Model of high-frequency hopping photoconductivity of doped silicon

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A phenomenological model of high-frequency hopping photoconductivity along excited metastable states of an impurity in a semiconductor is developed. Polarization conductivity arises as a result of changes occurring in the dipole moments of pairs of ions of the majority and compensating impurities due to electronic transitions between ionized and excited atoms of the majority impurity in a high-frequency electric field. It is shown that complexes, including excited and ionized atoms of the majority impurity and an ion of the compensating impurity, contribute to the high-frequency hopping conductivity. Such complexes are formed as a result of the capture of photocarriers on dipoles and drift of ions of the majority impurity in the electric field of the ions of the compensating impurity. Simple expressions are obtained for the high-frequency hopping photoconductivity as a function of the excitation intensity, the concentrations of the majority and compensating impurities, and the temperature. Estimates are presented of the absolute magnitude of the high-frequency photoconductivity. The model satisfactorily describes the experimentally established laws of extrinsic photoconductivity of silicon, doped with group III and V impurities, with a microwave bias voltage.

1. INTRODUCTION

In Refs. 1–4 it was established that the kinetics of extrinsic photoconductivity of silicon, doped with a number of group-III and -V elements in concentrations above 10^{16} cm^{-3}, under a microwave (37 GHz) bias voltage is determined by two relaxation times—a short relaxation time τ_r (10^{-8}–10^{-10} sec), equal to the lifetime of free photocarriers under a constant bias voltage, and a long relaxation time $\tau^* \approx 10^{-3}$ sec. Slow relaxation is observed at temperatures $T \leqslant 15$ K. When the exciting radiation has a modulation frequency $f \ll 1/\tau^*$ the high-frequency photoresponse can be almost three orders of magnitude higher at 5 K than the photoresponse with a constant bias voltage. Slow relaxation of extrinsic absorption with time constants close to τ^* was observed in the same samples of doped silicon.[3]

The observed phenomena were explained by the existence of metastable excited states of the impurities. The long lifetime of these states causes charge carriers trapped by impurity ions in the process of recombination to accumulate on them. In the present paper we propose a phenomenological model of hopping photoconductivity in which metastable states participate. This model explains the basic laws established by experimental investigations of high-frequency extrinsic photoresponse of doped silicon.[1–4]

Silicon samples doped with boron in concentrations $N = 10^{15}$–$4 \cdot 10^{16}$ cm^{-3} and compensated with phosphorus in concentrations $N_+ = 10^{12}$–10^{14} cm^{-3}, were studied in greatest detail in Ref. 4. For this reason, we consider a model for the case of a hole semiconductor. In the case of impurity photoexcitation the holes are captured by negatively charged ions of the majority impurity in high excited states,[5] and by emitting and absorbing phonons they relax into a metastable state from which optical transitions into the ground state are forbidden. In the case of boron this can be the $1S(P_{1/2})$ state with an ionization energy of 23 meV.[6] This state is formed from wave functions of the branch of the valence band that is split off by the spin-orbital interaction. Under quasisteady excitation conditions the ratio of the concentration of free nonequilibrium holes to the concentration N_* of localized nonequilibrium holes should be equal to τ_r/τ^*, i.e., it should reach 5–7 orders of magnitude for the silicon samples investigated. This difference is apparently insufficient for hopping photoconductivity to predominate over photohole photoconductivity under a constant bias voltage. When a high-frequency bias is voltage is applied, however, polarization conductivity should appear.[7] Polarization conductivity appears as a result of a change in the dipole moments of the pairs of ions of the majority and compensating impurities in an electric field. In order to magnitude the time constant of this change is equal to the hopping time τ between ionized and neutral atoms of the majority impurity, and the total change in the polarization of the semiconductor in an alternating electric field with frequency ω is manifested as flow of an alternating current. In this case the dark ac conductivity is many orders of magnitude greater than the dc conductivity.[7] In the case of extrinsic photoexcitation the excited metastable states of atoms of the majority impurity with large Bohr radii a^* are included in the process, and this should strongly increase the polarization hopping conductivity.

2. PHENOMENOLOGICAL MODEL

Holes accumulate in metastable states as a result of being captured by both isolated ions with concentration N_- and dipoles with concentration N_{+-} (Ref. 5). In the case of capture by dipoles the excited atoms are localized near ions of the compensating impurity at a characteristic distance $r_{\rm av} = (4\pi N/3)^{-1/3}$, forming positively charged pairs with concentrations N_{+*}. When the frequency $\omega = 2.3 \cdot 10^{11}$ sec^{-1} of the ac electric field is high (Refs. 1–4), only hops between close ionized and excited atoms can contribute to the hopping conductivity $\sigma(\omega)$. Since both photoionization and capture are random processes, the probability of formation of such close pairs is small if the degree of excitation is low. However, the electrostatic attraction between isolated ions N_- and pairs N_{+*} should draw them together and re-

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sult in the formation of complexes in concentrations N_{+*-}, consisting of excited and ionized atoms of the majority impurity and an ion of the compensating impurity. The average distance between all partners in such complexes will also be close to r_{av}, determined by the concentration of the majority impurity N. It is these complexes that should make the main contribution to the polarization hopping conductivity.

The fraction of excited atoms localized near ions of the compensating impurity is determined by the distribution of the recombination fluxes of holes between ions and dipoles, and is equal to

$$C = \frac{\alpha_{+-}N_{+-}}{\alpha_{+-}N_{+-} + \alpha_{-}N_{-}}, \qquad (1)$$

where α_- and α_{+-} are the hole capture coefficients of ions and dipoles, respectively. Note that N_- is the total concentration of isolated ions of the majority impurity, which appear with thermal and optical excitation, including also holes captured by neutral acceptors with consequent formation of A^+ centers.[8] If the nearest negatively charged ion of the majority impurity is located at a distance R when N_{+*} pair is created, then it is drawn to the pair over the drift time

$$t_d = \frac{\varepsilon R^3}{3e\mu_h}, \qquad (2)$$

where e is the electron charge, ε is the dielectric permittivity, and μ_h is the hopping "mobility." The number of such ions in a spherical shell $(R, R + dR)$ is

$$dN_- = N_- \cdot 4\pi R^2 \exp(-R/R_{\text{av}})^3 dR, \qquad (3)$$

where $R_c = (\frac{4}{3}\cdot\pi\cdot N_-)^{-1/3}$. Since the excitation relaxes as $\exp(-t/\tau^*)$, complexes are formed efficiently only if $t_d < \tau^*$. For this reason the concentration dN_{+*-} of complexes formed as a result of drift of ions N_- to pairs N_{+*} from a distance R is

$$dN_{+*-} = N_* C \exp(-t_d/\tau^*) dN_- . \qquad (4)$$

In the case of steady excitation with rate G we have $N_* = G\tau^*$. Integrating Eq. (4) over all values of R, substituting Eqs. (1)–(3), we obtain

$$N_{+*-} = CG\tau^*(1 + \tau_M/\tau^*)^{-1}, \qquad (5)$$

where $\tau_M = \varepsilon/4\pi\sigma_h$ is the Maxwellian relaxation time of the charge due to hopping conductivity $\sigma_h = e\mu_h N_-$.

In the case of impulsive excitation with duration Δt, when $\tau_r \ll \Delta t \ll \tau^*$ only ions N_- which were able to traverse a distance $R_d = (3e\mu_h t/\varepsilon)^{1/3}$ over the time t_d will participate in the photoconductivity. Hence

$$N_{+*-} = CG\Delta t \exp\left(-\frac{t}{\tau^*}\right)\int_0^{R_d} 4\pi R^2 \exp\left(-\frac{R}{R_{\text{av}}}\right)^3 dR \qquad (6)$$

or

$$N_{+*-} = CG\Delta t \exp(-t/\tau^*)[1 - \exp(-t/\tau_M)] . \qquad (7)$$

The conductivity $\sigma(\omega)$ can be determined using the expressions obtained in Ref. 7 for polarization hopping conductivity, replacing the dipole concentration in them, which is equal to the concentration N_+ of the compensating impurity, by the concentration N_{+*-} of complexes from Eq. (5) or (7). Then according to Ref. 7, in an electric field with frequency ω the contribution to $\sigma(\omega)$ of electronic transitions between excited and ionized atoms of the majority impurity, which are located at a distance r from one another, in which the energy changes by ΔE in a hop, is

$$d\sigma(r, \omega, \Delta E) = \frac{N_{+*-}e^2r^2}{12kT\tau}\left(\frac{\omega^2\tau^2 + i\omega\tau}{1 + \omega^2\tau^2}\right)\frac{dN}{\text{ch}^2(\Delta E/2kT)}, \qquad (8)$$

where $dN = 4\pi\cdot Nr^2dr$, and the hopping time is determined by the relation

$$\tau \approx 5\cdot 10^{-13}\left(\frac{r}{a^*}\right)^{-3/2}\exp\left(\frac{2r}{a^*}\right)\text{th}\left(\frac{\Delta E}{2kT}\right). \qquad (9)$$

At the frequency $\omega = 2.3\cdot 10^{11}$ sec^{-1} the condition $\omega\tau > 1$ is satisfied essentially up to $r = a^*$. For this reason, in Eq. (8) the real part of the photoconductivity is much larger than the imaginary part, and the frequency dependence $\sigma(\omega)$ can be neglected. However, the change ΔE in the energy of a dipole accompanying a hop can be significant, since the hopping length is of the same order of magnitude as the distance $r_{\text{av}} = (4\pi N/3)^{-1/3}$ between an ion of the compensating impurity and the atom of the majority impurity, and $\Delta E/kT$ cannot be considered small at low temperatures. Including in Eqs. (8) the functional dependence of ΔE on the hopping length and the direction of a hop with respect to the arm of the dipole is an unwarranted complication of the problem. For this reason, we employ the following rough approximation to estimate the absolute value of $\sigma(\omega)$.

We assume that both the average distance between the ions of the majority and compensating impurities and the averaging hopping length are close to r_{av}. Then only hops with $\Delta E \leqslant kT$ occur. The fraction of hops in directions for which this condition is satisfied is approximately kT/E_c, where $E_{\text{av}} = e^2/\varepsilon r_{\text{av}}$. Then in expressions (8) and (9) we can set $\cosh^2(\Delta/2kT) \times \tanh(\Delta E/2kT) = 0.5\cdot\sinh(\Delta E/kT) < 1/2$, and the expression (8) becomes

$$d\sigma(\omega, r) = \frac{N_{+*-}e^2r^2(r/a^*)^{3/2}\exp(-2r/a^*)dN}{5\cdot 10^{-13}\cdot 6E_{\text{av}}}, \qquad (10)$$

which, after integrating over r, gives

$$\sigma(\omega) = \frac{N_{+*-}}{2r_{\text{av}}^2}\frac{\varepsilon}{5\cdot 10^{-13}}\left(\frac{a^*}{2}\right)^5\frac{\Gamma(13/2)}{2^{13/2}}. \qquad (11)$$

3. COMPARISON WITH EXPERIMENT

According to expression (11), the dependence of the quasistationary polarization photoconductivity $\sigma(\omega)$ on the degree of excitation and the concentrations of the majority and compensating impurities is determined by the concentration N_{+*-} of the complexes, given by (5).

In Ref. 4 it was established experimentally that, in agreement with Eq. (5), the quasistationary photoresponse in a microwave electric field $U_{\text{LF}} \propto \sigma(\omega)$ is a linear function of the excitation rate G. Such a linear relation should hold only for $N_* < N_+$. When, however, an excited and an ionized atom of the majority impurity are present near each ion of the compensating impurity, further increase of excitation should not cause $\sigma(\omega)$ to increase. Indeed, in Ref. 4 the photoresponse was observed to saturate for $G\tau^* \geqslant N_+$.

The dependence of $U_{\rm LF}$ on the concentration N_+ of the compensating donors is determined by the parameter C given by Eq. (1). This parameter describes the distribution of recombination fluxes between dipoles and isolated ions of the majority impurity. As shown in Ref. 5 and 9, at low temperature under conditions of thermodynamic equilibrium with a fixed concentration of the majority impurity the dipole concentration N_{+-} decreases as the concentration N_+ of the compensating impurity decreases. Using the results of Refs. 5 and 9, we calculated for the concentrations $N = 3.5\cdot 10^{16}$ cm^{-3} and $N_+ = 2\cdot 10^{12}$–10^{14} cm^{-3} and $T = 5$ K the parameter C for different values of the ratio α_-/α_{+-}.

Figure 1 displays the parameter C calculated in this manner as a function of N_+, the dependence being extremely strong $[\alpha_-/\alpha_{+-} = 60]$ for the indicated range of concentrations of the compensating donors. The figure also displays the experimental dependence[4] of the photoresponse $U_{\rm LF}$ on the concentration N_+. When the boron concentration is $N = 3.5\cdot 10^{16}$ cm^{-3}. It is evident from the figure that we have $U_{\rm LF} \propto N_+$, although the calculation for thermodynamically equilibrium conditions gives a much weaker dependence. This discrepancy is due to the strong departure from equilibrium under the conditions of the experiment, thanks to which N_{+-} and N_- are determined not only by N, N_+, and the temperature, but also by the background illumination, which was neglected in Ref. 5. This leads, in particular, to the formation of A$^+$ centers[8] and an increase in the concentration N_- of the charge compensating them. For this reason, in order to determine the parameter C we employ the experimental $U_{\rm HF}$ dependence of the microwave photoresponse at high modulation frequency of the exciting radiation $1/\tau^* \leqslant 2\pi f < 1/\tau_r$. We take into account the fact that $U_{\rm HF} \propto \sigma(0)$, i.e., it is proportional to the quasistationary photoconductivity with constant bias voltage.[1,2,4] In comparatively weakly compensated samples we have $N_+ \ll N$, and the mobility μ_p of free holes is determined by scattering by neutral impurities. For this reason $U_{\rm HF} \propto [\alpha_- N_- + \alpha_{+-} N_{+-}]^{-1}$ holds. At high temperatures $T \gg 25$ K the hole recombination flux on dipoles can be neglected and $U_{\rm HF} \propto [\alpha_- N_-]^{-1}$. For a sample with minimum concentration $N_+ = 2\cdot 10^{12}$ cm^{-3} the rate of capture on dipoles, $\alpha_{+-} N_{+-}$, calculated for the case of thermodynamic equilibrium according to Refs. 5 and 9, is negligibly small compared with $\alpha_- N_-$ at all temperatures down to 5 K, i.e., for this "reference" sample $U^0_{\rm HF} \propto [\alpha_- N_-]^{-1} = [\alpha_- N_+]^{-1}$. Then the discrepancy appearing between the dependences $U_{\rm HF}$ and $U^0_{\rm HF}$ as the temperature decreases is caused by the fact that dipoles with concentration N_{+-} in the samples with high compensation are engaged in the capture process. Matching the plots of $U_{\rm HF}$ for each sample with the reference plot $U^0_{\rm HF}$ at high temperatures (25–30 K), we can determine the fraction of the recombination flux on dipoles directly from Fig. 2 at any temperature

$$C = \frac{\alpha_{+-}/\alpha_-}{1 - \alpha_{+-}/\alpha_-} \left(\frac{U_{\rm HF}}{U^0_{\rm HF}} - 1 \right). \qquad (12)$$

Figure 1 displays the parameter C determined as a function of N_+ at 5 K by means of the procedure described above. It is evident that this relation is nearly linear, i.e., it agrees with the relation established in Ref. 4. We note that the temperature dependence of $U^0_{\rm HF}$ differs insignificantly from the temperature dependence of τ_r calculated in Ref. 5 neglecting the formation of dipoles. As the temperature increases, the dipoles dissociate, and this results in a decrease of $U_{\rm LF}$. The temperature dependences of $U_{\rm LF}$ in Fig. 2 are plotted so that for different samples they coincide at high temperatures. It is evident that the hopping photoconductivity $U_{\rm LF}$ starts to exceed the photoconductivity due to free holes $U_{\rm HF}$ precisely at the temperatures when capture of holes on dipoles starts to play a significant role, i.e., when the temperature dependence of $U_{\rm HF}$ and of $U^0_{\rm HF}$ disagree. This confirms the initial assumption that the microwave hopping photoconductivity is connected with capture of photoholes on dipoles. At high concentrations of the majority and compensating impurities capture on dipoles predominates up to 15 K, and as the temperature increases further the decrease of $U_{\rm LF}$ is determined by thermal ionization of the metastable state with activation energy close to 23 meV (Ref. 4).

In Ref. 4 it was established that at low temperatures the

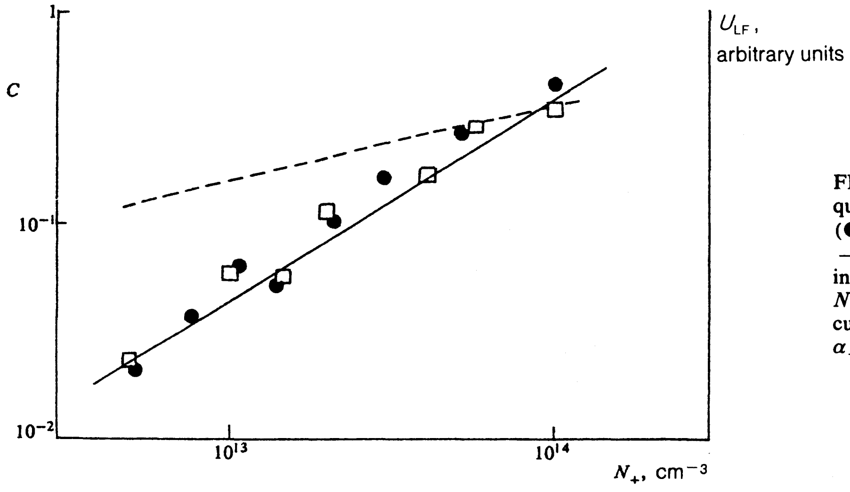

FIG. 1. Relative photoresponse at low modulation frequency $U_{\rm LF}$, of the exciting radiation, according to Ref. 4 (●), and relative value of the parameter $C = U_{\rm HF}/U^0_{\rm HF} - 1$ (□), as functions of the concentration of compensating donors N_+ in samples with the boron concentration $N = 3.5\cdot 10^{16}$ cm^{-3} at 5 K; the dashed line represents the curve of C calculated according to Refs. 5 and 9 for $\alpha_-/\alpha_{+-} = 60$.

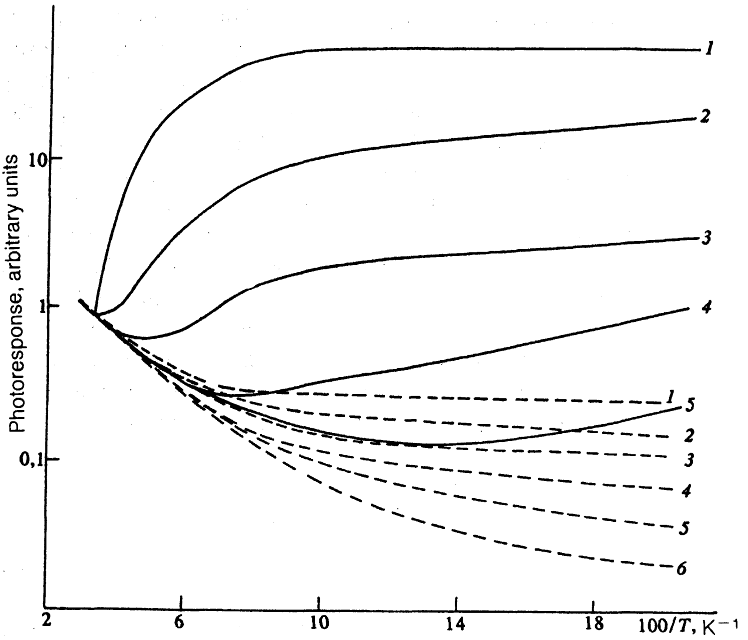

FIG. 2. Temperature dependence of the relative photoresponse with low $U_{\rm LF}$ (solid lines) and with high $U_{\rm HF}$ (dashed lines) modulation frequency of the exciting radiation for samples with boron concentration $N = 3.5\cdot10^{16}$ cm^{-3} and concentration of compensating donors (cm^{-3})

photoresponse $U_{\rm LF}$ with fixed concentration of N_+ of compensating donors ($N_+ = 10^{14}$ cm^{-3}) is virtually independent of the concentration of the majority impurity (boron) for $N \geqslant 2.7\cdot10^{16}$ cm^{-3}, but at lower concentrations

$$U_{\rm LF} \propto \exp(-r_{\rm av}/r_0), \tag{13}$$

where the optimal value is $r_0 = 1.7\cdot10^{-7}$ cm. According to the expression (5), the weak dependence of $\sigma(\omega)$ on N should be observed until $\varepsilon/4\pi\sigma_h = \tau_M < \tau^*$. At $\tau^* = 5\cdot10^{-4}$ sec this corresponds to a hopping conductivity $\sigma_h = 2\cdot10^{-8}$ $(\Omega\cdot\text{cm})^{-1}$ in a constant electric field. In the experiment of Ref. 4 the samples were illuminated with constant background illumination, which determined their conductivity. The free holes and the hopping mechanism made comparable contributions of the conductivity. At 5 K the conductivity of silicon samples with $N = (2.7\text{–}4)\cdot10^{16}$ cm^{-3} was at least 10^{-7} $(\Omega\cdot\text{cm})^{-1}$, i.e., it exceeded the indicated critical value. The exponential drop in $U_{\rm LF}$ for $N < 2.7\cdot10^{16}$ cm^{-3}, which we attribute to the exponential decrease of σ_h, should satisfy the relation $\sigma_h \propto \exp(-2.78 r_{\rm av}/a)$ established for the hopping conductivity,[10] where $a = 2.2\cdot10^{-7}$ cm is the Bohr radius of the ground state of the acceptor. For $r_0 = 1.7\cdot10^{-7}$ cm, however, we must set $a = 4.7\cdot10^{-7}$ cm in Eq. (13), which is approximately twice the Bohr radius. This discrepancy could arise for the following reasons. First, the coefficient 2.78 in the expression for σ_h was obtained in Ref. 10 in a calculation of charge transfer along an infinitely long chain of impurities (an infinite cluster). In our model the N_- ions drift toward the positively charged N_+ donors through atoms of the majority impurity, which form clusters of finite size less than $N_+^{1/3} \lesssim 10^{-4}$ cm. The conductivity of some clusters is significantly higher than the average equilibrium conductivity σ_h. For this reason, averaging over an ensemble of independent clusters can weaken the exponential dependence of σ_h on $r_{\rm av}$. Second, we must include the fact that the ions N_- drift in the electric field produced by the ions N_+, which for $R \leqslant 10^{-5}$ cm is greater than 100 V/cm and is not weak.[11] For this reason, the drift velocity may be higher than expected, which will also weaken the concentration dependence of $U_{\rm LF}$. On the basis of these remarks, it should be noted that both the transition to the exponential dependence of $U_{\rm LF}$ with $\tau_M \approx \tau^*$ and the value of r_0 in Eq. (13), in our opinion, support the proposed model.

We now estimate the absolute magnitude of the low-frequency hopping photoconductivity for a strongly doped sample with boron concentration $N = 4\cdot10^{16}$ cm^{-3} and $N_+ = 10^{14}$ cm^{-3} at 5 K, setting $C = 1$, $\varepsilon = 12$, $\tau^* = 5\cdot10^{-4}$ sec, and $a^* \approx 2a \approx 4\cdot10^{-7}$ cm. Substituting these values into Eq. (5), we obtain $U_{\rm LF} \propto \sigma(\omega) \approx 2\cdot10^{-24} G(\Omega\cdot\text{cm}^{-1})$. For photoconductivity due to free holes in such a sample we can set $\tau_r = 10^{-10}$ sec and take the mobility to be $\mu_p = 10^4$ cm^2/V·sec, whence $U_{\rm HF} \propto \sigma(0) = Ge\mu_r\tau_r = 1.6\cdot10^{-25} G(\Omega\cdot\text{cm})^{-1}$. Hence $U_{\rm LF}/U_{\rm HF} \approx 10$. The experimental value of the ratio $U_{\rm LF}/U_{\rm HF}$ is at least an order of magnitude larger.[4] In view of the roughness of the estimate presented, and primarily the possible uncertainty in calculating the hopping time τ and taking into account the change ΔE in the dipole energy accompanying a hop, this discrepancy is not in conflict with the proposed model.

In Refs. 12 and 13 a theory of hopping photoconductivity in a system with localized states was developed for the case when the equilibrium state is reached due to diffusion of localized charge carriers, but the possibility of charges of different signs drifting toward one another was neglected. Taking this into account in the case of a doped and partially compensated semiconductor seems to us to be the key to analyzing hopping photoconductivity. The proposed model of hopping photoconductivity along excited metastable states of impurities in silicon agrees, at least qualitatively, with experiment.[1–4] The quantitative discrepancies could be connected with the strong departure of the electronic system equilibrium, unavoidable under experimental conditions. Attractive features of the model are the physically clear mechanism by which the excited and ionized atoms of the majority impurity approach one another, i.e., the drift of the ionized atoms in the electric field generated by the ions of the compensating impurity, and the possibility of restoring a stationary state due to the hopping mechanism of transfer of the ion charge.[5]

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