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Orlova, EE orcid.org/0000-0003-0243-0074, Kelsall, RW orcid.org/0000-0003-4830-2457, Deßmann, N et al. (4 more authors) (2018) Cascade capture of charge carriers in highly doped semiconductors. Journal of Applied Physics, 124 (8). 085704. ISSN 0021-8979

https://doi.org/10.1063/1.5035301

Cascade capture of charge carriers in highly doped semiconductors

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(Dated: June 29, 2018)

We analyze the cascade capture of charge carriers due to the interaction with acoustic phonons in highly doped semiconductors using a model that describes the recombination of photo-ionized carriers as a continuous relaxation of carriers in the energy space both at positive and negative energies in the field of a set of impurity ions. Such description enables simultaneous calculation of non-equilibrium carrier distribution formed by interaction with acoustic phonons in the presence of impurity traps, and the time of recombination in a wide range of concentrations of capture centers and phonon temperatures. Additionally, we calculated the time of cascade recombination in the presence of fast scattering processes forming a Maxwellian distribution of free carriers. We show, that experimentally observed concentration and temperature dependence of carrier life times in highly doped semiconductors can be described within the model of the cascade capture to uniformly spaced capture centers, and the main factor that determines the regime of cascade capture is the ratio of the thermal energy and the energy of the overlap of impurity potentials.

PACS numbers: 72.20.J,78.47.D-,71.55.-i

I. INTRODUCTION

The states of shallow impurity atoms in semiconductors are analogous to those of free atoms. As such, they provide the possibility to bring the rich variety of quantum effects observed in atoms and in molecular gases to the solid state, where the benefits of semiconductor device fabrication technologies and combined electrical and optical control can be used. A number of quantum electronic applications based on impurity atoms in semiconductors have been developed during the past two decades [1, 2], such as lasers on shallow impurity transitions [3, 4], and qubits based on impurity atoms [5–11]. Optical and electrical manipulation of qubits based on impurity atoms, and optical pumping of impurity lasers lead to ionization of a considerable fraction of impurity centers. Heating of the carrier distribution leads to reduction of the population of localized impurity states, to enhancement of impact interactions, and to reduction of coherence times of impurity states. The adverse effect of high free carrier concentrations on the performance of impurity based lasers and qubits can be reduced in highly doped samples with high compensation levels due to faster recombination. Fast recombination is also important for improvement of the speed of semiconductor detectors based on impurity transitions. However, a thorough theoretical treatment of recombination processes in highly doped semiconductors has not been developed so far. Such analysis is especially timely now due to a recent advance in time resolved spectroscopy investigations of relaxation to impurity centers [8, 12–15], providing the necessary experimental basis for verification of the proposed approach.

Recombination to shallow impurity centers in semiconductors was investigated extensively due to their role as a source of free carriers in semiconductor electronic devices. It was found that the main recombination process in a wide range of temperatures and concentrations of impurity centers is cascade capture due to the interaction with acoustic phonons [16, 17]. The characteristic energy of the emitted phonon is much smaller than the ionization energy, thus excited carriers relax gradually through the ladder of excited impurity states. The model of cascade capture was originally developed for isolated Coulomb centers [16, 18]. This model implies that the capture rate is proportional to the concentration of impurity ions. The measurements of impurity photoconductivity [19] have shown that the concentration dependence of the carrier life time becomes weaker in highly doped samples. A

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similar weak concentration dependence of recombination time has been observed recently in pump probe investigations of impurity absorption relaxation [15] A qualitative model of cascade capture in highly doped semiconductors have been proposed earlier in [17], explaining the weak concentration dependence of the carrier life times by the influence of the non-uniformity of the spatial distribution of impurity ions. This model implies that the non-uniformity of impurity ion distribution leads to fluctuations of potential at the bottom of the band [20], and the carriers with the energy below the amplitude of potential fluctuations can be regarded as captured, as they do not take part in conductivity. Capture time is then determined as the energy relaxation time of the carriers with the energy equal to that of the amplitude of potential fluctuations. However, this model is not appropriate for the analysis of recombination times obtained from the measurements of impurity absorption relaxation [15], as the carriers relaxing below the level of potential fluctuations still influence the absorption modulation. Moreover, the non-uniform potential of impurity centers is not the only factor that determines cascade capture at high doping concentrations. The overlap of the orbits essential for cascade capture occurs also in the case of a uniform distribution of impurity ions. Additionally, fast cascade capture at high doping concentrations can lead to formation of non-equilibrium carrier distributions, with reduced population of the states near the band edge that are responsible for cascade recombination [21]. However, quantitative analysis of these factors has not been performed yet.

Here we investigate the dependence of cascade recombination on the doping level using the quasi-classical approach for the calculation of the carrier relaxation in the field of a set of impurity ions [22]. This approach enables the account of the overlap of the potentials of the neighboring impurity centers and the analysis of non-equilibrium distribution of photo-ionized carriers formed by acoustic phonon assisted relaxation and recombination. Using this model, we derive integral expressions describing the cascade capture time in a wide range of doping concentrations and temperatures. Additionally, we calculated the time of cascade recombination in the presence of fast scattering processes forming a Maxwellian distribution of free carriers. We show that concentration and temperature dependence of recombination time observed in highly doped semiconductors [15, 19] can be described within the model of uniformly spaced impurity ions (Fig. 1.). We explain the different regimes of cascade capture by the balance of the fluxes of charge carriers in the energy space: the relaxation flux due to spontaneous emission of acoustic phonons and the opposing thermal diffusion flux due to induced phonon assisted transitions. We show that this balance is determined by the ratio of the thermal energy and the energy of overlap of impurity potentials, that determines the energy scale of the change of the density of states in the field of impurity centers.

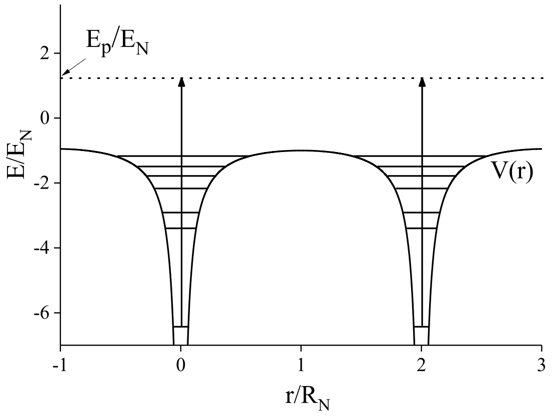

FIG. 1: Potential of a set of uniformly spaced charged attractive centers. $E_N = e^2/\varepsilon R_N$ is the energy of the overlap of impurity potentials, $2R_N$ – the distance between the neighboring centers, E_p – energy of photo-ionized carriers.

II. ACOUSTIC PHONON ASSISTED RELAXATION IN THE FIELD OF A SET OF ATTRACTIVE COULOMB CENTERS

We calculate the time of cascade capture in the field of a set of attractive Coulomb centers due to interaction with acoustic phonons using the quasi-classical approach, based on the assumption that the scattering probabilities of a charge carrier in the field of impurity centers are the same as that of a free carrier with the energy equal to the kinetic energy of a charge carrier in the given point of space. Quasi-classical description of cascade capture can be used when the energy gap between impurity states does not exceed the characteristic energy of emitted phonons. This approach has been proposed in [16], and further refined in [18], where the rate of the cascade capture to isolated attractive Coulomb centers was calculated assuming equilibrium distribution of free carriers.

Carrier distribution $f(\mathbf{r},\mathbf{p})$ in the field of attractive centers is described by the Boltzmann kinetic equation. Momentum relaxation due to interaction with acoustic phonons is much faster than energy relaxation, thus we assume that the distribution function f depends only on the total energy in the field of impurity ions:

$$E = \mathcal{E} + V(\mathbf{r}), \tag{1}$$

where $\mathcal{E}$ is kinetic energy of charge carriers and $V(r)$ is the potential of a set of attractive impurity centers. In order to account for the overlap of the orbits of the neighboring attractive Coulomb centers and for the formation of non-equilibrium distribution of photo-ionized carriers, we modify the procedure proposed in [18], describing the process of recombination as a continuous relaxation of charge carriers in the space of both positive and negative total energies in the field of a set of attractive centers

(Fig. 1.). We describe the source of photo-ionized carriers by a delta-function at the energy E_p assuming the width of the radiation spectrum much smaller than the energy of overlap of impurity potentials and the thermal energy. Since the energies of emitted acoustic phonons are usually much smaller than the electron kinetic energy, collision integral describing interaction with acoustic phonons can be presented in the differential form:

$$\tilde{\rho}\frac{\partial f}{\partial t} = \frac{\partial J}{\partial E} + I\delta(E - E_p). \tag{2}$$

Here I is the number of photo-ionized charge carriers in the unit time in the unit volume, J is the relaxation flux of carriers in energy space due to interaction with equilibrium acoustic phonons with thermal energy T [18]:

$$J = \tilde{B}(E)\left(f + T\frac{\partial f}{\partial E}\right). \tag{3}$$

Here we assume that the carrier flux J in the direction of smaller energies is positive. The first term in equation (3) describes the flux of carriers in the energy space due to spontaneous emission of acoustic phonons (J_s), it is analogous to drift motion under a constant force. The second term, proportional to the derivative of the distribution function, is the carrier flux due to induced phonon assisted transitions (J_d). This flux describes thermal diffusion of carriers in the energy space. The drift coefficient in the energy space $\tilde{B}(E)$ and the density of states $\tilde{\rho}(E)$ are obtained by averaging of the corresponding functions of kinetic energy $\mathcal{E}$ of charge carriers over the surface of a constant total energy E in the field of a set of attractive impurity centers:

$$\tilde{\rho}(E) = \frac{1}{V}\int\limits_V \int \rho(\mathcal{E})\delta(E - \mathcal{E} - V(\mathbf{r}))d\mathcal{E}\mathbf{dr}, \tag{4}$$

$$\tilde{B}(E) = \frac{1}{V}\int\limits_V \int B(\mathcal{E})\delta(E - \mathcal{E} - V(\mathbf{r}))\, d\mathcal{E}\, \mathbf{dr}. \tag{5}$$

Here $\rho(\mathcal{E})$ and $B(\mathcal{E}) = \rho(\mathcal{E})\mathcal{E}\tau_{\mathcal{E}}^{-1}(\mathcal{E})$ are the density of states and the energy drift coefficient of free carriers with the energy $\mathcal{E}$, and $\tau_{\mathcal{E}}(\mathcal{E})$ is the time of the energy relaxation due to spontaneous emission of acoustic phonons. The meaning of the energy drift coefficient B is that being multiplied by f it gives the carrier flux in the energy space with the velocity of energy relaxation due to spontaneous emission of acoustic phonons: $v_{\mathcal{E}} = \mathcal{E}\tau_{\mathcal{E}}^{-1}(\mathcal{E})$.

The stationary solution of equation (2) corresponds to the constant relaxation flux J everywhere except for the vicinity of the energy of the source of photo-ionized carriers E_p. The relaxation flux independent on the energy is a manifestation of the conservation of the number of carriers under conditions of continuous relaxation in the energy space. Imposing zero boundary condition at large positive energies, reflecting the presence of only one source of carriers at E_p, and at a negative energy E_1, $|E_1| >> T$, where carriers can be considered as captured, we get:

$$f(E) = \begin{cases} \frac{J}{T}\int\limits_{E_1}^{E} \frac{\exp((x - E)/T)}{\tilde{B}(x)}dx & E \le E_p \\ \frac{J}{T}\int\limits_{E_1}^{E_p} \frac{\exp((x - E)/T)}{\tilde{B}(x)}dx & E \ge E_p \end{cases} \tag{6}$$

with the stationary flux of recombining carriers equal to the flux of photo-ionized carriers: $J = I$.

The recombination time is expressed in terms of the distribution function using $J = n\tau_{rec}^{-1}$, where $n = f(-E_N)N_z$ is the concentration of free carriers, the effective number of free carrier states is:

$$N_z = \int_{-E_N}^{\infty} \tilde{\rho}(E)\frac{f(E)}{f(-E_N)}dE, \tag{7}$$

and E_N is the energy of overlap of impurity potentials:

$$\tau_{rec} = \frac{f(-E_N)}{J}N_z \tag{8}$$

Fast impact processes at high free carrier concentrations may lead to formation of quasi-equilibrium distribution of free carriers: $f_T(E) = \exp((-E - E_N)/T_e)n/N_{z,T}(T_e)$ with the effective number of free states for thermalized carrier distribution:

$$N_{z,T_e} = \int_{-E_N}^{\infty} \tilde{\rho}(E)\exp\left(\frac{-E - E_N}{T_e}\right)dE. \tag{9}$$

The effective temperature of electron distribution can differ from the lattice temperature $T_e \ne T$ in the case of electric field heating. Probabilities of impact processes decrease fast with the increase of the energy of transition between the localized states. Carrier distribution over the localized states at the energies below $E = -E_N$, where acoustic phonon assisted transitions are dominant, can be obtained as a stationary solution of the Fokker-Planck equation:

$$\tilde{\rho}\frac{\partial f}{\partial t} = \frac{\partial J}{\partial E}, \tag{10}$$

satisfying zero boundary condition at large negative energy E_1 and continuity boundary condition at $E = -E_N$. This solution is given by (6) at $E \le -E_N$ with $J = n\tau_{rec,T_e}^{-1}$ determined from the boundary conditions at $E = -E_N$, leading to the expression for the recombination time of the thermalized carriers:

$$\tau_{rec,T_e} = \frac{f(-E_N)}{J}N_{z,T_e}. \tag{11}$$

For comparison, the recombination time $\tau_{rec,i}$ for isolated centers in the case of the equilibrium carrier distribution obtained in [18] coincides with that given by expression (11) with $T_e = T$ in the limit of $E_N << T$. As we will show further, acoustic phonon assisted relaxation and recombination of photo-excited carriers forms a Maxwellian distribution with $T_e = T$, and the recombination time τ_{rec} is close to $\tau_{rec,i}$ when the energy of overlap of impurity potentials is much smaller than the lattice temperature $E_N << T$.

Let us now make assumptions about the band structure. We will imply a spherical non-degenerate parabolic band with effective mass m, and the density of states (without account of spin) $\rho(\mathcal{E}) = 4\sqrt{2}\pi m^{3/2}\sqrt{E}/(2\pi\hbar)^3$. The energy relaxation time in a parabolic band due to spontaneous emission of acoustic phonons for energies $\mathcal{E} >> ms^2$ producing the main impact to integral (5) is:

$$\tau_{\mathcal{E}}(\mathcal{E}) = L_0\sqrt{m/(2\mathcal{E})}, \qquad (12)$$

with L_0 being the scattering length [24].

We calculate the density of states $\tilde{\rho}$ and the drift coefficient in the energy space $\tilde{B}$ implying the uniform distribution of impurity ions in space. This approach, first proposed in the model of silicon donor lasers [22], and later used for investigation of dynamics of cascade capture [23], excludes the influence of potential fluctuations claimed as the main factor that determines the capture rate at large doping concentrations [17]. On the other hand, our results are valid also for non-uniform spatial distribution of impurity ions as long as it does not lead to a significant change of the integral parameters $\tilde{\rho}$ and $\tilde{B}$. Further, we approximate the integrals in (4) and (5) replacing the volume belonging to each impurity center by a sphere with the radius

$$R_N = (4\pi N_+/3)^{-1/3}, \qquad (13)$$

with the potential inside this volume $V(r) = -e^2/\varepsilon r$, and the energy of the overlap of impurity potentials

$$E_N = e^2/\varepsilon R_N. \qquad (14)$$

This approximation involves modifications in the regions with relatively small kinetic energy of charge carriers, producing a relatively small impact on the integrals (4) and (5).

These approximations allow us to express the density of states $\tilde{\rho}$ and the energy drift coefficient $\tilde{B}$ in elementary functions of E/E_N (see Appendix) with the correct behavior at negative energies below the energy of the overlap of potentials of impurity centers $E = -E_N$, and at large positive energies $E >> E_N$ (Fig. 2.). At $E < -E_N$ $\tilde{\rho}(E)$ and $\tilde{B}(E)$ are equal to the corresponding parameters of the isolated hydrogen like centers $\rho_i = \dfrac{E_B^{3/2}}{2|E|^{5/2}}$ and $B_i = \dfrac{8}{3\pi}\dfrac{E_B}{\hbar}\dfrac{e^2}{L_0\varepsilon|E|}$ ($E_B = e^4m/2\varepsilon^2\hbar^2$ is Bohr energy) multiplied by the concentration of impurity ions, while at $E >> E_N$, $\tilde{\rho}(E)$ and $\tilde{B}(E)$ approach the corresponding parameters of the free carriers $\rho(E)$ and $B(E)$. The fastest relative change of $\tilde{\rho}(E)$ and $\tilde{B}(E)$ occurs at the energies near the overlap of impurity potential, with $\tilde{\rho}(-E_N) = (3\pi/16)\rho(E_N)$ and $\tilde{B}(-E_N) = B(E_N)$, and the characteristic energy range where these parameters change considerably is about E_N (see the inserts in Fig. 2). As we show below, it is the maximum of the relative speed of change of the energy drift coefficient $\tilde{B}(E)$ that plays the role of the bottle-neck for relaxing carriers, controlling the balance of the carrier fluxes and the carrier distribution in the energy space. And the main factor that causes the decrease of the drift coefficient with reduction of energy is the decrease of the density of states in the field of impurity ions.

The distribution function of photo-ionized carriers (A.3) obtained introducing (A.1) and (A.2) in (6), normalized to the occupation number at the energy of the overlap of impurity potentials $f(-E_N) = J(T/E_N + 1)/B(E_N)$, is presented on Fig. 3. The shape of the

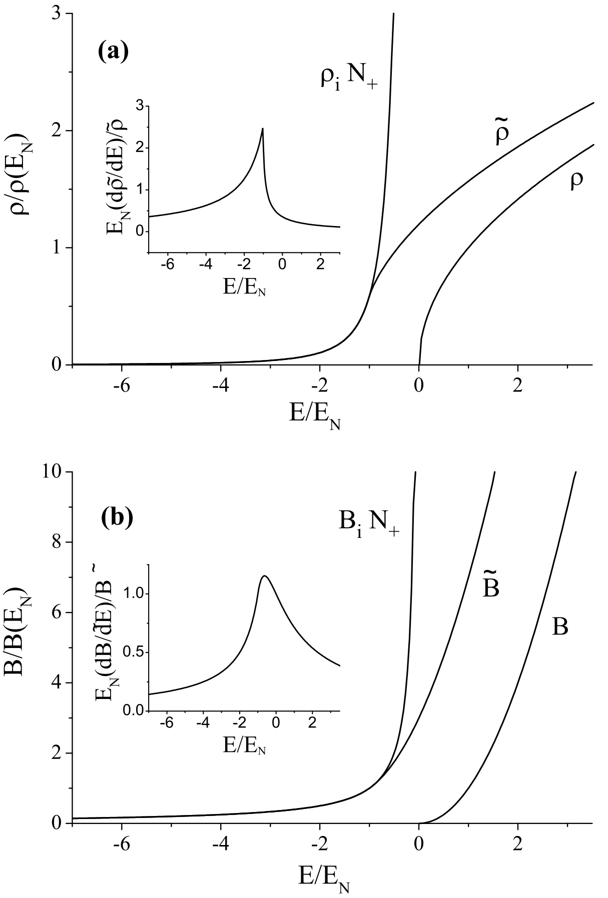

FIG. 2: The densities of states: ρ – for continuum with parabolic dispersion law, ρ_i – quasi-continuum of localized states of an isolated Coulomb center, $\tilde{\rho}$ – in the field of uniformly spaced Coulomb centers with the concentration N_+ (A.1) (a). The drift coefficient in the energy space: B – continuum , B_i –in the field of an isolated Coulomb center, $\tilde{B}$ – in the field of uniformly spaced Coulomb centers (A.2) with the concentration N_+ (b). Inserts: relative speed of change of $\tilde{\rho}$ and $\tilde{B}$ in the energy space normalized to E_N^{-1}.

energy dependence of the distribution function at the bottom of the band strongly depends on the ratio of the temperature to the energy of the overlap of impurity potentials T/E_N. At $T/E_N << 1$, the energy drift coefficient $\tilde{B}$ does not change much within thermal energy T, and, at the energies below E_p, the first term of (3), corresponding to the relaxation flux due to spontaneous emission of acoustic phonons J_s, is much larger than the second one, corresponding to thermal diffusion flux $J_d(E)$ (Fig. 4). The distribution function at $E < E_p$ is then close to

$$f \simeq J/\tilde{B}(E), \tag{15}$$

with the characteristic energy range of the population decrease $E_N >> T$. This indicates the reduction of the population of the low energy states due to a fast cascade process. The ratio of the fluxes $J_d/J_s = T\frac{\partial f}{\partial E}/f$ is than determined by the relative change of the energy drift coefficient within the thermal energy $T\frac{\partial \tilde{B}}{\partial E}/\tilde{B}$, and the maximum of the latter is about T/E_N (Fig. 2.). At $T/E_N >> 1$, the fast change of the energy drift coefficient $\tilde{B}$ within the thermal energy leads to a drastic increase of thermal diffusion flux towards higher energies (Fig. 4 (b)). The opposing carrier fluxes than have close magnitudes at the energies above $-E_N$, much larger than the resulting recombination flux: $|J_d(E > -E_N)|/J > T/E_N >> 1$. The carrier distribution for $-E_N < E < E_p$ is then close to the solution of (3) with $J = 0$, a Maxwellian function $f \sim \exp(-E/T)$. This indicates the formation of a bottleneck for relaxing carriers and their accumulation and thermalization at the energies above that of the maximum of relative speed of change of the energy drift coefficient $\frac{\partial \tilde{B}}{\partial E}/\tilde{B}$. The carrier distribution above the pumping energy E_p is always described by a Maxwellian function due to the zero stationary flux of carriers in this energy region (the sharp change of the slope of the distribution function and of the carrier flux curves at $E = E_p$ for $T << E_N$ is due to the delta-function approximation of the energy dependence of the photo-ionization flux). Thus, the characteristic energy range of decrease of free carrier distribution function is equal to the lattice temperature T at $T >> E_N$, and at $T << E_N$ it extends to E_N or $E_p + E_N + T$, whichever is smaller.

The calculated capture times are presented on Fig. 5. The capture time with account of non-equilibrium distribution of relaxing carriers is obtained by substitution of (A.1) and (A.3) in (8):

$$\tau_{rec} = \tau_{\mathcal{E}}(E_N)\left(\frac{T}{E_N}+1\right)F_N\left(\frac{T}{E_N},\frac{E_p}{E_N}\right), \tag{16}$$

and the capture time in the model of thermalized free carrier distribution is obtained by substitution of (A.1)

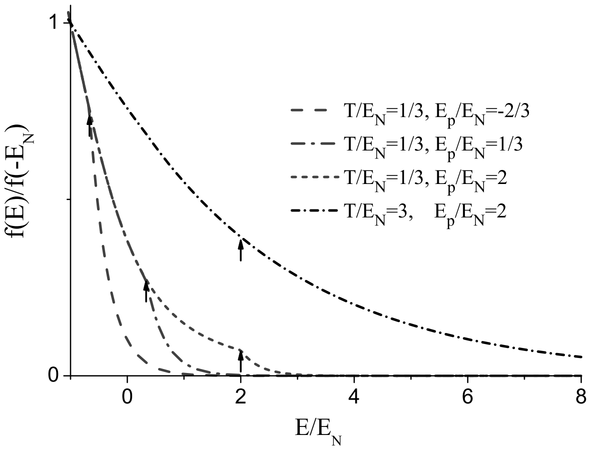

FIG. 3: Normalized distribution function for several values of T/E_N and Ep/E_N (a). Energies of the photo-ionized carriers are indicated by arrows. Note, that Maxwellian distribution with $T_e = T$ is formed when $T >> E_N$, and at any T when the pumping energy E_p is close to the energy of overlap of impurity potentials.

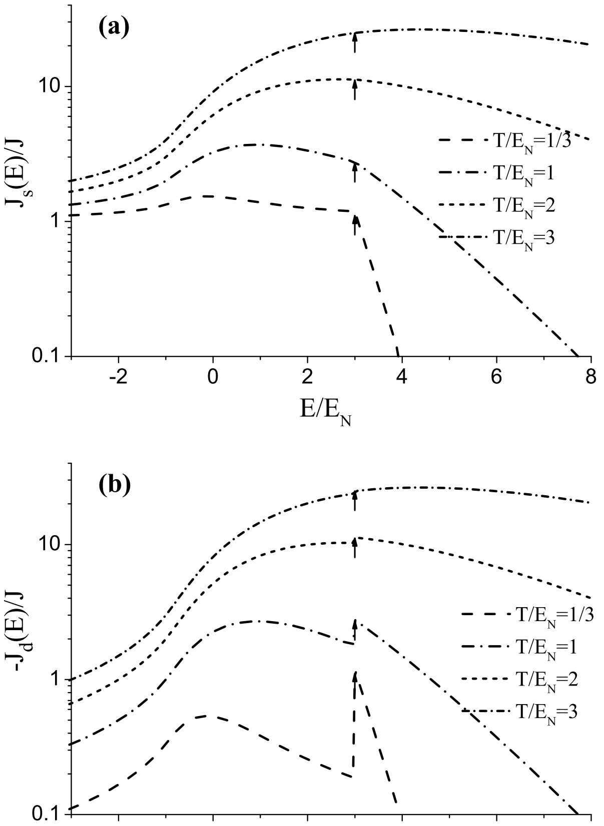

FIG. 4: Normalized relaxation flux due to spontaneous emission of acoustic phonons (a), and normalized thermal diffusion flux (b) for several values of T/E_N, with $E_p = 3E_N$. The energy of the photo-ionized carriers are indicated by arrows.

in (11):

$$\tau_{rec,T_e} = \tau_{\mathcal{E}}(E_N) \left(\frac{T}{E_N} + 1 \right) F_{N,T_e} \left(\frac{T_e}{E_N} \right). \qquad (17)$$

Here $\tau_{\mathcal{E}}$ is given by (12), and the dimensionless functions

$$F_N(y,z) = \int_{-1}^{\infty} F_\rho(x) F(x,y,z) dx / (T/E_N + 1), \qquad (18)$$

and

$$F_{N,T_e}(y) = \int_{-1}^{\infty} F_\rho(x) \exp\left(\frac{-x-1}{y} \right) dx \qquad (19)$$

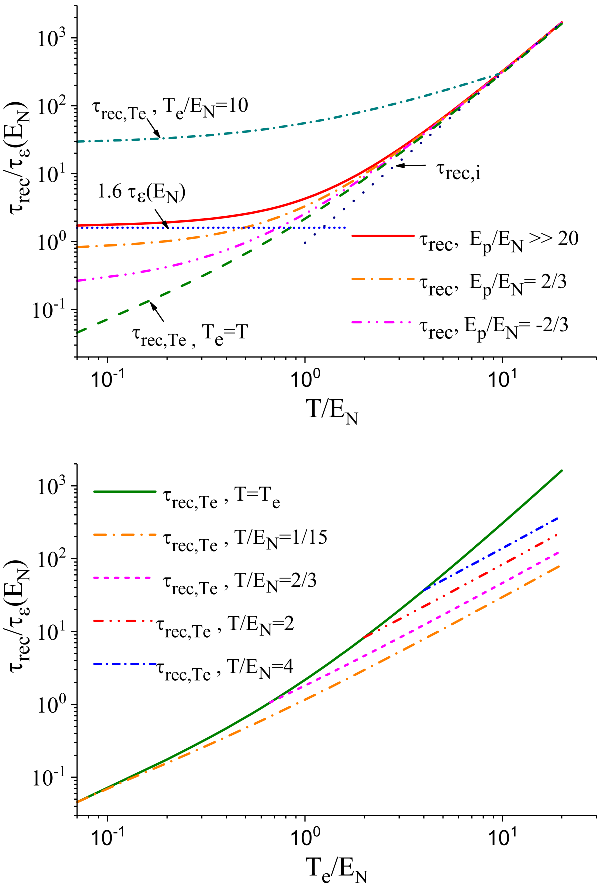

FIG. 5: (Color online) The time of cascade capture calculated using different approaches, normalized to the free carrier energy relaxation time due to spontaneous emission of acoustic phonons $\tau_{\mathcal{E}}$ at the energy of overlap of impurity potentials E_N: τ_{rec} – with account of the overlap of impurity potentials and non-equilibrium distribution of photo-ionized carriers (16); τ_{rec,T_e} – with account of the overlap of impurity potentials assuming Maxwellian distribution of free carriers (17) with the effective temperature $T_e \geq T$, (T – the lattice temperature); $\tau_{rec,i}$ – recombination time within the model of isolated centers [17].

represent the effective numbers of free carrier states normalized to $\rho(E_N)E_N$. The growth of these functions with the increase of the energy range of the free carrier distribution reflects the influence of the growth of the fraction of high energy carriers that do not participate in cascade capture. The factor $T/E_N + 1$ in (16) and (17) is the ratio of the relaxation flux due to spontaneous emission of acoustic phonons at the energy of overlap of impurity potentials to the total relaxation flux.

An important feature of the cascade capture times given by expressions (16) and (17) is that the temperature and the concentration of impurity ions enter $\tau_{rec,T_e}/\tau_{\mathcal{E}}$ and $\tau_{rec}/\tau_{\mathcal{E}}$ at $E_p/E_N >> 1$ only in the combinations T/E_N and T_e/E_N. This means that the temperature dependence and the concentration dependence of recombination time are linked, and the proportional change of T, T_e and $E_N = e^2(4\pi N_+/3)^{1/3}/\varepsilon$ leaves the ratio of the recombination time to the relaxation time at the energy E_N unchanged.

The dependence of capture times on T/E_N calculated within both models becomes weaker at small T/E_N. However, the decrease at small T/E_N of the capture time calculated with account for non-equilibrium distribution of relaxing carriers, τ_{rec}, is much slower. The behavior of τ_{rec} at small T/E_N is in agreement with the slowing of the concentration and temperature dependence observed at high concentrations of capture centers [15, 19], and with the fact that slowing down of the temperature dependence commences at higher temperatures in the samples with higher impurity ion concentrations [19]. The detailed comparison of the calculated results with experimental data [15, 19] will be given in the next section.

Let us now consider cascade capture in the limiting cases of high and low T/E_N and T_e/E_N.

For low concentrations of impurity ions $T/E_N >> 1$, when impurity potential creates a bottleneck for relaxing carriers, and recombination flux is much smaller than thermal diffusion flux forming a Maxwellian distribution of free carriers with $T_e = T$, the effective number of free carrier states is $N_z \simeq (\sqrt{\pi}/2)\rho(T)T$, leading to a recombination time:

$$\tau_{rec} = \tau_{\mathcal{E}}(E_N) \frac{\sqrt{\pi}}{2} \left(\frac{T}{E_N} \right)^{5/2}. \qquad (20)$$

This time is inverse proportional to the concentration of impurity ions and coincides with the cascade capture time in the limit of the isolated impurity centers [17].

At high concentrations of impurity ions with $T/E_N << 1$ and $E_p/E_N >> 1$, the energy range of decrease of the non-equilibrium distribution function is about E_N, and the effective number of states of free carriers $N_z \simeq 1.6\rho(E_N)E_N$. Recombination time is then given by:

$$\tau_{rec} \simeq 1.6\, \tau_{\mathcal{E}}(E_N). \qquad (21)$$

This time does not depend on the temperature, and decreases very slowly with the increase of impurity ion concentration as $\tau_{rec} \sim N_+^{-1/6}$. For comparison, the time of cascade capture in the limit of high concentrations of impurity centers defined in [17] as the relaxation time at the percolation energy (the amplitude of potential fluctuations due to non-uniformity of impurity ion distribution) $E_0 = 0.3K^{-1/12}e^2 N_+^{1/3}/\varepsilon$, where K is the compensation level, differs from that given by expression (21) by a factor $\tau_{\mathcal{E}}(E_0)/1.6\,\tau_{\mathcal{E}}(E_N) \simeq 1.5K^{1/24}$ that varies within the range $1.1 \div 1.5$ for K above 10^{-3}. Thus, in the limit of high concentration of capture centers, our model produces the result close to that of the qualitative model of [17], although our explanation does not involve the non-uniformity of spatial distribution of impurity ions and capture of carriers in the wells formed by potential fluctuations. Within our model the change of the concentration dependence of recombination time is explained by the change of the energy dependence of the density of states, leading to the change of the balance of the fluxes of carriers in the energy space. At $T/E_N << 1$ the density of states and the energy drift coefficient do not change much within the thermal energy, impurity potential does not form a bottleneck for carrier relaxation. Thermal carrier flux is then negligible, and the life time of a free carrier is determined by the slowest time of energy relaxation due to spontaneous phonon emission. This time corresponds to the minimum average kinetic energy of carrier states in the field of a set of impurity ions, and the latter is equal to the energy of the overlap of impurity potentials.

Recombination time at high concentrations of impurity ions decreases when carriers are excited with the energies slightly above the energy of the overlap of impurity potential $-E_N < E_p < -T$. The effective number of free carrier states is then determined by $Nz \simeq 0.6\rho(E_N)(E_p + E_N + T)$, and the recombination time:

$$\tau_{rec} \simeq \tau_{\mathcal{E}}(E_N)\,0.6\left(\frac{E_p}{E_N} + \frac{T}{E_N} + 1\right). \qquad (22)$$

In the case of formation of a quasi-equilibrium population of free states with effective temperature T_e, at low concentrations of impurity centers with $T_e/E_N >> 1$, recombination time is given by:

$$\tau_{rec,T_e} = \tau_{\mathcal{E}}(E_N)\frac{\sqrt{\pi}}{2}\left(\frac{T}{E_N} + 1\right)\left(\frac{T_e}{E_N}\right)^{3/2}. \qquad (23)$$

For lattice temperatures $T >> E_N$, this time coincides with the time of cascade capture of hot carriers obtained in the model of isolated impurity centers [25].

In the limit of high concentration of impurity ions with $T_e/E_N << 1$ and $T/E_N << 1$ the effective number of free states for a Maxwellian carrier distribution is determined by $N_{z,T_e} \simeq 0.6\,\rho(E_N)T_e$, and the recombination time is:

$$\tau_{rec,T_e} \simeq \tau_{\mathcal{E}}(E_N)\,0.6\,\frac{T_e}{E_N}. \qquad (24)$$

In contrast to the high concentration limit of the cascade capture time from [17], this time grows linearly with temperature T_e and exhibits a faster dependence on the concentration of impurity ions: $\tau_{rec,T_e} \sim N_+^{-1/2}$.

Thus, we derived the expressions for cascade capture time in the model of uniformly spaced impurity ions. A similar description of carrier relaxation in the field of a set of ions can be used for an arbitrary ion distribution. Let us estimate the influence of non-uniformity of impurity ion distribution on recombination time. We suppose, that the spatial average of impurity ion concentration is $\overline{N_+}$, and the distribution of ions is described by an even probability density function: $P(N_+ - \overline{N_+})$. The mean recombination rate can then be determined by:

$$\overline{W} = \int W(N_+)P(N_+ - \overline{N_+})dN_+, \qquad (25)$$

where $W(N_+) = \tau_{rec}^{-1}$ is calculated in the model of the uniform ion distribution. Assuming the with of the maximum of $P(N_+ - \overline{N_+})$ smaller than characteristic range of change of $W(N_+)$, we can present the latter as a power series of $(N_+ - \overline{N_+})$. The impact of the term proportional to the first derivative of $W(N_+)$ to the mean recombination rate is zero for an even distribution $P(N_+ - \overline{N_+})$. Thus, the correction to the recombination rate due to the non-uniformity of impurity ion distribution is determined by:

$$\overline{W} - W(\overline{N_+}) \simeq \frac{1}{2}\left.\frac{\partial^2 W}{\partial N_+^2}\right|_{\overline{N_+}} \overline{(N_+ - \overline{N_+})^2}. \qquad (26)$$

This magnitude is zero for small concentrations of impurity ions $E_N/T << 1$ due to a linear dependence $W(N_+)$. For highly doped semiconductors with $E_N/T >> 1$ the relative correction of the capture rate is $(\overline{W} - W(\overline{N_+}))/W(\overline{N_+}) \simeq -0.07(\overline{(N_+ - \overline{N_+})^2}/\overline{N_+}^2)$, that is much smaller than unity even in the case of the root-mean-square deviation of the ion concentration comparable to the mean ion concentration value.

III. COMPARISON WITH EXPERIMENTAL DATA

The amount of experimental data on cascade capture times in semiconductors is vast, with majority of investigations performed with relatively low doped semiconductors. These data has been reviewed in [17], and a good agreement with the model of cascade capture to isolated impurity ions [18] has been demonstrated. Recombination time calculated using our model coincides with that of [18] when the energy of the overlap of ion potentials is smaller than thermal energy. Only few experimental investigations [13–15, 19] were dealing with relatively high concentrations of capture centers. Below we verify our approach to calculation of the cascade capture in highly doped semiconductors using the temperature dependence of recombination time obtained from photo-conductivity

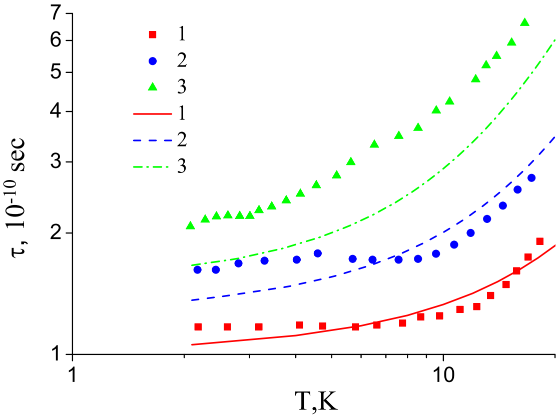

FIG. 6: Temperature dependence of the life time of photo-excited carriers in p-Si. The measured life times in Si:B [19] (simbols) with donor concentrations N_d, cm^{-3}: 1 – 2.3×10^{15}, 2 – 6.0×10^{14}, 3 – 2.3×10^{14}, and the cascade capture times in p-Si calculated using (16), with $E_p >> E_N$ (lines), N_+, cm^{-3}: 1 – 2.3×10^{15}, 2 – 6.0×10^{14}, 3 – 2.3×10^{14}.

investigations of p-Si [19] and the concentration dependence of recombination time obtained from the measurements of the power dependence of the relaxation of transmission modulation in p-Ge [15].

Figure 6 presents the comparison of recombination time calculated using (16) with the data of [19]. Parameters used in the calculations are: effective mass equal to the mass of the density of states of Si valence band $m = 0.5m_0$ with m_0 – the electron mass, the scattering length $L_0 = 3\times10^{-4}$ cm, and the dielectric constant of Si $\varepsilon = 11.9$. The figure shows that both the character of the temperature dependence and the absolute values of the life times calculated using our model are in good agreement with recombination times observed in p-Si with high levels of doping [19]. A small mismatch of experimental data with the curve with $N_+ = 2.3\times10^{14}$ cm^{-3} can be explained by a decrease of the number of Coulomb capture centers due to a correlation of location of charged acceptors and compensating donor centers, that becomes important at small temperatures [20, 26].

The low compensation of Ge sample used in pump probe experiment [15] enables a wide range of variation of the concentration of charged acceptors N_+ by changing the power of excitation pulse (Fig. 7.). The influence of acceptor-donor dipoles in pump probe experiments, where the concentration of charged acceptors is determined by optical ionization and exceeds considerably the donor concentration, is small. A Ge sample used in pump probe experiments [15] was doped with Ga acceptor with concentration $N_a = 2\times10^{15}$ cm^{-3} and a residual donor concentration N_d was below 2×10^{13} cm^{-3}. The investigated sample was cooled to about 5 K. The capture time was determined as the decay time of transmittance modulation measured after the excitation by a 10 ps pulse of laser radiation providing photon energies slightly exceeding the ionization energy of Ga centers in Ge, with $E_p = 0.5$ meV. We calculate the concentration of the

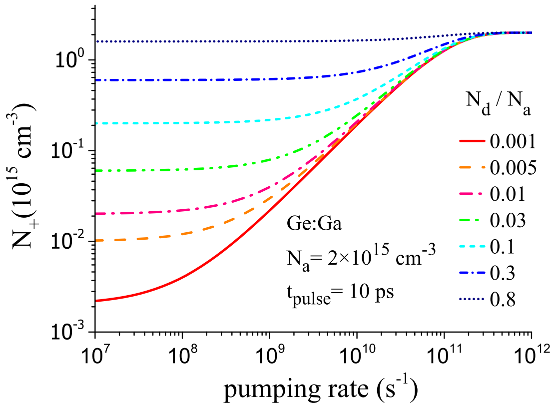

FIG. 7: (Color online) Concentration of charged impurity centers in Ge:Ga sample after the pump pulse for different compensation levels.

charged acceptors $N_+ = N_d + n$ taking into consideration that the duration of the excitation pulse (10 ps) is shorter than the recombination time, and assuming that excited impurity states are not populated within the pulse duration. Under these conditions, the population of the ground acceptor state $n_{g.s.}$ is described by the equation:

$$\frac{dn_{g.s.}}{dt} = -W_p n_{g.s.}, \tag{27}$$

where $W_p = P\sigma$ is the pumping rate, P – the photon flux, and σ – the cross section of optical ionization (ionization cross section of the ground state of Ga acceptor in Ge determined from absorption spectra [27] is $\sigma \simeq 9\times10^{-15}$ cm^{-2}). Solving (27) with $n_{g.s.}(t=0) = N_a - N_d$ and using the condition of charge-neutrality $N_a = N_d + n + n_{g.s.}$, we obtain:

$$N_+ = (N_a - N_d)(1 - \exp(-W_p t)) + N_d. \tag{28}$$

The dependence of the concentration of the ionized centers on the pumping rate depends strongly on the compensation level. Indeed, without pumping N_+ is equal to the concentration of compensating impurity N_d; it grows linearly at small pumping intensities and saturates at the level $N_+ = N_a$ when the optical ionization time becomes shorter than the pump pulse duration and the ground impurity state is depleted. According to Fig. 7, the concentration of ionized centers in the sample investigated in [15], with the compensation below $K = 0.01$, was growing almost linearly throughout the whole range of pumping rates, varying between 2×10^{13} cm^{-3} and 2×10^{15} cm^{-3}.

Figure 8 shows the concentration dependence of the cascade capture times in p:Ge at $T = 5$ K obtained using our approach, with account of formation of non-equilibrium distribution of photo-ionized carriers, for Maxwellian carrier distribution with $T_e = T$, and with the models proposed in [17]. Here we use the parameters: effective mass equal to the mass of the density of states of Ge valence band $m = 0.38m_0$, the scattering

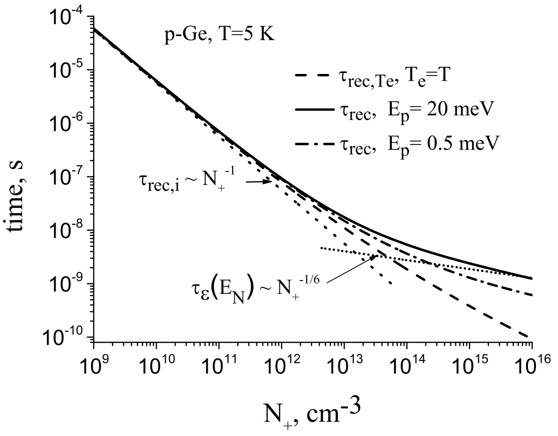

FIG. 8: Dependence of the cascade capture time in p-Ge on the concentration of impurity ions calculated using different approaches: τ_{rec,T_e} – with account of overlap of impurity potentials assuming equilibrium distribution of free carriers (17), τ_{rec}– with account of overlap of impurity potentials and non-equilibrium distribution of photo-ionized carriers (16), $\tau_{rec,i}$ – within the model of isolated centers [17]; $\tau_{\mathcal{E}}(E_N)$ is the energy relaxation time due to spontaneous emission of acoustic phonons at the energy of the overlap of impurity potentials.

length $L_0 = 4.3 \times 10^{-3} cm$, and the dielectric constant of Ge $\varepsilon = 16$. According to our calculations, the influence of the overlap of impurity potentials becomes significant already at the concentrations of charged acceptor centers in Ge about 10^{12} cm^{-3}. At concentrations about 1.2×10^{16} cm^{-3} the energy of potential barriers between impurity ions approaches the binding energy of the second excited impurity state, and at higher concentrations of impurity ions the model of cascade capture is not adequate. The minimum cascade capture time (about 100 ps at $T = 5$ K) can be reached at the concentration of charged acceptors in Ge about 10^{16} cm^{-3} in the case of the thermalized carrier distribution with $T_e = T$.

Let us now compare the measured recombination time dependence on the pump power [15] with that calculated using different models proposed in this paper and in [17]. First we compare the experimental results with the model proposed in [17] for high concentrations of capture centers, and with that of the model of cascade capture of thermalized carriers with $T_e = T$ to a set of Coulomb centers (17) (Fig. 9). We see that, although the recombination times calculated using the model of [17] have the right order of magnitude, their dependence on the pump power differs strongly from the observed one. Recombination times calculated using (17) are faster than the measured recombination times, and the difference increases with the increase of the concentration of impurity ions, indicating the significant role of heating of free carrier distribution caused by fast recombination. Indeed, our model that accounts for the formation of non-equilibrium distribution of photo-ionized carriers (16) provides a much better fit with the experimental data (Fig. 10). In agreement with out model, the measured recombination time decreases fast at small pumping levels, where the concentration of capture centers is relatively small. The decrease of the capture time slows down before the number of ionized centers saturates. This indicates the influence of the overlap of the potentials of impurity centers and non-equlibrium distribution of photo-ionized carriers. Overall, we can conclude, that our model (17) describes well the capture of photo-ionized carriers in p-Ge within the wide range of charged acceptor concentrations from 2×10^{13} cm^{-3} to 2×10^{15} cm^{-3} that have been realized in [15].

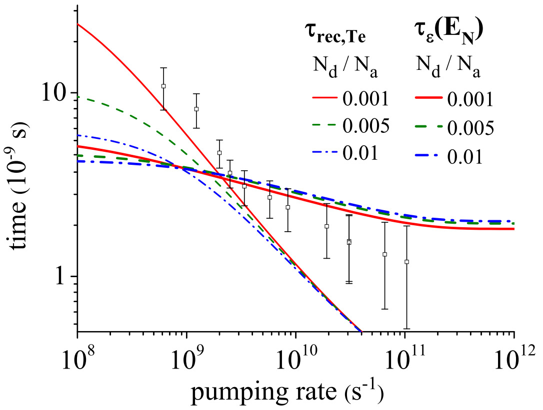

FIG. 9: (Color online) Comparison of the experimental pump power dependence of recombination time in Ge:Ga (Ge:Ga, $N_a = 2 \times 10^{15}$ cm^{-3}, $Nd/Na < 0.01$, $T = 5$ K, $E_p = 0.5$ meV)[15] with the cascade capture time calculated for several compensation levels below 0.01, with charged acceptor concentrations from Fig. 7, τ_{rec,T_e} – with account of overlap of impurity potentials assuming a Maxwellian distribution of free carriers with $T_e = T$ (17); $\tau_{\mathcal{E}}(E_0)$ – using the model proposed in [17] for high concentration of capture centers.

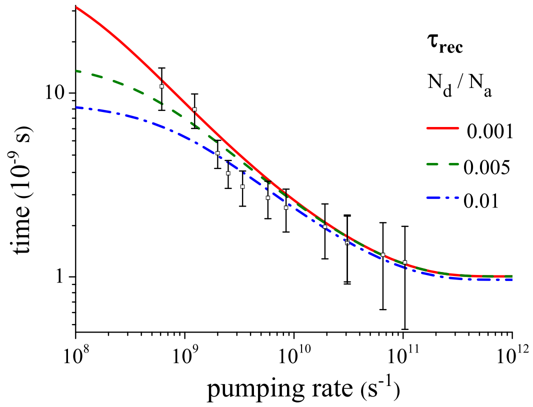

FIG. 10: (Color online) Recombination time: experimental results (Ge:Ga, $N_a = 2 \times 10^{15}$ cm^{-3}, $Nd/Na < 0.01$, $T = 5$ K, $E_p = 0.5$ meV) [15], and cascade capture time calculated with account of the overlap of impurity potentials and non-equilibrium distribution of photo-ionized carriers (16) with the concentration of impurity ions from Fig. 7.

However, the minimum life time obtained within our model is much longer than the recombination times (30 − 170 ps) measured in pump-probe experiments when the number of capture centers was above 10^{16} cm^{-3} [14]. Such increase of recombination rate may be due to optical phonon assisted recombination. This recombination channel is known to be dominant in p-Ge doped by deep impurities [28], and for shallow acceptors in the conditions of heating of charge carriers by strong electric field [29]. It has been mentioned that optical phonon assisted recombination may play a bigger role at high doping levels [30]. Indeed, optical phonon assisted transitions bring carriers directly to ground and first excited states with localization radii much smaller than that of the states essential for cascade capture to isolated impurity centers. This may lead to a slower dependence of the cross section of the optical phonon assisted recombination on the concentration of impurity ions than that of cascade capture.

IV. SUMMARY AND CONCLUSIONS

We analyzed the dependence of cascade recombination due to interaction with acoustic phonons on the doping level using the quasi-classical approach for the calculation of the relaxation of photo-excited carriers in the field of a set of impurity ions [22]. This model is an extension of the quasi-classical model of cascade capture to isolated Coulomb impurity centers proposed in [18]. Our model enables simultaneous calculation of non-equilibrium carrier distribution function formed by interaction with acoustic phonons in the presence of impurity traps, and the time of recombination in a wide range of capture center concentrations and temperatures. Additionally, we calculated the time of cascade recombination in the presence of fast scattering processes forming a Maxwellian distribution of free carriers. We show that the time of cascade capture in highly doped materials decreases when the energy of the source of photo-ionized carriers is close to the energy of potential barriers between the impurity centers, and in the presence of fast scattering processes that cause thermalization of the free carrier distribution.

We show that the concentration and temperature dependence of recombination time observed in highly doped semiconductors [15, 19] can be described within the model of uniformly spaced impurity ions. We explain the different regimes of cascade capture by the balance of the fluxes of charge carriers in the energy space: the relaxation flux due to spontaneous emission of acoustic phonons and the opposing thermal diffusion flux due to induced phonon assisted transitions. We show that this balance is determined by the ratio of the thermal energy T and the energy of the overlap of impurity potentials E_N, that determines the energy scale of the change of the density of states and the energy drift coefficient in the field of impurity centers. At low concentration of impurity ions when the $E_N/T << 1$, the fast change of the density of states within the thermal energy forms a bottle-neck for relaxing carriers, the opposing carrier fluxes have close values much larger than the resulting recombination flux. In this case a Maxwellian distribution of free carriers is formed, and the recombination time coincides with that of the model of isolated impurity centers [18]. At high doping concentrations $E_N/T >> 1$, the density of states in the field of impurity centers does not change much within the thermal energy, the thermal diffusion flux of carriers is much smaller than the relaxation flux due to spontaneous emission of acoustic phonons, and the fast relaxation forms a highly non-equilibrium carrier distribution. In this case the recombination time is determined by the energy relaxation time due to spontaneous phonon emission at the energy of the overlap of impurity potentials. Our result in the limit of high concentrations of impurity ions is close to that obtained in [17] within a qualitative model, based on the assumption of non-uniform distribution of capture centers. Contrary to that model, we show that the assumption of the non-uniform distribution of capture centers is not needed for explanation of the characteristic features of cascade capture in highly doped semiconductor. Moreover, we analyze the influence of non-uniformity of spatial distribution of impurity ions, and show that its effect on capture time in highly doped semiconductors is small.

V. ACKNOWLEDGEMENTS

This research has been funded by the joint RFBR-DFG project 18-502-12077, N.D. gratefully acknowledges support by the Helmholtz Research School on Security Technologies. We are grateful to P. Michel and the FELBE team for their dedicated support.

APPENDIX

Density of states in the field of a set of uniformly spaced Coulomb centers calculated using spherical approximation for integration in (4):

$$\tilde{\rho}(E) = \rho(E_N) F_\rho(E/E_N)$$

$$F_\rho(x) = \begin{cases} (3\pi/16)|x|^{-5/2} & x < -1 \\ \Phi_1(x) & -1 < x < 0 \\ \Phi_2(x) & x > 0 \end{cases}, \qquad \text{(A.1)}$$

with $\Phi_1(x) = \frac{3}{8x^{5/2}}\left(\phi(x) - \frac{\sin(4\phi(x))}{4} - \frac{\sin^3(2\phi(x))}{3}\right)$, $\Phi_2(x) = \frac{3}{8x^{5/2}}\left(\psi(x) - \frac{\sinh(4\psi(x))}{4} - \frac{\sinh^3(2\psi(x))}{3}\right)$, $\phi(x) = \arcsin(\sqrt{x})$ and $\psi(x) = \operatorname{arcsinh}(\sqrt{x})$.

The energy drift coefficient in the field of a set of uniformly spaced Coulomb centers calculated using the spherical approximation of the integral in (5):

$$\tilde{B}(E) = B(E_N)F_B(E/E_N)$$

$$F_B(x) = \begin{cases} 1/|x| & x < -1 \\ x^2 + 3x + 3 & x > -1 \end{cases} . \qquad \text{(A.2)}$$

Distribution of photo-ionized carriers in the field of a set of uniformly spaced Coulomb attractive centers at energies $E + E_1 >> T$ calculated using (6), (A.1) and (A.2):

$$f(E) = \frac{J}{B(E_N)} F\left(\frac{E}{E_N}, \frac{T}{E_N}, \frac{E_p}{E_N}\right)$$

$$F(x,y,z) = \begin{cases} y + |x| & x < -1 \\ \exp\left(\frac{-x}{y}\right)\Phi_3(x,y) & -1 < x < z \\ \exp\left(\frac{-x}{y}\right)\Phi_3(z,y) & x > z \end{cases} , \qquad \text{(A.3)}$$

with $\Phi_3(x,y) = \Psi_1(y) + \Psi_2(x,y)$, $\Psi_1(y) = \exp(-1/y)(1+y)$ and $\Psi_2(x,y) = \int_{-1}^{x} \exp(-u/y)/(u^2 + 3u + 3)du/y$.

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