Free and bound excitons in $TlInS_2$ crystal

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It is established for the first time that the photoluminescence of a $TlInS_2$ crystal with monoclinic modification near the fundamental absorption edge at 1.8 K is due to radiative recombination of free and bound excitons.

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Layered single crystals of $TlInS_2$ crystallize in six polytypical structures:two monoclinic, one triclinic, and three hexagonal modifications.[1] Investigations of the absorption of $TlInS_2$ at low temperatures show that the fundamental absorption edge of the crystal has a complicated structure due to the formation of free excitons.[2,3] There are no data on absorption below 5 K and on emission.

In this paper we present the first results of investigations of the photoluminescence and absorption near the fundamental absorption edge of the crystal $TlInS_2$ at 1.8 K. The photoluminescence, excited by LG-69 argon laser radiation ($\lambda_{exc} = 4765$ Å), was investigated from the side of the laser-illuminated surface of the specimen in a direction close to the normal to the layer which contains the second-order "**b**" axis. An incandescent lamp was used as a light source in studying the absorption. The investigation light signal was polarized and, after the transmission through the DFS-12 spectrometer, was recorded by a synchronous detection system. We prepared the specimens directly prior to performing the measurements by the method cleaving along the cleavage surface from single crystals grown by Bridgman's method. An x-ray

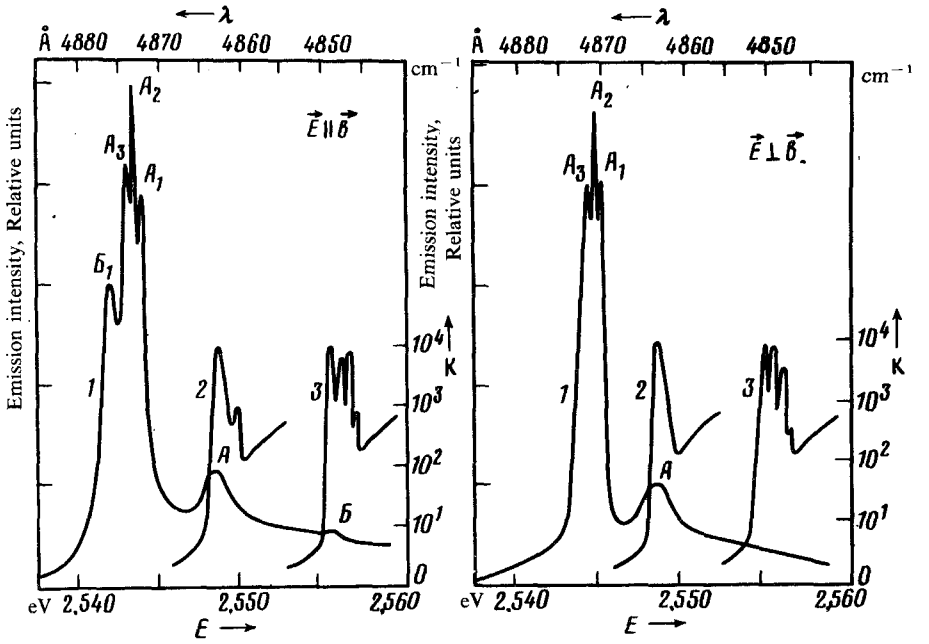

FIG. 1. Photoluminescence (1) and absorption (2,3) spectra of a $TlInS_2$ single crystal at 1.8 K for polarizations E||b and E⊥b.

structural analysis of the specimens revealed the presence in them of two polytypes of the monoclinic modification of $TlInS_2$ I and $TlInS_2$ II (space symmetry group C_s^4), according to well-known data in Ref. 3.

Figure 1 shows the photoluminescence[1] and absorption[2,3] spectra of the $TlInS_2$ crystal at 1.8 K for polarization E||b and E⊥b. The differences in the absorption spectra observed from the energy position of the fundamental absorption edge with the fine structure resulting from the formation of free excitons are explained by the presence of two polytypes in the single crystals investigated :$TlInS_2$ I in Fig. 1 (Ref. 2) and $TlInS_2$ II in Fig. 1 (Ref. 3).

As was clarified by numerous absorption experiments, the section of the crystal containing only one polytype of the monoclinic modification has dimensions not exceeding $\sim 0.3\times 0.3\times 0.003$ mm. It was impossible to make a reliable recording of the photoluminescence of a section of the specimen of this size, which was located within a helium cryostat with several light-scattering glass barriers, so that we recorded the luminescence from the larger area of the same specimen encompassing both polytypes.

The energy positions of the observed photoluminescence lines are as follows: 2.5490 eV (A), 2.5569 eV (B), 2.5443 eV (A_1), 2.5438 eV (A_2), 2.5434 eV (A_3), and 2.5422 eV (B_1). The coincidence of the energy positions of the luminescence lines A and B and the peaks of the exciton absorption at 1.8 K indicates that these emission lines are associated with resonance annihilation of free excitons in the two polytypes of the monoclinic modification of a $TlInS_2$ single crystal.

The photoluminescence lines can be separated into two groups on the basis of their polarization dependence: A, A_1, A_2, A_3 and B_1, B. We note that at 4.2 K, the lines A_1, A_2, A_3, and B_1 merge, and disappear at 10 K; in this case, the intensity of lines A and B increases insignificantly. As the experiment showed, the intensities of the lines A_1, A_2, A_3, and B_1 vary linearly as a function of the excitation level. The lines A_1, A_2, A_3, and B_1 are more intense than the lines A and B, have half-widths 0.5, 0.4, 0.4, and 1.5 meV, and are separated from A and B by 0.0047, 0.0052, 0.0056, and 0.0147 eV, respectively. Comparison of these values with the IR absorption data and data from Raman scattering of the single-crystal $TlInS_2$[4] excludes the possibility of explaining the luminescence lines by the exciton-phonon-inter-action mechanism. Therefore, the dependence of the intensities of the lines A_1, A_2, A_3, and B_1 on the level of excitation, their energy positions and half-widths, as well as the temperature and polarization dependences indicate that the most likely mechanism for the appearance of these lines is the radiative recombination of bound excitons in two polytypes of the monoclinic modification of the $TlInS_2$ single crystal.

The results of investigations of absorption of a $TlInS_2$ single crystal will be reported separately.

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