

H⁻-like centers and delocalization of electrons in semiconductors

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(Submitted 28 February 1983)

Zh. Eksp. Teor. Fiz. **85**, 746–763 (August 1983)

The localization-delocalization transition for weakly bound ionic (H⁻-like) states in a disordered system of neutral impurity atoms is studied. The delocalization-density threshold N_{cr} is determined mainly from the photoconductivity spectra in the particular case of doped and weakly compensated silicon. Because of the large falloff length of the wave function ($a_i \approx 4.2a_0$, where a_0 is the effective Bohr radius), the delocalization of the ionic states and formation of a conducting impurity band of the upper Hubbard band (UHB) type occurs at significantly lower neutral impurity densities (N_{cr}) than for the insulator-metal transition (N_M) ($N_{cr} \approx 0.015 N_M$). The effect of the charge-center density on N_{cr} is investigated. It is shown that at $N \gtrsim N_{cr}$ the delocalized states and localized states of the H⁻ – H⁺ type coexist. The effect of excited impurity states on the lifetimes of the nonequilibrium carriers in the UHB is studied. It is shown that when the energy positions of the UHB and of the excited impurity states coincide the excited-state localization decreases with increasing N .

PACS numbers: 71.55.Dp, 72.40. + w, 72.20.Jv

1. INTRODUCTION

One of the most widely used concepts in recent physics literature devoted to disordered system is “electron localization” (or its inverse, “delocalization”). Localization is discussed in several reviews,^{2–5} in which, in particular, a number of unsolved problems are formulated. A typical example of localization is the metal-insulator transition that occurs in doped semiconductors when the density (N) of shallow donors is decreased.¹ Such a transition manifests itself in experiment, for example, by the change of the static conductivity σ with N as $T \rightarrow 0$.¹ In a theoretical analysis of this transition one considers an assembly of hydrogenlike impurity atoms, each having a single electron with a wave function

$$\psi(r) \propto \exp(-r/a_0), \quad a_0 = \hbar / (2mE_0)^{1/2},$$

(a_0 is the effective Bohr radius, E_0 is the Bohr energy of the ground state). In uncompensated semiconductors the transition takes place at an impurity density $N = N_M$ satisfying the condition (Mott transition)

$$N_M^{1/3} a_0 \approx 0.25. \quad (1)$$

This article is devoted to the study of another transition from localized to delocalized states in a disordered system. It takes place at a neutral-center density $N = N_{cr} \ll N_M$. This transition is possible because the neutral impurity atom can connect also an “extra” electron into a state called ionic, with a binding energy E_i much lower than E_0 ($E_i \approx 0.55E_0$, Ref. 6) and with a wave function $\psi_i(r)$ whose fall-off length (a_i) exceeds a_0 :

$$\psi_i(r) = (2\pi a_i)^{-3/2} \frac{\exp(-r/a_i)}{r}, \quad a_i = \frac{\hbar}{(2mE_i)^{1/2}} \approx 4.2a_0. \quad (2)$$

The result is a system such as the negative hydrogen ion H⁻, namely a negatively charged donor D^- or a positively charged acceptor A^+ (see, e.g., Ref. 6 and the literature cited there). Of importance to the “excess” electron are the short-

range potential of the neutral atom and the Coulomb potential. The latter is due to the presence of compensating impurities and (at a degree of compensation $K \rightarrow 0$) to those charged donors which have acquired or lost an electron. The small (compared with E_0) binding energy of the extra electron is attributed in essence to the repulsion potential of the “main” electron. The classification of the states of an assembly of disordered centers with short-range potential and shallow density, $N^{1/3} \ll a_i^{-1}$, was carried out by Lifshitz.⁷ He has shown that in this case there can exist localized states on isolated centers and on pairs. States of this type for extra electrons of isolated H⁻-like centers and complexes analogous to the negatively charged hydrogen molecule H_2^- were observed in experiment in doped semiconductors with $N^{1/3} \ll a_i^{-1}$ and $K \ll 1$ (see Ref. 6). Also observed⁶ were states due to the Coulomb and short range potential of the type of hydrogen molecule H⁻–H⁺ in the ionic state; these states have energies inside the Mott-Hubbard gap.

It is clear that when the density of the neutral centers is increased (in the absence of charged ones) at $N = N_{cr} = \beta a_i^{-3}$, where $\beta \approx 1$, the overlap of the “ionic” states should become large enough for their delocalization.³ This produces in the disordered system a conducting impurity band with nonactivated conduction—the analog of the upper Hubbard band (UHB) or of the D^- band.¹ It is assumed that it is precisely in this band that the so-called static σ_2 conduction is produced, with an activation energy ε_2 :

$$\sigma_2 = \sigma_{02} \exp(-\varepsilon_2/kT), \quad (3)$$

which is observed in many semiconductors in a narrow range of impurity densities, $0.1N_M < N < N_M$, preceding the Mott transition.¹ The proof of strong overlap of the state in the UHB is taken to be the fact that according to the experimental data σ_{02} increases with N in accord with a power law (see Ref. 9 and the references therein):

$$\sigma_{02} \propto N^\gamma, \quad \gamma = 0.75 \quad (K < 0.5). \quad (4)$$

A comparison of (4) with the threshold dependence of the conductivity of the "basic" electrons on N near the metal-insulator transition at $N \gtrsim N_M$ (Ref. 10) ($\sigma(N) \propto (N/N_M - 1)^\nu$, $\nu \sim 1$) shows directly that the conductivity (3) is observed only at densities $N \gg N_{cr}$, i.e., far from the delocalization threshold. The apparent reason is that to observe the σ_2 conduction it is necessary that it exceed the competing true band conduction ($\sigma_1 = \sigma_{01} \exp(-\varepsilon_1/kT)$) and the conduction due to hopping between states near the Fermi level ($\sigma_3 = \sigma_{03} \exp(-\varepsilon_3/kT)$).⁵

It was already noted in Ref. 11 that more favorable for the determination of N_{cr} are measurements of the photoconductivity (PC) due to transitions of the excess electrons from the localized states or from the UHB into the conduction band. The present paper is a continuation and development of Ref. 11: the PC spectra are used to study the delocalization of the excess electrons and to determine the density threshold N_{cr} of the delocalization in n - and p -type silicon (Si:P and Si:B) with compensation $K < 10^{-2}$ and density $N = (5 \cdot 10^{-3} - 5 \cdot 10^{-1})N_M$. We study the effect of the temperature and of the density N_i of the ionized impurities on N_{cr} . It is observed that, in analogy with the case of the Mott transition in compensated semiconductors,⁵ N_{cr} increases with increasing N_i . At small values of N_i the value of N_{cr} is $\approx 2 \times 10^{-2} N_M$, or approximately one-tenth of those values of N at which the conductivity (3) is observed (see Refs. 12 and 13 and Table I). We study the influence of a magnetic field on the PC at $N \approx N_{cr}$. It is shown that at $N \gg N_{cr}$ delocalized and localized states of the type $H^- - H^+$ exist; the presence of the latter produces the fluctuation potential that "hampers" delocalization. We study the factors that determine the lifetimes of photoexcited carriers in the UHB and in localized states. It is observed that at $N \gtrsim N_{cr}$ the essential channel for the annihilation of complexes such as $H^- - H^+$ is ejection of the electron from the complex in the UHB, followed by migration over this band to the attracting center and by capture of the electron by the center.

Absorption was studied in n -Si with $N \approx 2-5 \cdot 10^{-2} N_M$ and it was shown that it has characteristic peaks at frequencies corresponding to transitions of the electrons from the ground state to the excited states (ES) $2p_0$ and $2p_{\pm}$. It was observed that in these optical transitions, when the UHB

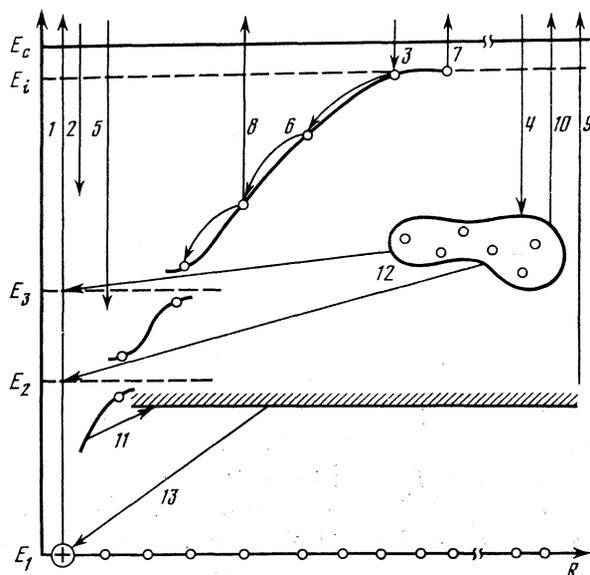


FIG. 1. Model and basic physical processes. The transition 14 (see the text) is not shown in the figure.

coincides with the energies of the corresponding ES, a photoconductivity is produced by the non-activating transition of the electrons from the ES to the UHB. In this connection, the question of the presence in one energy region of both ES of the impurity centers and localized and delocalized ionic states.

We note that at present there is no sufficiently complete theoretical analysis of the UHB (or the D^- band) in doped semiconductors. Thus, in Refs. 14 and 15, and in Refs. 16 based on Ref. 14, ε_2 and N_{cr} were calculated without allowance for the long-range Coulomb potential, and in analogous calculations¹⁷ an ordered arrangement of the impurities was assumed.²⁾ The experimental studies devoted to the group of questions considered here will be discussed below.

2. BASIC PHYSICAL PROCESSES

We analyze qualitatively the processes that can be important in the study of delocalization under nonequilibrium conditions, using as an example an n -type semiconductor (see Fig. 1).

TABLE I.

		Si:P= n															
№		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
N	$10^{17}, \text{cm}^{-3}$	0.2	0.35	0.55	0.70	0.90	1.2	1.6	1.9	2.5	3.5	4.2	5.8	6.7	10	12	14
ε_1	meV	44	44	44	44	44	44	44	44	43	40	35	26	22	18	13,9	9,5
ε_2	meV	3.4	3.9	4.5	5.0	5.2	5.7	6.3	6.7	7.5	8.0	7.2	5.9	5.1	4.0	2.4	2.0
ε_3	meV																
		Si:B= p															
№		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	
N	$10^{17}, \text{cm}^{-3}$	0.12	0.20	0.33	0.45	0.68	0.96	1.2	1.8	3.0	3.3	3.7	6.2	8.0	9.8	16.5	
ε_1	meV	45	45	45	45	45	43	42	40	36	34	32	27	23	22	21	
ε_2	meV	2.4	3.0	3.7	5.1	6.0	6.2	7.0	7.8	21	21	20	16	12.5	10	6.0	
ε_3	meV									8.3	8.3	8.2	6.8	6.0	5.5	5.0	

Background radiation (from the warm parts of the light pipe) of intensity Φ causes photoionization (1) of the neutral centers: positively charged donors and electrons are produced in the conduction band (with respective densities N^+ and n). Several mechanisms of electron capture from the conduction band by isolated impurities and by impurity formations are possible: by an attracting donor [direct recombination (2) with cross section σ_+] (see Ref. 18); by a neutral donor (3) whose ionic state overlaps weakly the ionic states of the neighbors (process (3) with cross section σ_{is} , see Refs. 19 and 20); in this case negative donors D^- are produced with density N^- . Also possible is capture (4) by a group of neutral donors whose ionic states overlap (finite cluster, cross section σ_{cl} , Ref. 17); capture by a complex of the type $H^- - H^+$ (5) with distance $R \approx N^{-1/3}$ between the nuclei ("dipole," cross section σ_D , Ref. 18). The latter is produced because of the hopping (6) of the excess electrons over the neutral centers towards the attracting D^+ (Ref. 6). The cross sections σ_+ , σ_{cl} , and σ_D depend, generally speaking, on the distance between the neighboring centers. The ratio of these cross sections is known¹⁸:

$$\sigma_+ > \sigma_D > \sigma_{is}. \quad (5)$$

The cross section σ_{cl} depends on the size and properties of the cluster, and is unknown. For all the listed capture mechanism there exist inverse processes—thermal and background ejection of an electron into the conduction band. Their probabilities are $W_T \propto \exp(-E/kT)$ and $W_b \propto \Phi$ (E is the electron binding energy). Estimates and experimental data offer evidence that $n \ll N_i$ and $N^+ \ll N$.⁶

As regards the cluster properties it can be noted that the most probable is formation of a cluster of centers far from a charged one ("small-displacement" centers⁶). With increasing N the cluster increases in size, but it decreases with increasing density N^+ or N^- of the ionized centers, owing to the fluctuation straggling of the levels.

The ground states of the impurities at $N \ll N_M$ are assumed to be almost isolated. The states ("ionic" for the "excess" electron can be either localized (H^- , H_2^- , $H^- - H^+$) or delocalized. In the latter case they form a UHB. It is assumed that the lower edge of the band lies higher in energy than the localized states. Three optical transitions (not shown in Fig. 1) are possible from the ground states of the transition: to excited states of the same center, to an ionic state of a neighboring center with formation of a complex of the $H^- - H^+$ type,²¹ and to a finite cluster. The first of them will be discussed in Sec. 4. The last two transitions make no contribution to the PC and will not be considered here; experimental data and estimates of the probabilities of these transitions are given in Ref. 21.

A contribution to the PC can be made by transitions of several types with different frequency dependences of the cross sections. The frequency dependences $\sigma_{H^-}(\hbar\omega)$, $\sigma_{H^- - H^+}(\hbar\omega)$ of the transitions (7) and (8) from localized states of type H^- or $H^- - H^+$ into the conduction band is characterized by a short-wave falloff at frequencies such that $\hbar\omega - E \gg \hbar^2/2ma^2$, where a is the falloff length of its wave function. This falloff is due in essence to the decrease of the overlap of the initial (localized) and final (delocalized) states

of the photoelectron with increasing photon frequency. The square of the transition matrix decreases then with $\hbar\omega$ more strongly than the final-state density increases. The cross sections for the optical transitions from the UHB (9) or from the final cluster (10) to the conduction band are not known; qualitative ideal concerning the frequency dependences of the corresponding cross sections are given in Sec. 4.

A contribution to the PC can be made, in addition, by transitions from the localized states, the ground state or the complex $H^- - H^+$ to the UHB (11). The frequency dependences of the corresponding cross sections are likewise unknown. They should be determined by the sign of the dispersion in the UHB at energies comparable with the energy gap between the bottom of the UHB and the localized states.

Besides the mechanisms indicated above, a contribution to the PC could be made in the indicated frequency band by transitions not shown in Fig. 1, from the excited impurity states to the conduction bands, as well as by transitions corresponding to their photothermal ionization.²² Under the conditions of our experiments with n -Si and p -Si and of experiments with n -Ge (see, e.g., Ref. 23), however, no PC corresponding to the transitions is observed, although the excited impurity states are still fully resolvable in the investigated density range.²¹ Since the PC is proportional to the number of electrons in initial states, it must be assumed that the number of electrons in excited impurity states is much less than in localized states of the type H^- and $H^- - H^+$, and also in UHB. Under background excitation this is possible if the times of the intracenter background transitions from the excited states to lower ones (τ_1) are much shorter than the lifetimes of the electrons in the complexes $H^- - H^+$ (τ_2), in the final clusters (τ_3), and in the UHB (τ_4). Therefore the times τ_2 to τ_4 should be determined by the corresponding probabilities of the phonon transitions to excited impurity states (transitions 14, 12, and 13 in Fig. 1). These probabilities are unknown. It can be shown that the probability for transition 14 should be largest (of the order of the probability of an intracenter transition) if the energy of the electron in the $H^- - H^+$ system coincides with the energy of the excited impurity state) pseudocrossing of the terms.⁶ Away from the pseudocrossing, the probability contains a small factor $\propto \exp(-2R/a^*)$, where a^* is a combination of the falloff lengths of the initial and final states. The probability of a transition of the excess electron in a complex of the type $H^- - H^+$ directly to the ground state of the donor is usually small, $\propto (qa_0)^{-1}$, $l > 1$, where q is the wave vector of the phonon emitted in the transition, $q^{-1} \ll a_0$.

3. EXPERIMENTAL PROCEDURE AND DATA REDUCTION

The physical situation described above, chosen for the solution of the principal problem, the study of delocalization H^- -like states, predetermined the experimental procedure and conditions. We investigated mainly the manifestation of delocalized "excess" carriers in the spectra of the long-wave infrared ($\lambda = 40$ – $1000 \mu\text{m}$) photoconductivity PC, in the static conductivity at low temperatures ($T = 1.5$ – 20 K), and in impurity (background) excitation of the free carriers in a wide range of electric ($E = 1$ – 250 V/cm) and magnetic

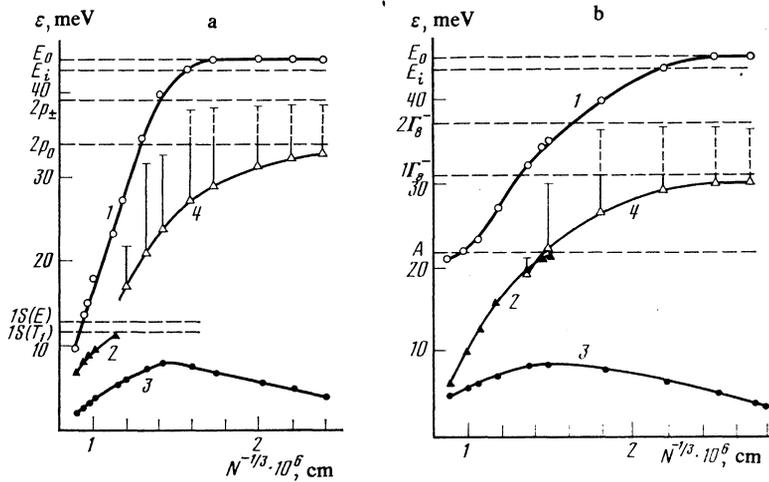


FIG. 2. Dependence of the characteristic values of $\varepsilon_1(1)$, $\varepsilon_2(2)$, $\varepsilon_3(3)$, as well as of $\varepsilon_{2\text{opt}}$ (4) on the impurity density at the minimum values of N^- and N^+ . The range of variation of $\varepsilon_{2\text{opt}}$ with increasing N^- and N^+ is indicated; a) Si:P, b) Si:B.

($H = 0-50$ kOe) fields. We used in the experiments weakly compensated ($K \leq 10^{-3}$) Si crystals doped with phosphorus or boron ($N = 10^{16}-10^{18}$ cm $^{-3}$). The donor and acceptor densities N_D and N_A , as well as the characteristic values of the energies ε_1 , ε_2 , ε_3 , were determined from standard measurements of the temperature dependences of the Hall constant and are given in Table I as well as in Figs. 2a, b. The samples were thin plates measuring $5 \times 1 \times 0.2$ mm, to produce uniform carrier generation in the crystal thickness, and the contacts were placed on the long side to decrease the resistance. The spectral measurements were made with a Fourier spectrometer (photon energy range $\hbar\omega = 1-30$ meV) and an echelette monochromator ($\hbar\omega = 3-50$ meV). The PC spectra were normalized to the number of photons. We measured the stationary conductivity σ_c and the integral PC ($\delta\sigma/\sigma_c$) produced by radiation of energy $\hbar\omega = 1-30$ meV. To determine the conductivity σ_b due to the impurity background Φ under conditions when the static (dark) conductivity is significant, we investigated simultaneously two identical samples. One was shielded by a metallic foil, and σ_b was defined as $\sigma_c - \sigma$.

In the study of the influence of the charged centers on the delocalization of the ionic states in a given sample, the values of N^+ and N^- were varied by varying the external factors Φ , E , and T . The value of Φ was varied by placing in front of the sample cold filters of polyethylene with different lampblack contents. The relative density N^-/N of the "excess" electrons in the localized (n_L/N) and delocalized (n_d/L) states ($N^- = n_L + n_d$) was determined from the values of $\delta\sigma$ and $\delta\sigma/\sigma$ in three particular cases.

1. At small N , when the annihilation of complexes of the type $H^- - H^+$ is negligible compared with the other destruction processes ($W_A < W_T + W_b$), and no UHB has yet been produced:

$$\sigma_b = \sigma_0, \quad N^- = n_L, \quad \frac{\delta\sigma}{\sigma} \propto \frac{W_c}{W_b}, \quad \delta\sigma \propto N^-/N, \quad (6)$$

where $W_c N^-$ is the integral intensity of carrier generation by long-wave infrared radiation.

2. At large N , when conduction through the UHB (σ_{cond}) is possible, annihilation is significant and the PC is due to electron transitions from UHB or localized states to the conduction band:

$$\sigma_b = \sigma_0 + \sigma_{\text{thr}}, \quad \delta\sigma \propto W_c N^- \tau_0 (\mu_0 - \mu_d), \quad (7)$$

where τ_0 , μ_0 , and μ_d are respectively the lifetime of the free carriers and the mobilities in the conduction band and over the delocalized states of the UHB.

If $\sigma_0 \gg \sigma_{\text{thr}}$, then

$$\frac{\delta\sigma}{\sigma_b} \propto \frac{N^- W_c}{N W_b}, \quad \frac{N^-}{N} \propto \frac{\delta\sigma}{\sigma_b} \frac{W_b}{W_c}. \quad (8)$$

3. With further increase of N , when $\sigma_{\text{thr}} > \sigma_0$

$$\frac{\delta\sigma}{\sigma_b} \propto W_c \mu_0 \tau, \quad \sigma \text{ and } \delta\sigma \propto n_L. \quad (9)$$

Relations (6)–(9) make it possible to determine the relative changes of N^- from the experimental dependences of $\delta\sigma/\sigma$ and $\delta\sigma$ on T , E , Φ , and N .

4. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

It was already noted in Ref. 11 that the delocalization of H^- -like states the critical values of the corresponding parameters (the density N_{cr} and the temperature T_{cr}) can be assessed from the qualitative change of the frequency dependence of the PC spectra, viz., the PC peak with the characteristic short-wave descent begins to give way to a monotonic increase of the PC with frequency. We begin with a determination of the values of N_{cr} and T_{cr} .

1. Critical density N_{cr} and temperature T_{cr}

Figures 3a, b show the PC spectra of several samples at $T = 4.2$ K. We note that the σ_2 conduction is observed only in the most strongly doped samples $p-9$ and $p-11$ (see Table I and Fig. 2). It can be seen that the character of the spectrum changes with increasing N . Thus, for n -Si the slope of the short-wave descent decreases at $N = 4.2 \times 10^{17}$ cm $^{-3}$, and for p -Si at $N = 3 \times 10^{17}$ cm $^{-3}$. At larger values of N the PC

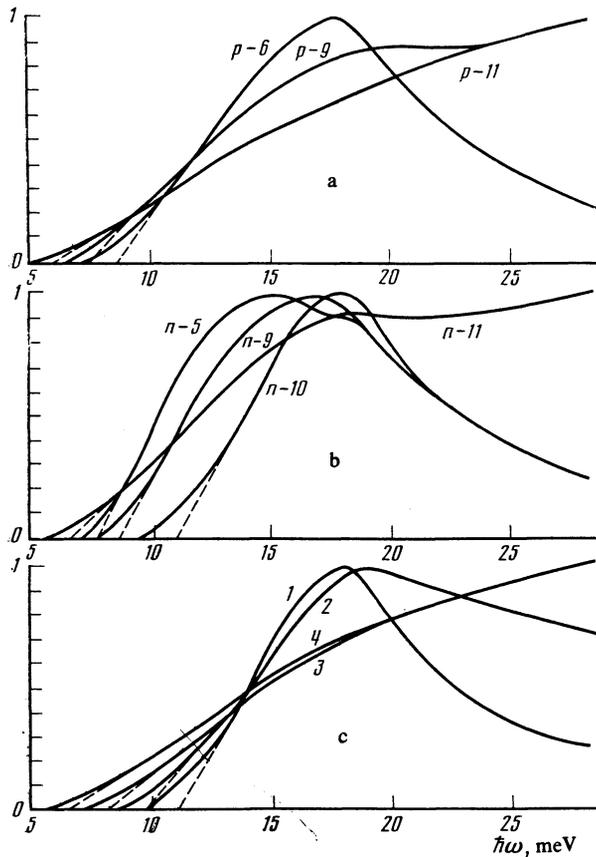


FIG. 3. Change of the form of the PC spectrum with change of the impurity density at $T = 4.2$ K; a) Si:B; b) Si:P; c) with temperature (sample $n-10$) $T = 2$ (1), 4.8 (2), 6 (3), and 8 K (4).

increases monotonically with frequency. We note that similar changes of the PC spectra with increasing N were observed also for Ge:Sb samples ($T = 1.46$ K, $N \gtrsim 2.6 \cdot 10^{16}$ cm^{-3}).²³

Thus, for all the investigated materials, at sufficiently large values of Na_0^3 , a monotonic increase of the PC (and hence of the absorption cross section) with frequency is observed. This increase is typical of transitions between delocalized states. A qualitative change of the form of the spectrum occurs at values of Na_0^3 smaller by approximately an order of magnitude than those at which the static σ_2 conduction is observed.

Let us discuss the possible cause of the change of the frequency dependence of the PC. It was already indicated above that in the case of an optical transition from a localized state to delocalized states the cross section decreases at photon energies $\hbar\omega$ corresponding to the condition $\hbar\omega - E \gg \hbar^2/2ma^2$. Therefore the change of the frequency dependence of the PC (curves 1–3 of Fig. 3a) following an insignificant change of the threshold energies³⁾ E_{thr} offer evidence that with increasing N the initial states cease to be localized and that the spatial modulation of their wave functions is negligible, i.e., three wave functions are of appreciable magnitude in the macroscopic region. Two situations are then possible: the electron is located in a cluster that contains a large number of centers, or the electrons is deloca-

lized, and a strip of states with band conduction was formed. In the latter case the frequency dependence of the PC (Figs. 2a, b) can be explained in the same way as for interband transitions.²⁴ To ascertain which of the possibilities is realized, it is necessary to know either the theoretical expressions for the cross section of the transition from the cluster to the band, or perform additional measurements of the static conductivity (see Sec. 4). A variation of the form of the PC spectrum with N can be assumed. The unexpected result was that a similar spectrum transformation occurs in sufficiently doped samples when the temperature is raised, at $T \gtrsim T_{\text{cr}}$, where T_{cr} is the temperature at which the slope of the short-wave descent decreases by $\sim 10\%$. Figure 3c shows by way of example the PC spectra of sample $n-10$ at different temperatures. We note that with increasing N the values of T_{cr} decrease, and the temperature interval ΔT in which a substantial change takes place in the form of the spectrum increases noticeably. Thus, for sample $n-5$ the temperature is T_{cr} and $\Delta T \approx 0.2$ K, while for sample $n-10$ we have $T_{\text{cr}} \approx 4.5$ K and $\Delta T \approx 1$ K.

The influence of the temperature on the form of the spectrum and the $T_{\text{cr}}(N)$ dependence can be explained by assuming the simultaneous existence of both localized states (of the type $H^- - H^+$ at $N < 0.05N_M$ and more complicated when N is increased) and delocalized ionic states higher in energy than the localized ones, but substantially lower than the bottom of the conduction band (see Fig. 4). Since the energy gap (δE) between the localized states and the delocalized ones (the bottom of the UHB) is less than the distance to the bottom of the conduction band, the thermal spread, which increases with increasing T , takes place mainly in the UHB. The probability of the thermal spike W_T is determined by the value of δE , which can be estimated at $\delta E = \Delta - I$, where $\Delta = e^2/\kappa R_c$ is the shift of the ionic level by the Coulomb potential of the nearest center $R_c = (\frac{4}{3}\pi N)^{-1/3}$ (Ref. 6) (and I is the exchange-interaction energy). The value of I itself depends here, generally speaking on the fluctuation spread Δ_{ij} of the levels of the neighboring centers due to the presence of charged impurities. Neg-

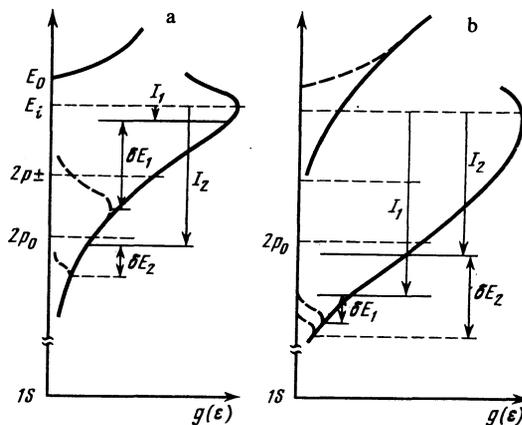


FIG. 4. Dependence of the state density on the energy (schematic) for samples $n-5$ (a) and $n-10$ (b). Variation of the density of the localized states (dashed lines) and of mobility edge with temperature. The values of δE_1 and I_1 correspond to $T = 2$ K; δE_2 , and I_2 correspond to 8 K.

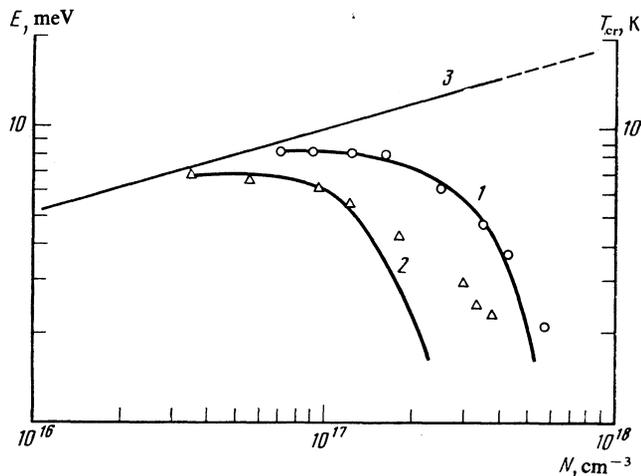


FIG. 5. Dependence of the critical temperature T_{cr} on the impurity density (O)— n -Si, Δ — p -Si and the calculated values of δE for n -Si—1, p -Si—2, Δ —3.

lecting the influence of Δ_{ij} on I , we assume that the dependence of I on N is of the form

$$I(N) \propto \exp(-\alpha/N^{1/2} a_i), \quad \alpha \propto 1.$$

Then, by determining I from the value of $\varepsilon_2(\varepsilon_2 = \varepsilon_1 - E_i - I, \text{Ref. 1})$ obtained from galvanomagnetic measurements for the samples with the smallest densities (n -13 and p -9), we can estimate δE . Figure 5 shows plots of $\delta E(N)$ and $T_{cr}(N)$ for Si:P and Si:B. The $\delta E(N)$ and $T_{cr}(N)$ curves are approximately similar.

The transition of the carriers from localized to delocalized states is thus determined by the ratio $\sigma E/T$. This confirms the assumption advanced above concerning the mechanism whereby the temperature influences the form of the PC spectrum. The calculation of δE must be regarded as

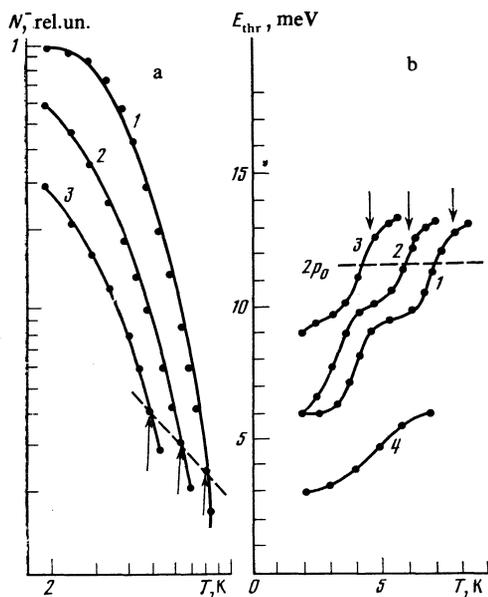


FIG. 6. Dependence of the charged-center density $N^- \approx N^+$ (a) and of the PC threshold energy (b) on the temperature for sample n -5 at different background excitation levels Φ : 1— Φ_0 , 2— $\Phi_0/5$, 3— $\Phi_0/25$. Curve 4 in Fig. 6b is a plot of $E_{thr}(T)$ for sample n -1 at $\Phi = \Phi_0$.

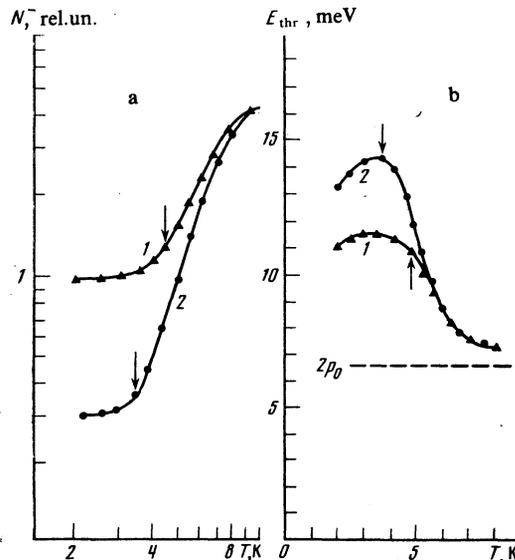


FIG. 7. Dependence of the charged-center density $N^- \approx N^+$ (a) and of the PC threshold energy (b) on the temperature for sample n -10 at $\Phi = \Phi_0(1)$ and $\Phi = \Phi_0/5 (2)$.

approximate, since the energy spread Δ_{ij} of the levels of the neighboring centers, as will be shown below, plays an important role in the location of the delocalized states.

2. Effect of charged-center density

To study the influence of the fluctuation potential on the energy positions of the delocalized states, the measurements were made under various background-excitation conditions, i.e., at different charged-center densities.

Figures 6 and 7 show plots of $N^-(T)$ and $E_{thr}(T)$ at different excitation levels Φ for the samples n -5 and n -10. The values of N^- were determined from relations (8) and (9), whose validity for samples n -5 and n -10 will be demonstrated in subsec. 4. We note in both samples the values of T_{cr} (marked by arrows) decrease with decreasing Φ , thus indicating a decrease of the energy gap δE between the localized and delocalized states. Since the binding energy of an excess electron in a state of the type $H^- - H^+$ is determined only by the density of the main impurity

$$E = E_i + e^2/\kappa R c_s,$$

the indicated decrease of the gap can be due to the fact that the delocalization "encompasses" deeper states because of the decrease of the fluctuation potential. The latter decreases with decrease of the density of the $H^- - H^+$ complexes.

From the fact that sample n -5 has at $T = T_{cr}$ close values of E_{thr} for all background illumination levels (see Fig. 6) it follows that at constant N the locations of the delocalized states are determined by the values of N^- and N^+ , and by the same token by the value of Δ_{ij} .

This is also clearly seen from Fig. 7b: a decrease of N^- leads to a deeper position of the delocalized states relative to the bottom of the conduction band.

We note that with increasing N (at constant Φ) the character of the $N^-(T)$ and $E_{thr}(T)$ dependences changes. At $N < 2 \times 10^{17} \text{ cm}^{-3}$ and $\Phi = \Phi_0$ (samples n -4 to n -8) N^- decreases with increasing T while E_{thr} increases, as is in fact

illustrated in Fig. 6. At $N > 2 \times 10^{17} \text{ cm}^{-3}$ (samples $n-9$ to $n-12$) the $N^-(T)$ and $E_{\text{thr}}(T)$ dependences are reversed (Fig. 7). The difference in the character of the $N^-(T)$ dependences causes δE to decrease with increasing T for samples $n-5$ to $n-8$, and to increase for samples $n-9$ to $n-12$. Therefore the decrease of δE with T leads to a change of the form of the spectrum in a narrow temperature range, while an increase of δE with T , conversely, lengthens the transition of the carriers to the delocalized states.

To explain the differences between the $E_{\text{thr}}(T)$ and $N^-(T)$ dependences ($\Phi = \Phi_0$) in samples with different N it is necessary to consider the processes of recombination of the "excess" and free carriers.

3. Processes of generation and recombination of free and "excess" charge carriers

The ionic states are populated under the experimental conditions by capture of free carriers by neutral centers. Hopping of the excess carrier over the neutral centers towards the attracting center produces complexes of the $\text{H}^- - \text{H}^+$ type. It was shown earlier (see Ref. 6) that at $N < N_{\text{cr}}$ the "excess" carriers are ejected predominantly back into the conduction band even at minimum background-excitation levels. Thus, at $N < N_{\text{cr}}$ the neutral impurities are adhesion centers, and the carrier lifetimes in the conduction band (τ_0) are determined by capture by dipoles of the $\text{H}^- - \text{H}^+$ type, whose relative density at $T \lesssim 2 \text{ K}$ is estimated at $N^-/N = f(N) = 0.05 - 0.15$. The $E_{\text{thr}}(T)$ and $N^-(T)$ dependences in such samples can be explained by starting from the equality $W_b = W_T(E_{\text{thr}})$ (Ref. 6).

The situation changes radically at $N > N_{\text{cr}}$. The $E_{\text{thr}}(T)$ and $N^-(T)$ dependences can no longer be explained by assuming thermal ejection of the electrons from complexes of the type $\text{H}^- - \text{H}^+$ into the conduction band. Thus, for the considered sample $n-5$, the calculated values of $W_T(E_{\text{thr}})$ at $\Phi = \Phi_3 = \Phi_0/25$ differ from the real ones by 10^7 times (cf. curves 4 with 1, 2, and 3 on Fig. 6b).

Since delocalized states are present in samples with $N > N_{\text{cr}}$, it is natural to assume that the electrons are ejected thermally from the $\text{H}^- - \text{H}^+$ complexes not into the conduction band but into the UHB, followed by recombination of an electron from this band with the freed attracting center. In this case the decrease of N^- or N^+ (say, on account of a decrease of Φ) should lead to a lowering of the bottom of the UHB and to an even earlier shift of the long-wave PC limit with increasing T . This is confirmed by the data of Fig. 6b, which shows plots of $E_{\text{thr}}(T)$ and $N^-(T)$ for three background-excitation levels. It can be seen that E_{thr} depends on Φ at constant T and that the difference in the values of E_{thr} at $T < T_{\text{cr}}$ reaches 3–4 meV. So large a change of E_{thr} points directly to an increased probability of thermal ejection from localized states of the $\text{H}^- - \text{H}^+$ type when the background illumination is decreased, i.e., to a change of the final states. These states cannot be the bottom of the conduction band, since $\varepsilon_1 = E_0 = \text{const}$ in the investigated sample. We note that the $\delta\sigma/\sigma(T)$ dependence turns out to be close to exponential

$$\delta\sigma/\sigma(T) \propto \exp(-\Delta E/kT),$$

and the values of ΔE do not exceed 2–3 meV, whereas $E_{\text{thr}} > 10 \text{ meV}$. This confirms once more that the thermal ejection is into states that are substantially lower than the bottom of the conduction band.

From a comparison of the $N^-(T)$ dependences in n -Si samples with different values of N it follows that the lifetime of the localized carriers depends substantially on the number of excited impurity-center levels that lie below the delocalized states. This explains why no delocalized carriers are observed in the PC spectra when the bottom of the UHB lies higher than the level $2p_{\pm}$, and the presence of the delocalized states manifests itself in the dependence of E_{thr} on T (see above). At $\varepsilon_2 > E_0 - E_{2p_0}$ the relative number of delocalized carriers is small ($n_d \ll n_L$), and they do not change the slope of the short-wave descent, but manifest themselves in the PC spectra as a weakly pronounced "pedestal" with $E_{\text{thr}} \gtrsim 6 \text{ meV}$, i.e., at $\varepsilon_2 < E_0 - E_{2p_{\pm}}$. In more strongly doped samples, such that the bottom of the UHB dropped below the last excited state (i.e., $\varepsilon_2 < E_0 - E_{2p_0}$), the density of the delocalized "excess" electrons, on the contrary, is relatively large. The reason is that in this case the capture of electrons from the UHB is possible only into a ground state of the impurity ($1S(A)$, $1S(E)$, $1S(T_1)$ in n -Si). The probability of such a capture is small, since the electron should emit a photon with a wave vector $q \gg a^{-1}$ (see §2). In such samples, the number of "excess" carriers increases with rising temperature, and the bottom of the band rises and approaches the $2p_0$ level; as a result, a transition from the UHB to the $2p_0$ level becomes possible, followed by recombination, and this hinders the increase of N^+ or N^- . The bottom of the band is therefore stabilized near the $2p_0$ level (see Figs. 7a, b). A similar role is played in Si:B by the excited states $1\Gamma_8^-$ ($E_{\text{thr}} \approx 14.5 \text{ meV}$) and $2\Gamma_8^-$ ($E_{\text{thr}} \approx 10.5 \text{ meV}$) (see Fig. 2b).

Figure 2 shows the value of $\varepsilon_{2\text{opt}}$ determined by an optical method ($\varepsilon_{2\text{opt}} = \varepsilon_1 - E_{\text{thr}}$) and the range of variation of $\varepsilon_{2\text{opt}}$ with changing N^- and N^+ . It can be seen from Figs. 2a, b that the UHB is formed at much lower impurity densities than those at which the static σ_2 conduction is observed, and the values of $\varepsilon_{2\text{opt}}$ near the delocalization threshold are not much smaller than $E_0 - E_i$. The experimental results allow us therefore to assume that the UHB is made up of small-shift H^- like states.

It was shown above that the trapped "excess" carriers can move rapidly over the delocalized states, reach the attracting center and recombine with it. Thus, at $N > N_{\text{cr}}$ the neutral centers cease to be adhesion centers and form a new indirect recombination channel, wherein the stationary carrier lifetimes⁴⁾ τ_0 can be shorter than the energy relaxation time,²⁰ $\tau_0 < \tau_e$. Since $\tau_0 \gtrsim \tau_e$ for cascade capture,¹⁸ the turning on of a new recombination mechanism should lead to a decrease of the static conductivity $\sigma_0 = \sigma_b - \sigma_{\text{thr}}$. Figure 8a shows the $\sigma_b(N)$ dependence for n -Si and various T (the $\sigma_b(N)$ dependence for p -Si is similar). The values of σ_b are normalized to depth of penetration of the radiation into the sample ($d \propto N^{-1}$). It can be seen that with increasing N , at $T = 2 \text{ K}$ and $N > N_{\text{cr}}$, the value of σ_b decreases abruptly, followed by a slow rise. We attribute the decrease of σ_b to the

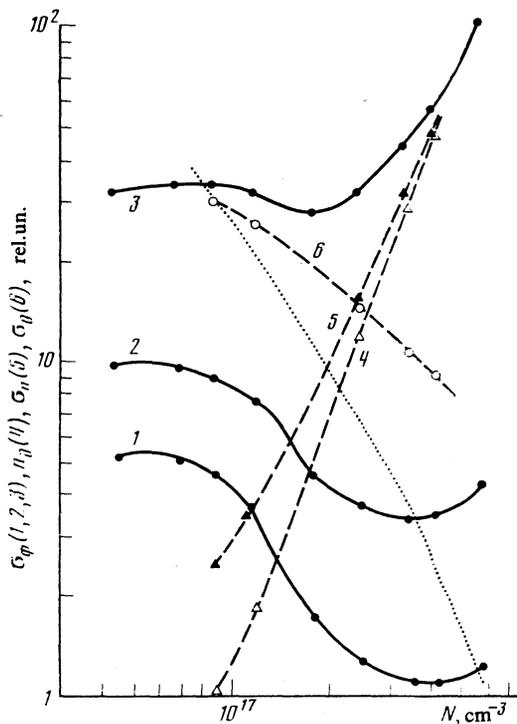


FIG. 8. Dependence of σ_b on N at $T = 2$ —(1), 4.2—(2), 8K (3). Dependences of n_d (4), σ_{thr} (5), and σ_0 (6) on N at $T = 8$ K.

turning on of the new carrier recombination mechanism; the subsequent rise, which becomes stronger with increasing T , will be shown below to be connected with the contribution made to the conductivity by carriers in delocalized states.

4. Conduction by carriers over delocalized states

As already noted, delocalized carriers appear at $N > N_{\text{cr}}$ and $T > T_{\text{cr}}$ in the PC spectra, i.e., their density (n_c) becomes comparable with n_L , the latter being estimated at $(5-15\%)N$ at $N < N_{\text{cr}}$ and $T \lesssim 2$ K.⁶ Therefore, notwithstanding their much smaller (compared with the band) mobility ($\mu_d/\mu_0 \ll 1$), they should obviously participate in the static conduction. It can be seen from Fig. 8 that at $T > T_{\text{cr}}$ the value of σ_b increases with increasing N and T . To cast light on the conduction mechanism, we measured the temperature dependence of the magnetoresistance $\Delta\rho/\rho$ of Si:P samples with $N > N_{\text{cr}}$ and $N < N_{\text{cr}}$ (see Fig. 9). It can be seen that at $N < N_{\text{cr}}$ (curve 1), when $\sigma_b = \sigma_0$, the ratio $\Delta\rho/\rho$ does not depend on T and Φ . At $N > N_{\text{cr}}$ the value of $\Delta\rho/\rho$ decreases with increasing T , and the start of the decrease varies with Φ and is close to T_{cr} . With increasing N and at $T \approx 10$ K the values of $\Delta\rho/\rho$ become even negative, and this is a distinguishing feature of static δ_2 conduction (conduction over the UHB, Ref. 1) i.e., σ_{thr} is an impurity conductivity of the band type. At lower values of N we have $\Delta\rho/\rho > 0$ and the mechanism of band conduction via the excess carriers is not so obvious. It can be assumed that at $\Delta\rho/\rho > 0$ the excess carriers are in a cluster and move through the crystal by hopping between clusters; in addition, photoexcited hopping conduction over the percolation level of the ground state can be produced following thermal breakup of the $\text{H}^- - \text{H}^+$ complex (i.e., at $T \gtrsim T_{\text{cr}}$). According to estimates, the value of

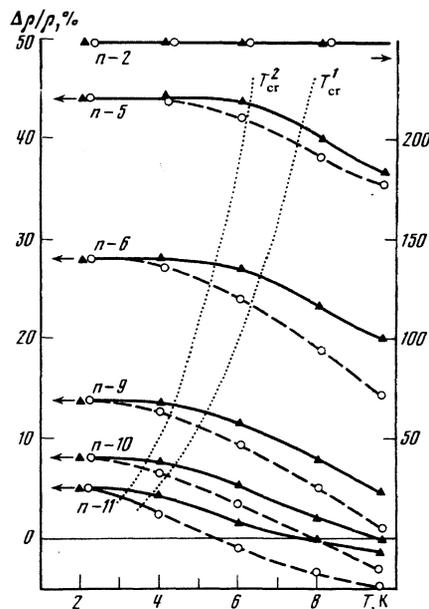


FIG. 9. Temperature dependences of the magnetoresistance of n -Si samples at $H = 45$ kOe and at Φ_0 (\blacktriangle) and $\Phi_0/5$ (\circ). The dashed line separates the region of observation of delocalized carriers in the PC spectra.

$\Delta\rho/\rho$ for these mechanisms should be less than that observed in experiment at $T = 2$ K, while σ_{thr} depends exponentially on N . We calculate the contributions of σ_{thr} and σ_0 to σ_b at $T = 8$ K, using the data of Figs. 8 and 9 and assuming that σ_{thr} is a band-type conductivity and is characterized by a negative magnetoresistance; for the sake of argument we assume that $(-\Delta\rho/\rho) = 5\%$ (see Fig. 8, curve 5). We estimate next the values of n_d under these conditions from the experimental values of $\delta\sigma/\sigma$ and $\delta\sigma$ (see Fig. 8, curve 4). Comparison of curves 4 and 5 leads to the conclusion that μ_d ($\mu_d \propto \sigma_{\text{thr}}/n_d$) does not increase exponentially with N , as should be the case for hopping conduction, but even decreases somewhat.

The experimental results allow us therefore to assume that at $N > N_{\text{cr}}$ the delocalization encompasses the entire crystal, from contact to contact, and an impurity band of the UHB is formed in Si:P at $N \approx (7-9) \times 10^{16} \text{ cm}^{-3}$ (i.e., at N smaller by an order of magnitude than observed in galvanomagnetic measurements).

5. UHB and the excited states of impurities

We have considered above (in subsecs. 1, 2, and 4) the overlap of the ionic state, the critical density N_{cr} , the factors that determine N_{cr} , and the conduction by the carriers, i.e., questions that are traditional in the study of delocalization.¹ In addition, we discussed a question peculiar to optical experiments, that of recombination of "excess" electrons; it was assumed that phonon transitions take place between the UHB and the excited states. In this section we shall dwell on a less studied problem, the interaction of excited states (ES) of impurity centers with ion states located in the same energy band, and in particular with delocalized states. At impurity densities lower than the delocalization threshold ($N < N_{\text{cr}}$), we noted manifestations of pseudocrossing of the ionic and

homeopolar terms of the impurity molecules.⁶ Now we shall discuss the interaction between ES and UHB, and in particular the degree of delocalization of the ES, at impurity densities $N_{cr} \lesssim N \lesssim 0.05N_M$, i.e., when the absorption lines corresponding to transitions from the ground to the excited states still have a resonant character.

The interaction of ES with ionic states was invoked only to explain the observed abrupt absorption line broadening with increasing N .^{21,25,26} Thus, in Ref. 26 this line broadening in n -Ge was analyzed from the viewpoint of a theory considered in Ref. 27 for transitions from ground states into slightly overlapping ionic states of neutral donors, as well as into a "mixture" of excited and ionic states of neighboring neutral centers.⁵⁾

In our opinion the question of the nature of the states that are final for an optical transition, and of the interaction between excited and ionic states, can be resolved by investigating the photoconductivity that occurs at the corresponding radiation frequency, and of the effect exerted on the PC by the magnetic field. To this end, the PC and absorption were measured in Si:P samples with $\epsilon_1 \approx E_0 (n-1 - n-8)$, see Table I). Figure 10a shows by way of illustration the absorption spectrum at frequencies corresponding to the $1S(A) \rightarrow 2p_{\pm}$ transition for sample $n-6$ at $T \approx 5$ K. We note that the absorption spectrum is of a form typical of intracenter transitions, but the linewidth is noticeably larger than in the case of isolated donors (see, e.g., Ref. 21). Thus, the absorption is determined mainly by optical transitions into excited impurity states, while the contribution of the nonresonant absorption due to transitions to the "tail" of the state density of the conduction band and directly to the UHB is small.

Figure 10b shows the PC spectrum under the same conditions. We note that the abrupt increase of the PC in the absorption band of the $2p_{\pm}$ band is observed at tempera-

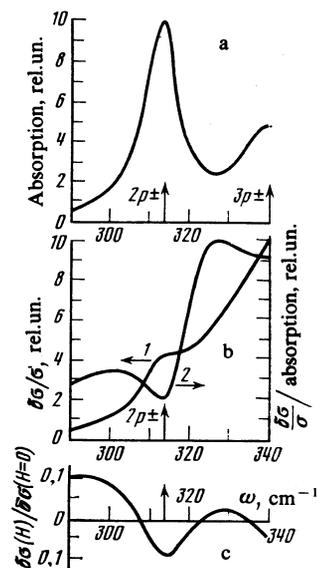


FIG. 10. Spectral dependences of the absorption (a), of the PC (b, 1), of the PC normalized to the absorption (b, 2), and of the change of $\delta\sigma$ (c) in a magnetic field $H = 10$ kOe. Sample $n-6$, $T = 5$ K, $\Phi = \Phi_0$, sample thickness $120 \mu\text{m}$.

tures at which the bottom of the UHB drops below the $2p_{\pm}$ level (for sample $n-6$ at $T \gtrsim 3.5$ K). Since the usual photothermal ionization of the impurities is observed at considerably higher temperatures ($T > 15$ K), it can be stated that the PC is effected over states located lower than the bottom of the conduction band. The influence of the magnetic field H ($H \approx 10$ kOe) leads under these conditions to an increase of $\delta\sigma$ at frequencies far from resonance (see Fig. 10c)—a negative magnetoresistance typical of conduction over the UHB is observed. Near resonance, on the contrary, $\delta\sigma$ decreases somewhat. We note that the PC referred to the absorption (Fig. 1b, curve 2) has a minimum at $\hbar\omega = E_{1S(A)} - E_{2p_{\pm}}$. With increasing N this ratio increases sharply, and negative magnetoresistance sets in also at a resonant frequency.

It appears that the absence of negative magnetoresistance near resonance (see Fig. 10c) is evidence that the electrons land first in the ES, and then go over to the UHB. In this case the contribution of the excited electrons to the conductivity is proportional to the ratio W_1/W_2 , where W_1 and W_2 are the probabilities of departure to the UHB and to the ground state, respectively. The mechanism of such a transition calls for further refinement, but the decrease of its probability (W_1) with increasing H allows us to assume that it is of the hopping type. Therefore at small W_1 the ES should be regarded as a localized despite the fact that they land in the UHB energy band. Since W_1 and the width of the absorption line are determined by the same interaction of the excited and ionic states, it follows that W_1 should increase sharply with increasing N .⁶⁾ Therefore when W_1/W_2 is increased the excited states should manifest themselves not only in recombination but also in conduction. The degree of delocalization of the excited state is apparently characterized by the absorption-line width and can be determined by investigating the effect of the magnetic field on the PC.

Under equilibrium conditions, when the bottom of the UHB drops to the ES with increasing N , a jumplike increase of the density of states takes place, and consequently of the carrier density near the UHB bottom. The contribution of these "additional" carriers to the UHB conduction is determined by the degree of delocalization of the ES. The participation of the ES in the formation of a conducting impurity band is confirmed by the following experimental fact: the maximum value of ϵ_2 determined from galvanomagnetic measurements is always somewhat smaller than the energy gap between the ground and nearest excited state or split-off ground state (see Table II).⁷⁾

5. CONCLUSION

The foregoing experimental results show that in a disordered system of neutral impurity centers the appearance of impurity conduction of the band type is due to delocalization of H^- -like states, which takes place at impurity densities smaller by almost two orders than N_M . The energy gap between the ground and the delocalized states (the Mott-Hubbard gap) is determined not only by the quantity N , but also depends strongly on the charged-center density. In a wide range of impurity densities there exist simultaneously localized and delocalized states, and their relative popula-

TABLE II.

Material	Si : P	Si : B	Ge : Sb	Ge : As
States	$1S(T_1)$	A	$2\sigma_0$	$1S(T)$
Energy gap meV	11.6	23	4.75	4.2
ε_2 , meV	11	22	4.6	3.9

tion varies with temperature. When the energy positions of the UHB and of the excited impurity states coincide the degree of localization of the latter decreases steeply with increasing N .

The lifetimes of the delocalized carriers depend on the mutual energy positions of the excited states of the impurity and the UHB. Delocalization of H^- -like states leads to formation of a channel for rapid transport of the captured "excess" carrier to an attracting center.

¹At low compensation ($K \rightarrow 0$) the energy ε_2 is half the Mott-Hubbard gap $(\Delta E)_{MH}$.¹

²The frequently cited Ref. 14 contains in our opinion a number of inaccuracies. Thus, the Hamiltonian for the electrons in the D^- band was constructed assuming $Na^3 \ll 1$, but ε_2 was calculated assuming $Na^3 \gg 1$; no account was taken (in the calculation of the downward shift of the D^- band) of the polarization potential of the neutral center.

³The values of E_{thr} (the PC threshold energy) were determined from the emission-energy quanta corresponding to cutoff of the PC.

⁴In the situation considered, τ_0 can differ substantially from the photoconductivity relaxation time.

⁵It is noted in Ref. 27 that what is studied is an optical transition from the ground state and excited states to localized ones. From the calculations in that reference, however, it can be seen that final state for the optical transition is taken to be an aggregate of weakly overlapping ionic states.

⁶The role of the exchange interaction between the excited and ionic states is apparently decisive in the absorption-line broadening: our reduction of the results of Ref. 25 shows that the line widths increase with N exponentially ($\propto \exp(-\alpha N^{1/3}/a)$).

⁷The interaction between delocalized ionic states and excited states, which leads to delocalization of the latter, is apparently the decisive factor in the formation of UHB in materials with $K > 0.1$.

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Translated by J. G. Adashko.