CONDENSATION OF NONEQUILIBRIUM CARRIERS IN SILICON

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We investigate recombination radiation produced in silicon at low temperatures and at high levels of stationary photoexcitation. The broad maxima of this radiation are shifted by 15 MeV towards lower energies compared to the free-exciton emission peaks. The intensity of the long wave radiation I_c increases with increasing excitation level, in proportion to the cube of the free-exciton radiation intensity I_e. With increasing temperature, I_c decreases sharply in a narrow temperature interval, whereas I_e increases exponentially with an activation energy of approximately 2 meV. The spectral distribution and the concentration and temperature dependences of the intensity of the long-wave radiation are attributed to the occurrence of a condensed phase consisting of spherical drops of degenerate plasma with electron and hole concentration 3.7×10^{18} cm^{-3}. The characteristic features of the recombination radiation of silicon doped with boron give grounds for assuming that the impurity atoms are the primary condensation centers.

1. INTRODUCTION

A new intense recombination radiation produced in germanium when threshold values of the temperature and of the photoexcitation level are reached was investigated in[1,2]. The origin and the main features of this radiation were attributed to the occurrence of a condensed phase of nonequilibrium carriers, the possible existence of which was pointed out in[3]. Similar radiation was observed earlier by Haynes in silicon[4], in an investigation of photoluminescence at low temperatures and high excitation levels. Figure 1 shows the spectrum of such radiation as recorded by us following electron excitation. It is seen from the figure that besides the well known narrow peaks of radiation with energies 1.136, 1.097, and 1.032 eV, due to annihilation of free excitons with emission of the transverse acoustic (TA), transverse optical (TO), and two TO phonons[5], the spectrum reveals broad emission lines shifted by 15 meV towards lower energies relative to the corresponding exciton peaks.

These new lines were attributed in[4] to formation and radiative decay of exciton molecules—biexcitons, consisting of two electrons and two holes. When an electron and a hole recombine in such a molecule, part of the energy is released in the form of radiation, and part is consumed in the ejection of the second pair of carriers into allowed bands, to which they impart a certain amount of kinetic energy. Since the kinetic energy can be different in different recombination acts, the energy of the emitted photons can also be different, and this should lead to the occurrence of broad radiation bands shifted towards lower energies relative to the exciton peaks. The observed shapes of the radiation bands were not compared with any calculation results. The binding energy of the exciton molecule can likewise not be determined. Starting from the binding energy of the free exciton E_e = 8 meV[5] it was assumed in[4], by analogy with the model of the positronium molecule, that the binding energy of the biexciton is close to $2 \times 10^{-2} E_e = 0.16$ meV.

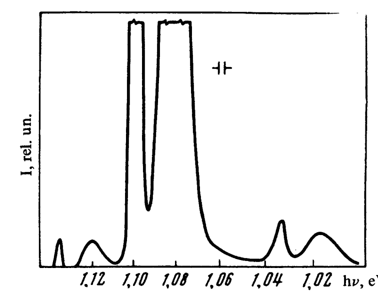

FIG. 1. Spectral distribution of recombination radiation of pure silicon at T ≈ 10°K and under electron excitation (15 kV, 0.5 mA, pulse duration 5 μsec).

Further, since the binding of the excitons into exciton molecules is a bimolecular reaction, one should expect a quadratic dependence of the biexciton concentration n_b on the exciton concentration n_e. It follows therefore that $I_b \sim I_e^2$, where I_e and I_b are the intensities of the exciton and biexciton radiations. In Haynes' opinion[4], it follows from his experimental data (Fig. 2a) that I_e increases linearly with the excitation level g, and I_b quadratically. We shall show later, however, that this conclusion is not sufficiently well founded.

We report in this paper a more detailed investigation of recombination radiation of silicon at low temperatures and at high photoexcitation levels. The results were not in agreement with the model proposed in[4], and were explained as being due to the occurrence of a condensed phase of nonequilibrium electrons and holes[2]. Preliminary results of these investigations were published in[6].

2. EXPERIMENTAL PROCEDURE

We investigated silicon single crystals obtained by crucible-less zone melting, with resistivity ρ from 20 to 3×10^4 ohm-cm, containing boron in concentrations from 3×10^{11} to 5×10^{14} cm^{-3}. When the measurements were made at temperatures 4.2–2°K, the

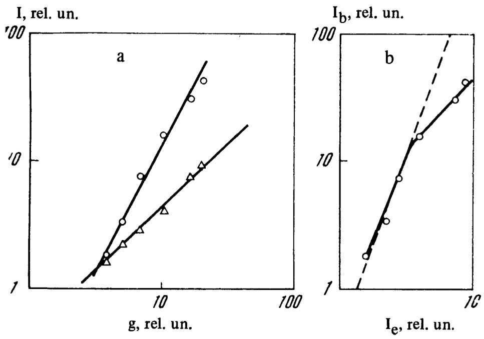

FIG. 2. a—Radiation intensity of free excitons, I_e, and of the long-wave radiation, I_b, vs. the excitation rate g [4]. b—Dependence of I_b on I_e, plotted by us from the data of Haynes [4]. Δ—exciton, O—biexciton, T ~ 3°K.

samples were placed in the helium bath of a metallic cryostat having glass windows for the entry of the exciting radiation and for the exit of the recombination radiation. The temperature was regulated by pumping off helium vapor, was stabilized with a manostat, and was measured by determining the saturated vapor pressure over the surface of the liquid helium. At temperatures above 4.2°K, we used an optical cryostat of the Swenson system[7]. The temperature of the radiating region of the silicon samples was determined in this case from the widths and shapes of the exciton-radiation peaks. The recombination radiation was analyzed by an MDR-2 monochromator and registered by a cooled photoresistor made of n-Ge doped with copper and having a threshold sensitivity of approximately 10^{-14} W-Hz$^{-1/2}$. The ac component of the photoresistor response was amplified, synchronously detected, and registered with an automatic recorder. The apparatus made it possible to record recombination-radiation spectra of silicon with a resolution of approximately 0.4 meV.

The "long-wave" recombination radiation of the silicon was produced only at sufficiently high density of the intrinsic excitation. However, pulsed electronic and optical excitation at a pulse power on the order of several watts led to overheating and to instability of the temperature during the pulse. To ensure a constant and controlled sample temperature, therefore, we used subsequently only stationary excitation with a cw GaAs laser[8] 1). Radiation of a laser with a power up to 300 mW has sharply focused on the surfaces of the samples, thereby ensuring sufficiently high density of stationary photoexcitation. To avoid instability of the temperature in this case, the recombination radiation was modulated. The intensity of the exciting radiation was attenuated in steps by using calibrated grids.

3. ENERGY SPECTRUM OF CONDENSED PHASE AND EMISSION SPECTRUM

Let us consider the change of the energy spectrum of the crystal when a condensed phase of nonequilibrium carriers is produced in it. We start from the following model:

At low temperatures and high excitation levels, the nonequilibrium carriers form two phases—a phase of free excitons and a condensed phase consisting of drops of a degenerate electron-hole plasma. The energy of such a plasma, consisting of mobile carriers of two signs, was not calculated. A similar problem, in which the carriers of equal sign are regarded as mobile, while the carriers of the other sign produce a homogeneous background ensuring electroneutrality of the system, has been solved theoretically quite completely (see, for example[9]). To use the results of these calculations, we shall henceforth assume that the plasma energy is the additive sum of the energies of the electron and hole gases.

In the stationary state, the concentration of the carriers in the condensed phase n_0 should correspond to the minimum of the free energy of the condensate. For a strongly degenerate electron-hole gas (F_e, F_h $\gg kT$), the free energy practically coincides with the total average energy per electron-hole pair, and can be written in the form[9]

$$\tfrac{3}{5}(F_e+F_h)+E_{ex}+E_{cor}. \qquad (1)$$

Here F_e and F_h are the Fermi energies for the electrons and holes, E_{ex} is the exchange energy, equal to the sum of the exchange energies of the electron and hole gases

$$E_{ex}=-2\frac{0.458e^2}{\varepsilon r_0}, \qquad (2)$$

ϵ = 12 is the dielectric constant of silicon, $4\pi r_0^3/3 = n_0^{-1}$, e is the electron charge, and E_{cor} is a correlation correction that takes into account the different types of interaction between electrons and between holes, other than exchange interaction.

The calculation of the correlation energy has been the subject of many papers (see, for example,[9]). E_{cor} can be calculated exactly only for small ($an_0^{1/3} \ll 1$) and large ($an_0^{1/3} \gg 1$) carrier concentrations (a is the Bohr radius). On the other hand, in the region $an_0^{1/3} \sim 1$, one uses different interpolation formulas, which lead, however, to close results. To calculate E_{cor}, we shall use an expression derived by Wigner ([9], formula (3.58)), which can be written in the form

$$E_{cor}=-\frac{0.88}{r_0/a_e+7.8}\frac{e^2}{2\varepsilon a_e}-\frac{0.88}{r_0/a_h+7.8}\frac{e^2}{2\varepsilon a_h}. \qquad (3)$$

Here $a_e = 2\times10^{-7}$ cm and $a_h = 1.4\times10^{-7}$ cm are the Bohr radii for the electrons and holes in silicon[10]. Recognizing that

$$F_e, F_h=\frac{\hbar^2}{2m_{e,h}}\left(\frac{9}{32\pi^2}\right)^{2/3}\frac{1}{r_0^2}, \qquad (4)$$

where $m_e = 1.08m$ and $m_h = 0.55m$ are the effective masses for the density of states of the electrons and holes in silicon[10], we get from (1) $r_0 = 3.9\times10^{-7}$ cm, corresponding to a minimum of the condensate energy. Substitution of this value into (2) and (4) yields $E_{ex} = -29$ meV, $E_{cor} = -6.2$ meV, and $(3/5)(F_e + F_h) = 14$ MeV. The electron and hole concentration corresponding to the minimum of the condensate energy is $n_0 = 4.2\times10^{18}$ cm^{-3}. Thus, the average energy per electron-hole pair in a condensed plasma is smaller by approximately 20 meV than the energy of the free electron-hole pairs, and by 10 meV than the energy of the free exciton. Consequently, formation of a con-

1)The authors are grateful to P. G. Eliseev and V. P. Strakhov for supplying a laser capable of stable operation for several hundred hours.

sequently, formation of a condensed phase of nonequilibrium carriers is favored energywise [2)].

Let us consider the spectral distribution of the radiation produced as a result of recombination of electrons and holes in a nonequilibrium plasma. Following[11], we shall assume that the radiative-transition probability is practically independent of the energy E of the electrons and holes. Then the radiation intensity should be determined only by the density of the initial and final states, for which we can put N_e, $N_h \sim E^{1/2}$. We also recognize that the law of quasimomentum conservation in radiative recombination of electrons and holes is satisfied as a result of emission of phonons with quasimomenta corresponding to the minima of the conduction band, and it is therefore necessary to consider all possible band-band transitions corresponding to a given phonon energy $h\nu$. In this case the spectral radiation density of the condensate is

$$I(h\nu - h\nu_0) \sim \int_A^B E^{1/2}(h\nu - h\nu_0 - E)^{1/2}\, dE \tag{5}$$

with integration limits

$$\begin{gathered} 0 \leqslant h\nu - h\nu_0 \leqslant F_e : A = 0, \quad B = h\nu - h\nu_0, \\ F_e \leqslant h\nu - h\nu_0 \leqslant 2F_h : A = 0, \quad B = F_e, \\ 2F_h \leqslant h\nu - h\nu_0 \leqslant F_e + F_h : A = h\nu - h\nu_0 - F_h, \quad B = F_e. \end{gathered} \tag{6}$$

Here $h\nu_0 = E_G + E_{ex} + E_{cor} - \hbar\omega$ corresponds to the long-wave edge of the emission band, E_G is the width of the forbidden band in "pure" silicon, and $\hbar\omega$ is the energy of the emitted phonon.

We note that expressions (5) and (6) depend only on the concentration n_0 for given values of the effective masses. The spectra calculated in accordance with (5) and (6) were tabulated and compared with the experimental spectra. Figure 3 shows part of the spectrum of the recombination radiation of pure silicon, accompanied by emission of a TO phonon with $\hbar\omega = 58$ MeV[5]. The circles in the figure represent the spectral radiation density calculated in accordance with (5) and (6) for $n_0 = 3.7 \times 10^{18}$ cm^{-3}, corresponding to optimal agreement between experiment and calculation. It is seen from the figure that agreement is actually observed everywhere except in the low-energy region. This discrepancy is natural, owing to the occurrence of "tails" of the allowed bands, a fact not taken into account in the calculation. One should also note the good agreement between the values of n_0 determined from the condition for the minimum of (1) and from the condition of optimal agreement between the experimental and calculated spectra.

The long-wave emission edge should correspond to $h\nu_0 = E_G + E_{ex} + E_{cor} - \hbar\omega$, with $E_G = 1.163$ eV[5]. Calculation according to (1) gives $E_{ex} + E_{cor} = -35$ meV, as against -38 meV in experiment. This agreement should also be regarded as satisfactory, if it is recognized that no account was taken in the calculation of the correlation between the electrons and holes; this correlation can greatly increase the binding energy of the condensate.

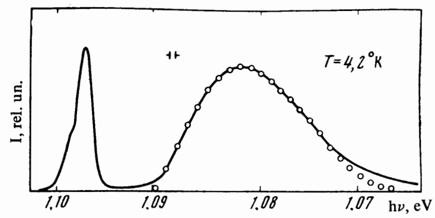

FIG. 3. Spectral distribution of the laser-excited recombination radiation of pure silicon ($\rho \approx 3 \times 10^4$ ohm-cm) at 4.2°K, accompanied by emission of a TO phonon. The spectrum calculated from formula (5) for $n_0 = 3.7 \times 10^{18}$ cm^{-3} is represented by the circles.

4. DEPENDENCE OF THE RECOMBINATION-RADIATION INTENSITY ON THE EXCITATION LEVEL

We shall assume that the condensed phase consists of spherical drops of radius R. Then the main features of the concentration and temperature dependences of the radiation intensity of the free excitons and of the condensed phase can be established by starting from the following simple considerations:

If R is significantly smaller than the exciton mean free path, then under stationary conditions[2] the flux of free excitons on the surface of the drop $\pi R^2 v n_e$ should be equal to the sum of the recombination flux inside the drop $4\pi R^3 n_0/3\tau_0$ and the flux $4\pi R^2 A \exp(-\varphi/kT)$ through the surface of the drop into the volume of the crystal as a result of thermal ejection of the carriers:

$$\pi R^2 v n_e = \frac{4}{3}\pi R^3 \frac{n_0}{\tau_0} + 4\pi R^2 A e^{-\varphi/kT}. \tag{7}$$

Here n_e is the concentration of the excitons, v their average thermal velocity, τ_0 the lifetime of the nonequilibrium carriers in the condensed phase, φ the work function, and A a coefficient that depends little on the temperature. At low temperatures, the thermal ejection of carriers can be neglected. Taking into account the fact that the intensity of the exciton radiation is $I_e \sim n_e$ and the intensity of emission of the condensed phase is $I_c \sim 4N\pi R^3 n_0/3\tau_r$, where τ_r is the radiative lifetime in the condensate and N is the concentration of the condensate drops, we find from (7) that

$$I_c \sim N I_e^3. \tag{8}$$

It can be assumed that the concentration N changes little when the excitation level or the temperature changes, and is given, for example, by the concentration of the condensation centers. Then the radiation intensity of the condensed phase I_c should be proportional to the cube of the radiation intensity of the free excitons: $I_c \sim I_e^3$. Such a dependence for one of the samples of pure silicon at 4.2°K is shown in Fig. 4. It was well satisfied also for all the other samples, so long as the photoexcitation power did not exceed the critical value. In the investigation of the dependence of I_c on I_e, there is no need to know the absolute change in the carrier excitation rate g. The dependence of I_e and I_c on g has a more complicated character (Fig. 5). However, even in the region of a strong variation of the dependences of I_e and I_c on g, relation (8) was well satisfied in the experiment.

2)If it is assumed that a significant contribution to the exchange energy is made only by electrons belonging to one valley of the conduction band, then the concentration calculated from the minimum of (1) is $n_0 = 2.2 \times 10^{18}$ cm^{-3}, and $E_{ex} + E_{cor} = -25$ meV.

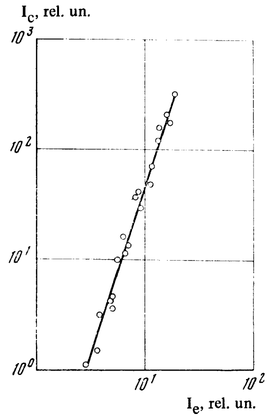

FIG. 4. Dependence of I_c on I_e for pure silicon ($\rho \approx 3 \times 10^4$ ohm-cm) at 4.2°K and laser excitation.

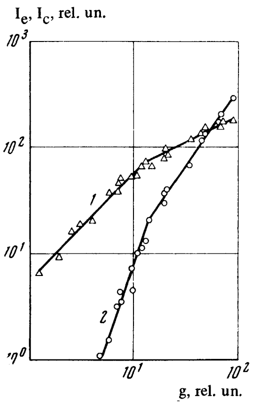

FIG. 5. Dependence of I_e (curve 1) and I_c (curve 2) of the sample of pure silicon on the excitation rate g at 4.2°K.

We note that a cubic dependence of I_c on I_b actually was obtained in Haynes' experiment[4] at low excitation levels. This dependence, plotted by us from Haynes' data, is shown in Fig. 2b. It is seen from the figure that the deviation from a cubic dependence is observed only at large excitation levels. A similar deviation from the cubic dependence (8) was observed by us also at high excitation power and was accompanied by overheating of the samples, causing a broadening of the exciton-radiation lines.

An appreciable overheating of the silicon samples apparently took place also in[12], where the photoexcitation source was a ruby laser. According to the estimates of the authors of[12], the overheating should not exceed 10°K. However, even at such a temperature rise the main laws governing the kinetics of the recombination radiation can be greatly distorted.

5. TEMPERATURE DEPENDENCE OF THE RECOMBINATION-RADIATION INTENSITY

The main problem in the investigation of the temperature dependence of the radiation intensity in the region 4.2–15°K lies in the actual measurement of the temperature of the radiating region. Indeed, in the indicated temperature interval it is impossible to immerse the sample in liquid hydrogen or helium so as to ensure a sufficiently intense heat conduction. On the other hand, placing the sample in a stream of helium gas does not ensure satisfactory thermal coupling between the sample and any thermometer. If we take into account the relatively large power dissipated by the sample when it is intensely excited, the ensuing difficulties can be readily understood.

In our opinion, the most correct method of measuring the temperature is to determine it from the widths and shapes of the emission lines of the free excitons. Indeed, as shown in[13], at relatively high temperatures the spectral distribution of the intensity of the excited radiation is well approximated by Boltzmann distribution. However, at low temperatures there appears a finite width of the exciton lines, connected both with the finite lifetime of the TO phonon emitted in radiative annihilation of the excitons, and with the splitting of the exciton lines into two closely lying peaks[14]. Therefore, to determine the temperatures we used the values of the half-width of the exciton line, determined experimentally for a number of fixed temperatures (2, 4.2, 20.4, and 78.3°K), interpolating linearly the temperature dependence of the half width between these points. This method also made it possible to eliminate the influence of the finite resolution of the spectral instrument.

Figure 6 shows the temperature dependence of I_e and I_c for a sample of pure silicon. This dependence can be explained on the basis of (7).

Indeed, Eq. (7) can be represented in the form

$$aI_e - bI_c^{1/3} = e^{-\varphi/kT}. \tag{9}$$

The constants a and b can be determined in the following manner: relation (7) is valid only in the region of existence of the condensed phase ($R \geq 0$, $I_c \geq 0$), when the temperature T is lower than the threshold value T^{thr}. Recognizing that I_e remains practically constant after the radiation of the condensate disappears, we denote the relative intensity of the exciton radiation at $T \geq T^{thr}$ by I_e^{thr}. An investigation of the temperature dependence of I_e and I_c at $T < 4.2$°K has shown that both intensities become constant at $T \leq 3$°K. We therefore denote I_e and I_c at low temperatures by I_e^0 and I_c^0, respectively. We can then write (9) in the form

$$\frac{I_e}{I_e^{thr}} - \frac{I_e^0}{I_e^{thr}}\left(\frac{I_c}{I_c^0}\right)^{1/3} = \exp\left[-\frac{\varphi}{k}\left(\frac{1}{T} - \frac{1}{T^{thr}}\right)\right]. \tag{10}$$

Since $I_c \leq I_c^0$ and $I_e \gg I_e^0$ the term containing I_c in (10) can be neglected in practically the entire temperature interval shown in Fig. 6. Then

$$I_e \sim \exp\left[-\frac{\varphi}{k}\left(\frac{1}{T} - \frac{1}{T^{thr}}\right)\right].$$

On the other hand, at high temperatures, when I_c decreases rapidly and $I_e \approx I_e^{thr}$, it follows from (10)

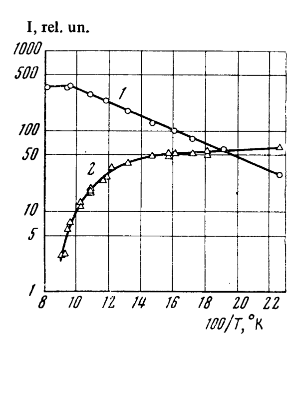

FIG. 6. Temperature dependence of I_e (curve 1) and I_c (curve 2) for a sample of pure silicon excited with a laser.

that

$$I_c \sim \left[1 - \exp\left\{-\frac{\varphi}{k}\left(\frac{1}{T} - \frac{1}{T^{\text{thr}}}\right)\right\}\right]^3.$$

Thus, with increasing temperature the decrease of the radiation intensity of the condensate I_c should be much sharper than the increase of the intensity of the exciton radiation I_e. This is clearly seen from Fig. 6. The work function φ, determined from the temperature dependence of I_e and I_c in Fig. 6, turned out to be approximately 2 meV. This value, however, is only tentative, since the temperature of the condensate drops may exceed the temperature of the crystal lattice of the silicon as a result of the release of the heat of condensation and as a result of the weak interaction between the degenerate electron-hole gas and the lattice[15].

As already noted, the binding energy of the biexciton, according to the estimate (4), should be on the order of 0.16 meV. It is seen, however, from Fig. 6 that the intensity of the long-wave radiation I_c remains practically constant up to 7°K. The existence of an exciton molecule with such a low binding energy up to 7°K is doubtful.

6. POSSIBLE NATURE OF THE CONDENSATION CENTERS

So far, in the analysis of the experimental results, we have considered only a stationary state in which dynamic equilibrium obtains between the condensed and "gaseous" phases. In analogy with other first-order phase transitions, one should expect an appreciable role to be played in the formation of the condensed phase by the condensation centers. Such centers may be, generally speaking, any crystal-lattice defects (impurity center, dislocation, etc.) causing a local deformation.

To verify this assumption, we investigated the recombination radiation of silicon doped with boron in relatively small concentrations. At a low photoexcitation level, the spectra of these samples revealed only radiation peaks of free excitons ($h\nu$ = 1.0982 and 1.0964 eV) and peaks with $h\nu$ = 1.0942 and 1.0924 eV, resulting from radiative decay of the excitons bound on the boron atom[5]. With increasing excitation level (Fig. 7), three new peaks appear in the spectrum, with $h\nu$ = 1.0903, 1.0881, and 1.0863 eV. These energies do not correspond to radiation peaks of the excitons bound on some impurity atoms of group III or V element[5]. An increase of the excitation level (Fig. 8) leads first

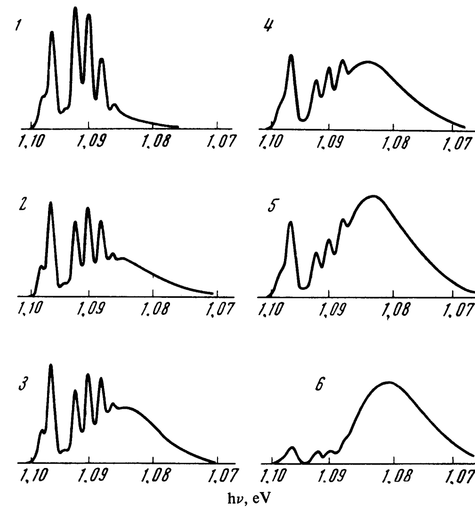

FIG. 8. Change of recombination-radiation spectra of silicon with $N_B = 5 \times 10^{12}$ cm^{-3} with changing laser focusing. The excitation density increases from spectrum 1 to spectrum 6. Temperature 2°K.

to the appearance of a long-wave "tail" of radiation, developing subsequently into a broad peak whose maximum shifts towards lower energies with increasing excitation level. At a high excitation level, this peak does not differ in shape or position from the corresponding emission peak of the condensate in pure silicon (Fig. 8b).

The foregoing results allow us to assume that when the excitation is increased complexes containing more than one exciton can be produced on the impurity atoms; this leads subsequently to the occurrence of condensate drops around such complexes. The interaction between the complexes and the carriers of the condensed phase leads to a broadening of the emission lines of the complexes, until the entire line structure of the radiation vanishes at large excitation levels. In silicon with a larger boron content (5×10^{14} cm^{-3}), the appearance of a condensate radiation peak with increasing excitation level is not accompanied by the occurrence of free-exciton radiation[6], so that the formation of the condensed phase occurs practically without the free-excitons taking part.

The shift of the maximum of the radiation of the condensate towards larger energies with decreasing excitation level may be connected with a decrease in the dimension of the condensate drops. Indeed, the distortion of the potential in the boundary layer of the condensed phase can lead to a lifting of the bottom of the potential well in which the condensed carriers are localized. At small drop dimensions, the influence of the boundary layer should appear more strongly, and this can lead to the observed shift of the maximum on the condensate radiation.

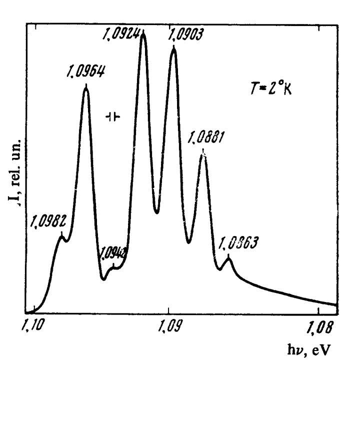

FIG. 7. Spectral distribution of TO component of the recombination radiation of silicon doped with boron ($N_B = 5 \times 10^{12}$ cm^{-3}) at 2°K and under excitation with defocused laser radiation.

7. CONCLUSION

The foregoing results show that the spectral distribution of the recombination radiation produced in silicon when the threshold temperature and excitation levels are reached, as well as the main dependences of the intensity of this radiation on the excitation level and on the temperature, can be quantitatively explained by using a model of a condensed phase consisting of spherical drops of a degenerate electron-hole plasma. The characteristic features of the spectra of doped silicon show that the primary condensation centers may be the impurity atoms.

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