Observation of resonance radiation in the region of the continuous spectrum of semiconductors

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Observation is reported, with indium and gallium monoselenides (InSe and GaSe) as examples, of resonance radiation due to transitions from states located in the interior of the conduction band and the valence bands and situated at distances commensurate with the width of the semiconductor forbidden band. The concept of a hyperbolic exciton is suggested as a possible explanation of the nature of the observed radiation.

Luminescence is usually observed in semiconductors from states near the principal absorption edge. Shorter-wavelength recombination radiation was also observed beyond the principal absorption edge and in its immediate vicinity. This includes the observed radiation near a direct transition in Ge[1] as well as luminescence in GaAs, which is attributed to the emission of the carriers heated by the light.[2] We report here observation, in semiconductors, of luminescence due to transitions from states located deep in the conduction or valence band and situated at distances commensurate with the width E_g of the semiconductor forbidden band. The experiments were performed on single crystals of indium monoselenide (InSe) and gallium monoselenide (GaSe) with E_g values ~1.3 and ~2.13 eV, respectively. The excitation was with a ruby laser ($\hbar\omega = 1.79$ eV) and the second harmonic of a neodymium laser; both lasers were Q-switched. The luminescence was registered with an MDR-2 monochromator, followed by synchronous detection and automatic recording of the spectra. The measurements were made in the temperature interval from 2.5 to 400 °K. When the InSe samples were excited by light directed along the C axis, we observed distinct emission lines in the 2.5—2.6 eV region, and in the case of GaSe at $T = 4.2$ °K we observed luminescence with photon energy 3.361 eV. The corresponding data on the spectral distribution of the observed emission in InSe at different temperatures are shown in Fig. 1. In this case, at low temperatures (in the interval from 2.5 to 77 °K) we observed two emission lines with energies $E_1 = 2.547$ and $E_2 \sim 2.613$ eV at $T = 4.2$ °K and $E_1 = 2.531$ and $E_2 \sim 2.588$ eV at $T = 77$ °K. With increasing temperature, the E_2 line decreased noticeably in intensity and vanished completely at temperatures above 100 °K. It should be noted that the energy E_2 differs somewhat from sample to sample, and the intensity of this line changes appreciably from crystal to crystal. As to the E_1 line, its energy position and the intensity stay the same in different samples within the limits of experimental error. At temperatures above 77 °K, some broadening of the E_1 line is observed, without a significant change in intensity, up to $T = 380$ °K. Plots of the position, the intensity I_1 at the maximum, the half-width ΔE_1, and the integrated intensity Q_1 of line E_1 against the temperature are shown in Fig. 2. For comparison, the figure shows the temperature dependence of the intensity I_2 and the position of the line E_2.

Attention is called to the relatively large half-width of the line (~40 meV) at low temperatures (the GaSe emission-line half-width is approximately the same), to the

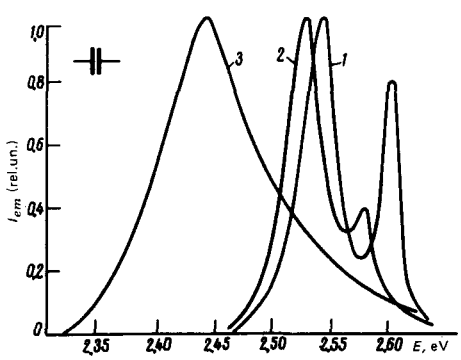

FIG. 1. Emission spectra of InSe at varous temperatures. $I_0 \sim (2-3) \times 10^{24}$ quanta/cm^2 sec: 1) $T = 4.2$ °K, 2) $T = 77$ °K, 3) $T = 293$ °K.

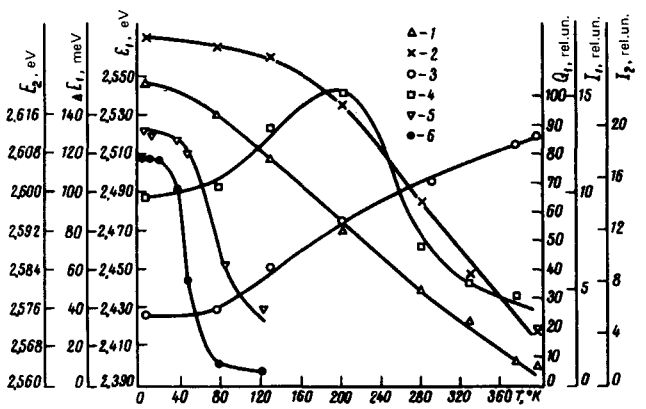

FIG. 2. Temperature dependences of the parameters of the observed emission. $I_0 \sim (5-6) \times 10^{24}$ quanta/cm^2 sec: 1) E_1, 2) I_1, 3) ΔE_1, 4) Q_1, 5) E_2, 6) I_2.

Emission (our data)			Absorption	
T, °K	E_1, eV	E_1, meV	E_1, eV [4]	E_1, eV [3]
4.2	2.547	37	–	–
15	2.546	37	2.546	–
77	2.531	39	–	2.534
90	2.526	42	2.527	–
200	2.470	83	2.470	–
293	2.440	104	2.420	2.440
376	2.402	125	–	–
	$\delta E_1/\delta T \cdot 10^4$ eV/deg		$\delta E_1/\delta T \cdot 10^4$ eV/deg	
2.3(4.2 – 77)°K			2.5 ±0.1(15 – 100)°K	4.2(77 – 300)°K
5.03(100 – 400)°K			5.2 ±0.2(100 – 200)°K	
			5.5 ±0.5(200 – 300)°K	

fact that it stays constant in the interval from 2.5 to 77°K, and to the relatively weak broadening with further rise in temperature. It is important to emphasize here that E_1 persists to very high temperatures (up to 400°K), and the integrated intensity at 400°K is lower by only a factor 2.5 than at helium temperature. As to the dependence of the intensity I_{obs} of the observed emission on the excitation intensity I_0, the lines E_1 and E_2 agree well with the relation $I_{obs} \sim I_0^m$, where $m = 1.5$ for E_1 and $m = 2$ for E_2. The line E_1 observed in emission correlates fully with the corresponding peak inside the intrinsic absorption region, which was observed in[3,4]. The parameters of this line, obtained by investigating the emission and absorption at different temperatures, are compared in the table.

We proceed to a discussion of the results. The fact that the position and intensity of the line E_2 differ in different samples and that there is no corresponding analog in the case of absorption[1)] seems to indicate that this line is connected with defects or impurities, which have, however, the distinguishing feature that their levels are located in a continuous-spectrum region that is higher than the edge of the intrinsic absorption edge by an amount on the order of E_g.

The resonant character of the line E_1 indicates without a doubt that the observed emission is brought about by excitons. The question of the critical point with which this exciton is connected cannot be answered unambiguously at present on the basis of our data and the data in the literature. It seems to us, however, that the results obtained in this paper, and particularly the large half-width of the line, its weak temperature dependence, the stability of the line up to 400°K, the weak dependence of the integrated intensity on the temperature, the asymmetry of the emission line with its steeper short-wave edge, which becomes much more pronounced at lower excitation intensities I_0, and also the results of an investigation of absorption in[3-5], can all be more consistently interpreted from the point of view of the concept of a hyperbolic exciton.[6-8]

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1)To be sure the absorption measurements were not made on the same samples in which the emission was observed.

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