SPECTRA OF PHOTOEXCITATION OF FREE EXCITONS BY SUBMILLIMETER RADIATION IN "ULTRAPURE" GERMANIUM

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A series of absorption lines corresponding to transitions from the ground state to excited states of a free exciton in germanium was registered for the first time in a wide submillimeter band (1.7 - 3.7 meV). The measurements were made at 1.5 - 4.2°K on a sample with a residual-impurity concentration 5×10^{10} cm^{-3}. Temperature measurements yielded for the splitting of the ground state of the indirect exciton a value $\Delta \approx 0.7$ meV.

Until recently, experimental investigations of excitons in semiconductors were performed exclusively in the region of interband transitions, i.e., in the optical region of the spectrum. Yet it is obvious that, just as in the case of shallow impurities, more complete information on the structure of the excitons and on their energy spectrum can be obtained from measurements in a region corresponding to the exciton binding energy, i.e., in the long-wave IR band in the case of most semiconductors. The first such experiments were performed on germanium [1], where absorption due to photoexcitation and photoionization of excitons was registered in the 2 - 5 meV region. Subsequently, such investigations were performed by a more sensitive high-resolution procedure using a backward-wave tube (BWT) as the monochromatic source of submillimeter radiation. This has made it possible to register a triplet with a maximum at 3.42 meV in the absorption spectrum of free excitons in germanium [2, 3]. Photoconductivity connected with the photothermoionization of free excitons in germanium was observed in the same region of the spectrum [3, 4]. The germanium samples used in all these experiments had a residual-impurity concentration 10^{12} - 10^{13} cm^{-3}, which is commensurate with the exciton concentrations typical of such experiments, 10^{12} - 10^{13} cm^{-3}, so that possible impurity effects were not completely excluded.

We present here the results of an investigation of spectra of exciton photoexcitation in a sufficiently wide submillimeter band in crystals of "ultrapure" germanium, in which the residual impurity content did not exceed 5×10^{10} cm^{-3} [5]. The measurements were performed with a BWT [6] in the wavelength intervals 340 - 455 and 510 - 730 μ, and also with the aid of a diffraction spectrometer for the far IR range 60 - 700 μ. The long-wave IR radiation was modulated in the measurements with the diffraction spectrometer [1]. In the experiments with the BWT we used a differential procedure with modulation of the radiation from an incandescent lamp that produced non-equilibrium carriers in the sample. The level of optical generation of electron-hole pairs was 10^{17} - 10^{18} cm^{-2}sec^{-1}. The experiments were performed in the temperature interval 1.5 - 4.2°K.

In the measurements at 1.5°K, which were performed with the aid of a diffraction spectrometer, we observed the spectrum of resonant absorption by electron-hole drops in "ultrapure" germanium; this spectrum differed little from that observed earlier in germanium with an impurity content 10^{12} - 10^{14} cm^{-3} [2]. On the edge of the spectrum, at a photon energy 2 - 4 meV, certain singularities were noted and became more pronounced when the temperature was raised to 2°K; they were shown to correspond to photoexcitation of free excitons [1, 2]. The results of a detailed investigation of the long-wave section of the spectrum, carried out with higher resolution and sensitivity by the BWT method at 4.2 and 1.6°K, are shown in Fig. 1. We see that the absorption spectrum of free excitons in germanium lies in the region 2.3 - 3.6 meV and consists of a series

of absorption lines with widths on the order of 0.05 meV (see, e.g., the line at 2.875 meV), which overlap in certain spectral regions. The positions of the individual lines and absorption maxima in the spectrum, measured at 4.2°K with accuracy ±0.005 meV, correspond to the energies 2.355, 2.410, 2.875, 2.965, 3.040, 3.135, 3.295, 3.350, 3.420, 3.495, and 3.540 meV. Similar spectra were observed with other samples of relatively pure n- and p-type germanium with residual-impurity concentrations 10^{12} - 10^{13} cm^{-1}.

Since the measured spectrum consists of narrow lines, it can be concluded that it is due principally to transitions from the ground state to excited states of the exciton. It follows obviously from Fig. 1 that the exciton binding energy in the ground state exceeds 3.7 meV in the case of germanium.

When the temperature is decreased to 1.6°K, as can be seen from Fig. 1, the intensity of the absorption bands in the region 3.135 - 3.540 meV decreases somewhat, whereas at longer wavelengths the absorption drops almost to zero. Such a change in the spectrum can be explained by assuming that these two groups of lines are due respectively to transitions from the lower and upper levels of the ground state of the exciton in germanium. Figure 2 shows the temperature dependences of the absorption intensities $\alpha_1 d$ and $\alpha_2 d$ (α_1 is the absorption coefficient, and $d \sim 1$ mm is the sample thickness) measured at two characteristic points of the spectrum, and also the temperature dependence of the ratio of these intensities. Since the latter is determined by the populations N_1 and N_2 of the two levels, $\alpha_1 d/\alpha_2 d = N_1/N_2 \sim \exp(-\Delta/kT)$, the energy difference Δ between the two levels of the ground state of germanium can be determined from the slope of the straight line in Fig. 2, viz., $\Delta = 0.7$ meV. The obtained value of Δ agrees satisfactorily with the theoretical estimates and with the measurements made in the interband-transition region [7, 8]. The decrease in the absorption intensity $\alpha_1 d$ when the temperature is lowered to 1.6°K is apparently connected with a transition of some of the carriers into the condensed phase, as is evidenced by the increase of the resonant absorption $\alpha_k d$, measured at $h\nu = 9$ meV.

Since the measured exciton-spectrum lines were quite narrow (~0.05 meV), it must be concluded that the fact that the excitons have a kinetic energy

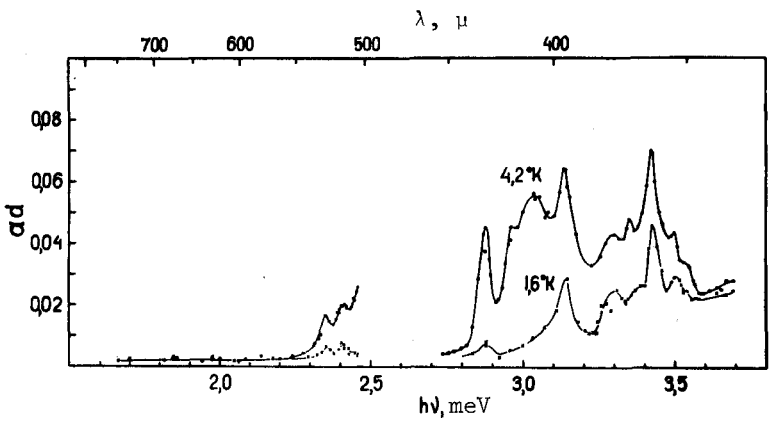

Fig. 1

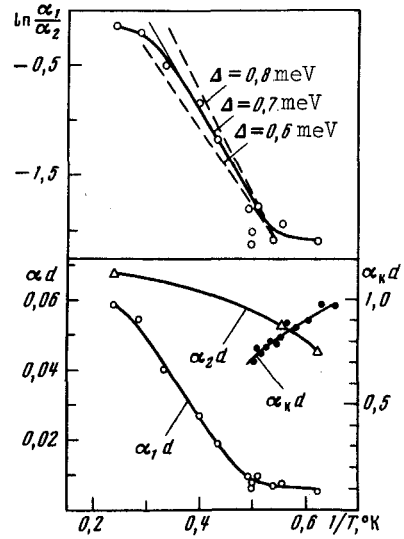

Fig. 2

Fig. 1. Spectrum of exciton absorption in "ultrapure" germanium.

Fig. 2. Induced absorption αd in different parts of the spectrum ($\alpha_1 d$ - 3.00 meV, $\alpha_2 d$ - 3.42 meV, and $\alpha_k d$ - 9 meV) and logarithm of the ratio α_1/α_2 vs. the reciprocal temperature.

($kT \sim 0.4$ meV at 4.2°K) makes no significant contribution to the broadening of the exciton-absorption lines, as proposed earlier [1]. Apparently, just as in the case of shallow impurities, the half-width of an exciton-absorption line is determined at low temperatures by the interaction of the excitons with the zero-point vibrations of the crystal lattice. Estimates based on the theory of [9] yield $\Delta h\nu \sim 0.01 \pm 0.02$ meV. At the same time the concentration averaged over the crystal volume reached $\sim 10^{13}$ cm^{-3} in our experiments[1]. A noticeable contribution to the line broadening can be made by interactions of the excitons with one another [2].

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ANOMALOUS BEHAVIOR OF THE MAGNETIC SUSCEPTIBILITY NEAR THE MAGNETIC-COMPENSATION POINT IN COMPOUNDS OF RARE-EARTH METALS WITH IRON

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In ferrimagnetic compounds of rare-earth metals with iron ($HoFe_3$, $ErFe_3$, and $ErFe_2$) we observed, near the compensation point T_c, maxima on the temperature dependence of the differential susceptibility χ, one at $T < T_c$ and another at $T > T_c$. Calculations of χ performed on the basis of the molecular-field theory have shown that such maxima on the $\chi(T)$ curves should appear in the noncollinear phase in fields weaker than the effective exchange field, if the magnetic field is directed along the easy-magnetization axis. It is shown that the theoretical value of the susceptibility in the noncollinear phase agrees with the experimental data.

In ferrimagnetic compounds of rare earth metals with iron, $ErFe_2$, $ErFe_3$, and $HoFe_3$, near the magnetic compensation temperature (490, 250, and 393°K,

[1] The volume lifetime of the excitons in the investigated "ultrapure" germanium samples, at the same exciton concentration and at 4.2°K, amounts to 5 - 10 μsec according to the data of B.V. Zubov and V.P. Kalinushkin (private communiation).