Plasma model of an exciton gas in a semiconductor

Z. A. Insepov, G. É. Norman, and L. Yu. Shurova

Moscow Power Institute
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A model of an exciton gas is developed in which termal dissociation of excitons into electrons and holes, formation of biexcitons, as well as adhesion of electrons and holes to excitons are taken into account. Allowance is made for the nonideality of such a plasma-like system. The composition of the exciton gas in germanium is calculated for temperatures 1-10°K and for excitation levels such that no electron-hole drops are formed. The conduction mechanisms in such systems are considered and agreement is obtained with available experimental data on the electric conductivity.

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At low temperatures, ~10 °K, the non-equilibrium electrons and holes produced in semiconductors by external excitation are pairwise bound to each other to form excitons.[1–7] The increased interest in these phenomena is due to the fact, predicted by Keldysh,[1] that the excitons condense at large concentrations and two phases appear, vapor and liquid.

It should be stated that the exciton gas consists not only of excitons (eh or ex). The excitons can be bound to one another to form biexcitons (e_2h_2 or b).[7] Thermal dissociation leads to the appearance of free electrons (e) and holes (h).[8,9] Free charges adhere to the excitons and form charged three-particle complexes (e_2h and eh_2 or ex^- and ex^+).[10] The following reactions take place in the system:

$$ex \rightleftarrows e+h, \qquad b \rightleftarrows 2ex,$$
$$ex^- \rightleftarrows ex+e, \qquad ex^+ \rightleftarrows ex+h.$$

Thus, the exciton gas is in fact a multicomponent system. The analogy with hydrogen is obvious: h—proton H^+, ex—hydrogen atom H, b—H_2 molecule, ex^- and ex^+—H^- and H_2^+ ions.

Calculation of the composition of the exciton gas is made difficult by the fact that the ideal-gas approximation turns out to be incorrect in a sufficiently large region below the saturation line. It is necessary to take into account the interactions of the charged particles with one another and with the neutral particles.

In this paper we construct a multicomponent model of an exciton gas. We use methods borrowed from the theories of real gases and of non-ideal plasma, and account is taken of the features peculiar to semiconductors. The composition is calculated for germanium. We discuss the conduction mechanisms and compare the results with the experimental data.

Multicomponent model of exciton gas. The chemical-equilibrium conditions

$$\begin{aligned} &\mu(ex)=\mu(e)+\mu(h), \qquad \mu(b)=2\mu(ex), \\ &\mu(ex^-)=\mu(ex)+\mu(e), \quad \mu(ex^+)=\mu(ex)+\mu(h), \end{aligned} \tag{1}$$

where μ are the chemical potentials, make it possible to connect the concentrations of the different components. Before writing down the corresponding equations for the dissociative (ionization) equilibrium, we note a number of features due to the fact that an exciton plasma exists in the solid.

1. The masses of the positive and negative particles are comparable.

2. The degeneracy of the extremum of the band at the point $k=0$ leads to the appearance of two types of holes, light and heavy (h' and h), and corresponding different types of excitons, three-particle complexes, and biexcitons should exist. The connection between the concentrations of the light and heavy particles can be obtained by noting that they are transformed into one another via emission or absorption of phonons (f): $h \rightleftarrows h' \pm f$, $ex \rightleftarrows ex' \pm f$ etc. Since the chemical potential of the phonons is equal to zero, we have $\mu(h)=\mu(h')$, $\mu(ex)=\mu(ex')$. Thus, the concentrations (without allowance for the interaction between the particles) are connected by the relations

$$n(h)/n(h')=(m_h/m_{h'})^{3/2},$$
$$n(ex)/n(ex')=(m_{ex}/m_{ex'})^{3/2}\exp(\beta I_{ex}-\beta I_{ex'})$$

etc. Here m and m' are the masses, I and I' are the binding energies, and $\beta=1/kT$.

3. The extremum of the condition band at the point $k\neq 0$ is degenerate in the number of valleys, i.e., the multiplicity of the degeneracy of the particles of the corresponding band increases by a factor N (N is the number of valleys). For example, the statistical weight of the electron in germanium is $g_e=g_e^*N$, where $g_e^*=2$ is the statistical weight of the free electron in vacuum, and $N=4$.

4. The masses of the particles in a solid can be anisotropic, $m_\parallel \neq m_\perp$. We can set approximately the effective mass equal to $m=(m_\parallel m_\perp^2)^{1/3}$, and for composite particles $m_\parallel$ ($m_\perp$) is taken to mean the sum of the corresponding number of $m_\parallel(m_\perp)$ of the electrons and holes. The values of the binding energies contain a different mass: $m_0=(m_\parallel^{-1}+2m_\perp^{-1})^{-1}$.

5. The composition of the gas exciton phase can be greatly influenced by impurity atoms. The impurity atoms existing in the ground state at $T\sim 10$ °K are not ionized (their binding energy is $\gtrsim 100$ °K) and influence the composition only via interaction with particles of the exciton phase.

Allowance for non-ideality. The free energy can be approximately written in the form

$$F=\sum_i F^0(i)+\sum_i N(i)I_i-\Phi-\sum_{ij}c(i,j)N(i)N(j)V^{-1}-\sum_{ij}a(i,j)N(i)N(j)V^{-1}. \quad (2)$$

Here $F^0(i)$ is the free energy of the i-th component of the ideal system, $N(i)$ is the number of particles of the i-th component, $c(i,j)$ and $a(i,j)$ are the virial coefficients that take into account the charge–neutral and neutral–neutral interaction (including the impurity atoms), Φ is the contribution of the Coulomb interaction, and nondegenerate systems are considered.

Recognizing that $\mu(i)=(\partial F/\partial N(i))_V$, we obtain from (1) and (2)

$$n(ex)=n(e)n(h)\left(\frac{2\pi\hbar^2\beta m_{ex}}{m_e m_h}\right)^{3/2}\frac{\Sigma_{ex}}{g_e g_h}\exp(\beta I_{ex}-\beta\Delta I_{ex}), \quad (3)$$

$$n(b)=n^2(ex)\left(\frac{2\pi\hbar^2\beta m_b}{m_{ex}^2}\right)^{3/2}\frac{\Sigma_b}{\Sigma_{ex}^2}\exp(\beta I_b-\beta\Delta I_b), \quad (4)$$

$$n(ex^-)=n(ex)n(e)\left(\frac{2\pi\hbar^2\beta m_{ex^-}}{m_{ex}m_e}\right)^{3/2}\frac{\Sigma_{ex^-}}{\Sigma_{ex}g_e}\exp(\beta I_{ex^-}-\beta\Delta I_{ex^-}), \quad (5)$$

$$n(ex^+)=n(ex)n(h)\left(\frac{2\pi\hbar^2\beta m_{ex^+}}{m_{ex}m_h}\right)^{3/2}\frac{\Sigma_{ex^+}}{\Sigma_{ex}g_h}\exp(\beta I_{ex^+}-\beta\Delta I_{ex^+}), \quad (6)$$

where Σ are the partition functions and g are the statistical weights. ΔI include the changes of the binding energy due to the interaction, for example,

$$\Delta I_{ex}=2\varphi+[c(e,ex)+c(h,ex)-2a(ex,ex)]n(ex)+[c(e,b)+c(h,b)]n(b)+[c(e,a)+c(h,a)-a(ex,a)]n(a)-c(e,ex)n(e)-c(h,ex)n(h)-c(ex^+,ex)n(ex^+)-c(ex^-,ex)n(ex^-).$$

The system (3)–(6), the electroneutrality equation

$$n(ex^+)+n(h)=n(ex^-)+n(e)$$

and the equation that determines the excitation level

$$n(h)+n(ex)+n(ex^-)+2[n(b)+n(ex^+)]=n_{cp}$$

constitute jointly the complete system of equations needed to calculate the composition of the exciton gas.

The quantity φ stems from the interaction of the free charges with one another, and we have used for it an expression that approximates the numerical results obtained for a non-ideal plasma[11]:

$$\beta\varphi=(\pi\gamma^3)^{1/2}-0{,}526\gamma^3-0{,}37\gamma^6+0{,}21\gamma^9,$$

where

$$\gamma=\beta e^2[n(e)+n(h)+n(ex^+)+n(ex^-)]^{1/3}\varkappa^{-1}$$

is the non-ideality parameter of the charged subsystem, and $\varkappa$ is the dielectric constant. The value of the coefficient $a(i,j)$ was estimated from the expression

$$a(i,j)=^{16}/_9\pi\varepsilon(i,j)\sigma^3(i,j),$$

where $\sigma(i,j)$ is the effective cross section and $\varepsilon(i,j)$ is the depth of the potential well for the interaction of the i-th and j-th particles. The coefficient $c(i,j)$ was taken in analogy with the interaction between a charge and an atom[12]:

$$c(i,j)=8\pi e^2\alpha/\varkappa R(i,j), \quad (7)$$

where α is the polarizability and $R(i,j)$ is the repulsion radius.

Composition of exciton gas in germanium. Inasmuch as the light particles in germanium amount to not more than 5%, the calculation was carried out only for the heavy particles. We use the following values of the effective masses[13,14] in units of the mass of the free electron in vacuum: $m_{e\parallel}=1.64$, $m_{e\perp}=0.082$, $m_e=0.22$, $m_{0e}=0.12$, $m_h=0.347$. The binding energies used have the following values (in °K): for the exciton—42,[15] for the complexes ex^-—2.2,[10] ex^+—3.2,[10] for the biexciton—1.5.[16] In view of the lack of sufficiently complete data on the excited states and their not too important role at low temperatures, the partition functions were assumed to be equal to the statistical weight of the ground state: for the electron—$2N$,[1)] for the hole—2,[1)] for the exciton—$4N$,[2)] and for the complexes ex^-, ex^+ and the biexciton—$4N$.[3)]

As shown by preliminary estimates, not all kinds of interactions turn out to be significant in the calculation of ΔI. For example, for $T\lesssim 4$ °K, a good approximation is

$$\Delta I_{ex}=2\varphi+[c(e,ex)+c(h,ex)]n(ex),\quad \Delta I_{ex^-}=[c(e,ex)-c(ex^-,ex)]n(ex),\quad \Delta I_{ex^+}=[c(h,ex)-c(ex^+,ex)]n(ex),\quad \Delta I_b=0. \quad (8)$$

The estimate $c(i,j)$ cannot be made sufficiently uniquely.[12,17,18] The values of $c(e,ex)$ and $c(h,ex)$ were chosen with allowance for the experimental data discussed below.[9] This quantity corresponds to reasonable values $R(i,j)=1.5a_0$ and $\alpha=4.5a_0^3$ in (7) (a_0 is the Bohr radius); $c(ex^+,ex)$, $c(ex^-,ex)$ and $c(i,b)$ are smaller, since the values of $R(i,j)$ for them are larger by 2 or 3 times.

The results of the calculation of the composition of the gas phase in germanium are shown in Figs. 1 and 2. The calculation was limited to the region $\gamma<1$.

The role of the Coulomb interaction increases with

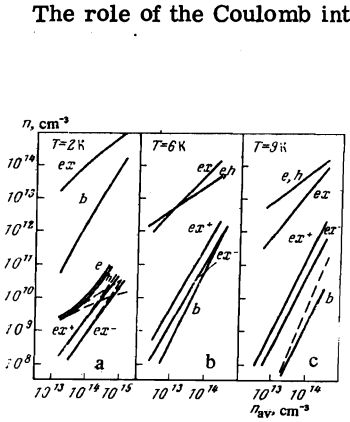

FIG. 1. Isotherms for germanium. The dashed lines show the results of calculation without allowance for the contribution of the charged–neutral interaction.

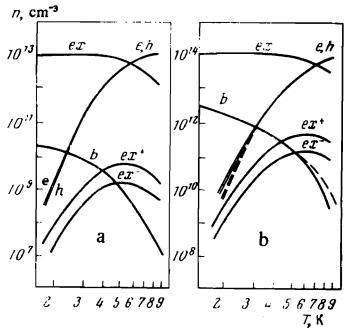

FIG. 2. Isochors for germanium. The dashed lines show the results of calculation without allowance for the contribution of the charged–neutral interaction.

temperature and becomes decisive at $T>4$ °K, in which case in the region $\gamma\approx 1$ the interaction in the charged subsystem increases the concentration of the electrons and holes by one order of magnitude. The charge–neutral interaction greatly increases at $T<3$ °K the concentrations of the electrons and holes and decreases the concentration of the three-particle complexes. In the case $T\gtrsim 8$ °K, when $n(ex)<n(e)$, the charge–neutral attraction exerts a weaker but opposite action: it decreases the concentrations of the electrons and holes and increases the concentration of the excitons. Calculation of the composition of the gas exciton phase in germanium was carried out without allowance for the influence of the impurity atoms. This is permissible if the impurity atom concentration is $n_{\text{imp}}\lesssim 10^{13}$ cm^{-3} and the impurity atoms are in the ground state.

Density–temperature diagram. In Fig. 3 are gathered the available experimental data on the density of the saturated exciton vapor in germanium. In the region $2\lesssim T\lesssim 4.2$ K the measurements were performed by various methods: by conductivity,[9,19] cyclotron absorption,[20,21] luminescence radiation,[22,23] and absorption of light.[24] The results are for the most part in agreement. We note that the data of Lo *et al.*[22] have been corrected in Fig. 3 in accordance with the exciton lifetime 7.7 μsec[26] and the diffusion length 0.04 cm.[3] At $T>4.2$ °K the only available results are those of Thomas *et al.*[23]

The composition calculated in the preceding section makes it possible to draw on Fig. 3 the constant-nonideality curves in the gas phase. The interaction of the charged particles with the neutral ones at $T\lesssim 3$ °K changes strongly the degree of dissociation of the excitons and shifts the position of the $\gamma=1$ curve (curve 9). The $\gamma=1$ curve turns out to be in the region of the start of the exciton condensation, and this serves as an indirect indication of the plasma character of this phase transition.[27] Curve 10 is the line of equal values of

$$\gamma_{ex}=\beta n(ex)\,[c(e,\,ex)+c(h,\,ex)+c(ex^-,\,ex)+c(ex^+,\,ex)\,].$$

Next, on curve 11 we have $\gamma_{ex,ex}=\beta a(ex,ex)n(ex)$, on 12 we have $\lambda_{ex}^3 n(ex)=2.6N$, and on 13 we have $\lambda_b^3 n(b)=2.6N$. In the two-phase region, curves 9–13 are drawn under the assumption that the system remains homogeneous.

Conductivity. The plasma model can be developed also to permit calculation of the conductivity of the exciton gas. In analogy with a plasma, we can consider the scattering of free carriers by excitons and by impurity atoms, and also Coulomb collisions in an electron–hole subsystem. We use here the multicomponent composition considered above for the exciton gas.

The connection between the electric conductivity of a weakly ionized plasma and the diffusion scattering cross section is given by the solution of the Boltzmann kinetic equations. In the Chapman–Enskog approximation the expression for the electric conductivity takes the form[28]

$$\sigma=3/4\,e^2 n_1/n_2 m\bar{v}\langle Q_{12}\rangle, \tag{9}$$

where $\langle Q_{12}\rangle = Q_{12}^{11}(T)/K_{\sigma 12}^{(n)}$ is the averaged diffusion scattering cross section in the n-th Chapman–Enskog approximation,

$$Q_{12}^{11}(T)=1/2\int Q_{12}^{1}(\varepsilon)\,e^{-x}x^2\,dx \tag{10}$$

is the averaged diffusion cross section in the first Chapman–Enskog approximation, $K_{\sigma 12}^{(n)}$ is the correction for the n-th approximation, $x=\varepsilon/kT$, $\varepsilon=mv^2/2$, v is the thermal velocity, m is the reduced mass of the test and field particles, $\bar{v}=(8kT/\pi m)^{1/2}$, n_1 is the concentration of the test particles, and n_2 is the concentration of the field particles.

For a theoretical estimate of $\langle Q_{ea}\rangle$ and $\langle Q_{ha}\rangle$ it is necessary to calculate the values of $Q_{12}^{11}(T)$ and $K_{\sigma 12}^{(n)}$ from the known diffusion cross section $Q_{12}^1(\varepsilon)$. In the isotropic approximation for $T\lesssim 5$ °K, the contribution of the p and d phases is small in comparison with that of the s phase, so that the diffusion cross section coincides with the total cross section. We have used for the theoretical estimates the dimensionless cross section of an electron on a hydrogen atom[29] and of a positron on a hydrogen atom.[30] The Bohr radius a_0 and the ionization potential I were used as scale factors.

Since the functions Q_{ea}^1 and Q_{ha}^1 admit of a power-law approximation ε^μ, it is possible to calculate $K_{\sigma 12}^{(n)}$ in the Lorentz approximation exactly for any value of n. Us-

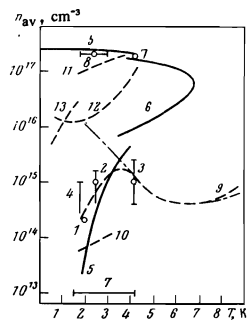

FIG. 3. Density–temperature diagram for germanium. Results of measurements of the coexistence line of the exciton phases: 1—[9], 2—[19], 3—[20], 4—[21], 5—[22], 6—[23], 7—[14], 8—[25]. The constant–non-ideality lines (calculated): 9—$\gamma=1$ (the dash-dot curve shows the results of calculation without allowance for the contribution of the charged–neutral interaction), 10—$\gamma_{ex}=1$, 11—$\gamma_{ex,ex}=1$, 12—$\lambda_{ex}^3 n(ex)=2.6N$, 13—$\lambda_b^3 n(b)=2.6N$.

ing the values of $K_{\sigma 12}^{(\infty)}$ from[31] and formula (10), we can find the cross sections for any temperature. For 2 °K we have $\langle Q_{ea}\rangle = 40\pi a_0^2$ and $\langle Q_{ha}\rangle = 9\pi a_0^2$, and the temperature dependence is weak. These quantities can be used to estimate the cross sections for the scattering of electrons and holes by the impurity atoms. Assuming for germanium $I = 100$ °K and $a_0 = 40$ Å, we obtain $\langle Q_{ea}\rangle = 2 \cdot 10^{-11}$ cm^2 and $\langle Q_{ha}\rangle = 4.3 \cdot 10^{-12}$ cm^2. Measurements[32] yield $\langle Q_{ea}\rangle = 1.35 \cdot 10^{-11}$ cm^2 for $T = 4.2$ °K and $\langle Q_{ha}\rangle = 2.5 \cdot 10^{-12}$ cm^2 for $T = 2$ °K. Somewhat lower values of the experimental scattering cross sections can be attributed to the influence of the anisotropy.

We can estimate in similar fashion the cross sections for the scattering of electrons and holes by excitons. By virtue of the exchange effects, these cross sections are close to each other. Estimates based on the cross section for the scattering of an electron by a hydrogen atom are less reliable in this case, for the role of the anisotropy increases as a result of the smaller value of I. In addition, initial exchange and annihilation effects appear. The experimental value for germanium can be obtained from the widths of the cyclotron-absorption peaks measured in[21]: $\langle Q_{eex}\rangle = \langle Q_{hex}\rangle = 3 \cdot 10^{-12}$ cm^2. The scattering cross section for Coulomb collisions was calculated from Spitzer's formula.[33] It was assumed that the individual scattering mechanisms make an additive contribution to the resistance. The electron and hole conductivities were calculated separately and were added.

The conductivity of the exciton gas in germanium was measured by Gurnee, Gliksman, *et al.*[9] The experimental points in Fig. 4 break up into three groups. At $n_{av} < 2\times 10^{14}$ cm^{-3}, in the opinion of the authors of[9], the exciton system exists in a single-phase state (gas); at $n_{av} = 2\times 10^{14}$ cm^{-3} electron–hole drops come into being, while the composition of the gas phase remains unchanged, so that the conductivity also remains constant. The section of the abrupt growth of the conductivity at $n_{av} > 5\times 10^{15}$ cm^{-3} can be attributed to the appearance of

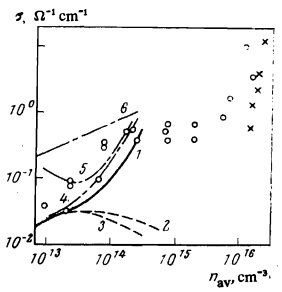

FIG. 4. Conductivity of exciton plasma in germanium at $T = 2$ °K as a function of the total concentration of the electron–hole pairs. Measurement results: —[9], ×—[19] ($T = 2.5$ K). Results of our present calculations—1. Results of calculations for approximate variance of the composition: for the composition obtained without allowance for the charged–neutral interaction—2; for the composition obtained in the ideal-gas approximation—3. The contributions of the individual mechanisms of conductivity to the plot 1: scattering by impurity atoms—4, scattering by free excitons—5, Coulomb scattering in electron–hole system (without allowance for anistropy)—6.

percolation over the contiguous drops. There are also data for this region.[19]

In the present paper we have considered conductivity in an exciton gas, i.e., for $n_{av} < 2\times 10^{14}$ cm^{-3}, see Fig. 4. The results of the calculation (curve 1) carried out using the aforementioned formulas, the experimental values of the scattering cross section, and of the calculated composition, agree satisfactorily with the measurement results. Curves 2 and 3 are shown in Fig. 4 to demonstrate that the composition calculated without allowance for the non-ideality cannot account for the measurement results. The principal role is played in this case in the region $n_{av} \gtrsim 10^{14}$ cm^{-3} by the interactions of the charges with the excitons. Curves 4–6 characterize the role of the different mechanisms. The largest contribution to the resistance, according to our estimates, is made by scattering by the impurity atoms and excitons.[4)] Of lesser significance are the Coulomb collisions (the anisotropy should cause curve 6 to lie even higher). The influence of other scattering mechanisms is negligible. We estimated the collisions of the carriers with phonons, scattering accompanied by transfer of the carriers to another Brillouin band, collisions with spin flips due to exchange and annihilation electron–hole interactions.[34] The contribution of $ex^{\pm}$ also turned out to be negligible at $n_{av} < 2\times 10^{14}$ cm^{-3}.

In the experiments described in[9], the impurity concentration was $n_{imp} = 2\times 10^{13}$ cm^{-3}. At $n_{imp} \lesssim 10^{12}$ cm^{-3}, the $\sigma(n_{av})$ dependence would take the form of curve 5 of Fig. 4, i.e., a section would appear in which the conductivity decreases with increasing pump level. At $n_{imp} \gtrsim 10^{14}$ cm^{-3} the interaction between the charge and the impurity atom becomes appreciable and leads to a noticeable increase of the electron and hole densities. This can also influence the position of the phase-transition point.

We note that the gas composition in the two-phase region can be in disequilibrium because of the appearance of Auger electrons from the electron–hole drops, and the conductivity changes accordingly. This effect is particularly important at low temperatures, when the equilibrium electron density is low.

We are sincerely grateful to L. V. Keldysh for interest in the work, a discussion of the results, and useful remarks.

[1)] The factor 2 takes the spin into account.

[2)] The factor 4 is the result of allowance for the electron and hole spins. The exchange electron–hole interaction does not lead to a change in the degree of degeneracy, since kT greatly exceeds the exchange splitting of the exciton level.

[3)] We assume that two particles of the same sign exist in one valley. Account is taken of the exchange degeneracy for two electrons or two holes. We assume that the splitting due to the exchange electron–hole interaction is smaller than kT.

[4)] We note that on curve 4 the conductivity is of the hole type and on curve 5 it is mainly of the electron type.

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An investigation of photoconductivity in tellurium at low temperatures

V. B. Anzin, Yu. V. Kosichkin, and A. I. Nadezhdinskiĭ

P. N. Lebedev Institute of Physics, USSR Academy of Sciences
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During an investigation of the photoconductivity spectra of tellurium in the 1.6–4.2K temperature range peculiarities have been discovered which indicate the existence of allowed states within the forbidden band near the bottom of the conduction band. The mobility of non-equilibrium electrons and its temperature dependence have been determined. It is shown that the rate of recombination of non-equilibrium carriers is described by a combination of terms linear and quadratic in the concentration.

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The phenomenon of photoconductivity in tellurium is characterized by very specific spectral and relaxation peculiarities which make the interpretation of the experimental results difficult. In preceding investigations at the temperature of liquid nitrogen (see, for example, [1–3]) the presence of a maximum near the intrinsic absorption edge, the position of which depends on the polarization of the exciting light, was revealed, and the non-linear character of the recombination of the non-equilibrium carriers was pointed out. Experiments at the temperature of liquid helium[4] led to the discovery of the existence of an additional maximum in the photoconductivity spectrum situated at energies less than the width of the forbidden band and differing markedly in its characteristics from the remaining parts of the spectrum.

Since the study of this peculiarity has opened up the possibility of clearing up the question of the existence of allowed energy states within the forbidden band, we have carried out a detailed investigation of photoconductivity in samples of tellurium with a hole concentration $p_0 = 2 \cdot 10^{14} - 2 \cdot 10^{16}$ cm^{-3} in the temperature range 1.6–4.2 K.

EXPERIMENTAL RESULTS

The work was carried out in the spectral band 3.5–4.2 μm using a diffraction-grating monochromator. The tellurium samples were placed directly in liquid helium,