuGaS₂, and the effective masses of electrons (m_{c1}^*) and holes $(m_{v1}^*, m_{v2}^*, m_{v3}^*)$. It is shown that the mass m_{v1}^* in the upper valence band at $\mathbf{k} \parallel c$ equals $(0.7-0.8)m_0$ and at $\mathbf{k} \perp c$, $1.87m_0$. © 2002 MAIK "Nauka/Interperiodica".

INTRODUCTION

Among the experimental and theoretical studies of the optical spectra of polaritons performed in the last decades, there are a number of experimental works on reflectivity spectra of polaritons under the conditions of oblique incidence [1–9]. The analysis of the reflectivity spectra light of polaritons depending on the angle of light incidence onto the differently oriented samples (geometry), wave vectors, and the polarizations of the incident and the reflected beam provides important information on the parameters of the exciton polaritons, such as the oscillation strength, the energy of the longitudinal and transverse excitons, the effective mass of excitons and its dependence on the orientation of the wave vector with respect to the axis of optical activity of the crystal, etc. [7, 8].

At present, the polariton spectra, the problems of spatial dispersion, and many other important characteristics of the behavior of exciton polaritons are mostly studied mainly for $A^{II}B^{VI}$ crystals (see, e.g., [1–9] and references there]. To confirm the universal nature of the "hot" polariton luminescence, elastic scattering of polaritons, and the optical orientation of the exciton spins, it is important to study these effects on the crystals of multicomponent materials, and, in particular, CuGaS₂ crystals, which show polariton luminescence [10] and resonance Raman scattering of exciton polaritons [11–14].

In this study, we obtained new data on the parameters of the long-wavelength exciton polaritons of the A series and studied the wavelength-modulated reflectivity spectra of the A, B, and C excitons, the luminescence spectra, and the reflectivity and the luminescence spectra as functions of the angle of incidence of the exciting beam of CuGaS₂ crystals in the range of the A exciton absorption at 77 and 8 K. We also obtained information on the parameters of excitons, their band structure, and anisotropy of the exciton mass.

EXPERIMENTAL

The experiments were performed on CuGaS₂ crystals grown by gas-transport reactions. The mirror-like unpolished 1×1 cm-large crystals were parallel to the *c*-axis [the (010) surface]. The $8 \times 8 \times 4$ mm-large crystals were prisms with perpendicular faces. Some of the crystals had two or three mirror-like faces. The dimensions and the quality of the natural faces of the crystals provided a reliable record of both reflectivity and luminescence spectra at the same spot of the crystal surface at different orientations of the polarization vector of the light and the wave vector k. Some crystals had mirrorlike surfaces tilted at angles $\approx 40^{\circ}-45^{\circ}$ with respect to the c axis. The reflectivity spectrum from such a surface in the exciton range was independent of the orientation of the electric vector **E** of the incident light wave, i.e., the surface behaved as an isotropic crystal. The optical reflectivity and luminescence spectra were recorded on a setup based on a double Raman DFS-32 spectrometer with the 1200 grooves/mm-gratings. The reflectivity spectra were measured with the use of the slit 0.015 Åspectral width. The samples were mounted on a cold finger of the cryostat LTS-22 C 330 of the Workhorse type and were kept at a temperature of 9 ± 0.5 K. The luminescence spectra were excited by the emission lines $\lambda = 4880$ and 4765 Å of an Ar⁺-laser. The exciting light power in the spot 1-3 mm in diameter did not exceed 100 mW. The reflection and the absorption (transmission) spectra were studied using the continuous spectrum of an incandescent lamp (≈ 100 W).