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Cross section for recombination of an electron with a positively charged center in a semiconductor

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A formula for the total cross section for the recombination of a conduction electron with positively charged center in a semiconductor is derived under the assumption that the excess electron energy is transferred to acoustic phonons. The cross section is $2kT/ms^2$ times greater than that obtained in the widely cited paper by M. Lax [*Phys. Rev.* **119**, 1502 (1969)] and has a different power-law dependence on the temperature. In contrast to Lax's conclusion that capture of electrons with energies $\sim kT$ by highly excited levels of the center with a binding energy $U \sim kT$ is predominant in the recombination process, it is shown that the main contribution to the total recombination cross section is made by capture of electrons with an energy near ms^2 by levels with a binding energy close to ms^2 .

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INTRODUCTION

When a conduction electron is captured by a positively charged center, it should transfer a large energy (on the order of the binding energy) to the lattice. In many cases, the principal channel through which energy is lost is the interaction with acoustic phonons, wherein the electron loses in each collision only a small fraction of its kinetic energy $\sim \epsilon (8ms^2/\epsilon)^{1/2}$ (ϵ is the electron kinetic energy, m is its effective mass, and s is the speed of sound in the crystal). In a well known paper,

Lax^[1] proposed a "cascade capture" model: the electron is captured by one of the highly-excited levels of the recombination center, which are quasi-continuously arranged, and "rolls down" over them, emitting acoustic phonons. For the case of a gas-discharge plasma, a similar problem concerning the capture of an electron by a positive ion in the presence of neutral atoms was solved by Pitaevskii by another method.^[2] We have adapted Pitaevskii's procedure to the case of recombination of a conduction electron in a semiconductor by a charged center, with emission of acoustic phonons, and

obtained for the total recombination cross section the expression

$$\sigma = 2^6 \sigma_1 \left(\frac{ms^2}{2kT} \right)^3, \quad \sigma_1 = \frac{1}{3} \left(\frac{e^2 Z}{\kappa ms^2} \right)^3 \frac{m^3 E_c^2}{\rho \hbar^4}, \quad (1)$$

where T is the temperature of the electrons which are in thermal equilibrium with the lattice, E_c is the deformation-potential constant, eZ is the charge of the recombination center, κ is the dielectric constant, and ρ is the crystal density. The cross section obtained by us exceeds the one obtained by Lax by a factor $2kT/ms^2$ (which amounts to two orders of magnitude at $T \sim 10^\circ \text{K}$), and depends on the temperature in accordance with a different power law. Both procedures are based on the idea of being able to describe the recombination of an electron that has landed in a bound state as diffusion in space of negative total energy, so one might think that the two results should coincide. In view of this discrepancy, we have analyzed Lax's paper and found an error in his calculations, due to an incorrect averaging of the transition probability in the collisions of the electron with acoustic phonons on the electron orbit in the field of the center. It follows from Lax's work that the principal role in recombination is played by captures of electrons of energy $\sim kT$ by levels with binding energy $\sim kT$, whereas correct averaging shows that the principal role is played by captures of electrons of energy $\sim ms^2$ by recombination-center levels with binding energy $U \sim ms^2$. After correcting this averaging error, the total recombination cross sections calculated by Lax's and Pitaevskii's methods are in agreement.

1. CALCULATION OF THE TOTAL RECOMBINATION CROSS SECTION BY PITAEVSKII'S METHOD

We consider the kinetic equation for the distribution function f of an electron in the field of a positively charged center with potential $U(r)$

$$\frac{\partial f}{\partial t} + v \nabla_r f + e \frac{\partial U(r)}{\partial r} \nabla_r f = \left(\frac{\partial f}{\partial t} \right)_{\text{coll}}, \quad U(r) = \frac{eZ}{\kappa r}. \quad (2)$$

We take into account in the collision integral only the interaction of the electrons with the acoustic phonons. Recognizing that the kinetic energy of the electron changes little in each collision, we represent, as is customary, the collision integral in differential form

$$\left(\frac{\partial f}{\partial t} \right)_{\text{coll}} = -[\rho(\epsilon)]^{-1} \frac{\partial}{\partial \epsilon} \epsilon \rho(\epsilon) \frac{2ms^2}{kT} v_a(\epsilon) \left[f + kT \frac{\partial f}{\partial \epsilon} \right], \quad (3)$$

where $\rho(\epsilon) d\epsilon$ is the number of states in the kinetic-energy interval $d\epsilon$:

$$\rho(\epsilon) = \frac{8\sqrt{2} \pi m^3 V}{(2\pi \hbar)^3} \epsilon^{3/2} \quad (4)$$

(here V is the volume of the crystal), and $v_a(\epsilon)$ is the frequency of the elastic collisions of the electron with the acoustic phonons:

$$v_a(\epsilon) = \frac{2^{1/2} E_c^2 m^3 kT}{\pi \hbar^4 \rho s^2} \epsilon^{1/2} \quad (5)$$

(the notation is the same as in formula (1)).

We assume that the distribution function of an electron situated in the field of the center depends only on the total energy E :

$$f = f(E), \quad E = \frac{p^2}{2m} - \frac{e^2 Z}{\kappa r}. \quad (6)$$

Averaging the kinetic equation with the collision integral (3) in the phase-space layer between the hypersurfaces $E = \text{const}$ and $E + dE = \text{const}$, we obtain

$$\rho(E) \frac{\partial f(E)}{\partial t} = - \frac{\partial}{\partial E} j(E),$$

$$j(E) = B(E) \left[f(E) + kT \frac{\partial f(E)}{\partial E} \right], \quad (7)$$

where $\rho(E) dE$ is the number of states in the total-energy interval dE , and $B(E)/\rho(E)$ has the meaning of the coefficient of dynamic friction in energy space:

$$\rho(E) = \int_0^{r_{\text{max}}} \frac{\rho(\epsilon)}{V} 4\pi r^2 dr, \quad (8)$$

$$B(E) = \int_0^{r_{\text{max}}} \frac{\epsilon \rho(\epsilon) v_a(\epsilon)}{V} \frac{8ms^2}{kT} \pi r^2 dr.$$

We solve Eq. (7) under the stationary conditions ($\partial f / \partial t = 0$) in the region of negative total energy ($E < 0$), where

$$\epsilon = e^2 Z / \kappa r - |E|, \quad r_{\text{max}} = e^2 Z / \kappa |E|. \quad (9)$$

Equation (7) then goes over into

$$B(E) \left[f(E) + kT \frac{\partial f(E)}{\partial E} \right] = j, \quad B(E) = \frac{16}{3\pi^2} \frac{m^4 E_c^2 (e^2 Z)^3}{\kappa^3 \hbar^7 \rho |E|} \quad (10)$$

with the boundary condition

$$f(E) \rightarrow 0 \quad (E \rightarrow -E_1), \quad E_1 \gg kT. \quad (11)$$

The constant j , which has the meaning of the constant flux in a region of negative total energy, will be determined from the condition that the distribution function tends to a Boltzmann function as $E \rightarrow 0$. These conditions are discussed in Pitaevskii's paper,^[2] where it is shown that the final result does not depend on the value of E_1 and in the expression for the flux j we can replace $-E_1$ by $-\infty$. Following the Pitaevskii procedure, we solve Eq. (10) and obtain the flux j (with the distribution function renormalized to one particle in the volume):

$$j = \frac{2^{1/2} E_c^2 (e^2 Z)^3 m^{3/2}}{3\sqrt{\pi} \kappa^3 \hbar^4 \rho V (kT)^{3/2}} \quad (12)$$

If this flux is divided by the concentration ($n = 1/V$) and the average electron velocity is calculated with a Boltzmann distribution function ($\langle v \rangle = (8kT/\pi m)^{1/2}$), then we obtain the total recombination cross section σ given in formula (1).

In concluding this section we recall once more the physical assumptions on which our deduction is based. First, the distribution function depends only on the total

energy; second, in collisions between the electron and the acoustic phonons, a small fraction of the kinetic energy of the electron is transferred to the phonons; third, the upper excited levels of the charged center are quasi-continuous up to energies $\sim -kT$.

The first assumption is valid if

$$r \ll l, \quad r \approx e^2 Z / \chi kT, \quad (13)$$

where r is the characteristic dimension of the orbit of an electron with binding energy $\sim kT$ and l is the electron mean free path. The second assumption is reasonable at all energies except $E \sim ms^2$. The third assumption can be naturally made if the distance between neighboring levels in the noted energy interval $\Delta \varepsilon$ is much less than the characteristic energy $\Delta \varepsilon_{\text{phon}}$ of the emitted phonons. Using a hydrogen-like spectrum, we obtain the inequality

$$\frac{\Delta \varepsilon}{\Delta \varepsilon_{\text{phon}}} \approx \frac{kT}{(2ms^2 e_b)^{1/2}} < 1, \quad \varepsilon_b = \frac{e^4 m}{2\chi^2 \hbar^2}, \quad (14)$$

satisfaction of which makes the third assumption valid.

2. CALCULATION OF THE TOTAL RECOMBINATION CROSS SECTION BY LAX'S METHOD

In Lax's model, the process of electron capture by a center is divided into two stages. First, the electron moving past the center emits a phonon as a result of a single collision, and ends up in a bound state on one of the highly-excited levels. Next, emitting and absorbing phonons, the electron changes its total energy and either goes into the region of positive energies and moves away from the center, or else "collapses" to the ground state and "sticks" to the center. Both in the capture by a highly-excited level and in the subsequent diffusion in energy space, the electron is regarded as a classical particle.

For a quantitative description of the behavior of the electron in a bound state with binding energy U , Lax introduced a sticking function $P(U)$, which is defined as the probability that the captured electron will not go off to the region of positive energies. The sticking function is close to unity when U exceeds kT .

Lax's results for the total recombination cross section can be obtained from the following simple considerations. Assume that initially the electron with kinetic energy E_0 is far (at $+\infty$) from the charged center. In a positive-charge field it is accelerated and acquires at the distance r_0 a large kinetic energy:

$$\varepsilon = E_0 + e^2 Z / \chi r_0, \quad (15)$$

retaining a total energy equal to E_0 . The energy of the phonon emitted by the electron amounts to a small fraction of its kinetic energy:

$$\hbar\omega \approx 2(2ms^2 \varepsilon)^{1/2}. \quad (16)$$

In order for the electron to be able to go over into a state with negative total energy $E = -U$ as a result of a

single collision, it should emit a phonon with energy

$$\hbar\omega = E_0 + U. \quad (17)$$

Naturally, in this case it is necessary to have a large kinetic energy, i.e., it suffices to come close to the charged center. The maximum distance r_0 from the recombination center, at which the electron can go over from a state with positive energy E_0 as a result of emission of one phonon into a bound state with binding energy U can be easily obtained from relations (15)–(17):

$$r_0 \approx 8ms^2 e^2 Z / \chi [E_0 + U]^2. \quad (18)$$

The cross section of this process is determined by the formula

$$\sigma(E_0, U) \approx \pi b^2 r_0 / l, \quad (19)$$

where b is the impact distance for the orbit whose shortest distance to the attraction center is equal to r_0 :

$$b = r_0 [(E_0 + e^2 Z / \chi r_0) E_0^{-1}]^{1/2}. \quad (20)$$

The ratio r_0 / l , where l is the mean free path of the electron in the case of collisions with acoustic phonons, describes the probability of the emission of an acoustic phonon by an electron over a length r_0 . Ultimately we have

$$\sigma(E_0, U) = 2^2 \pi \left[\frac{e^2 Z}{\chi (E_0 + U)} \right]^3 \left(\frac{ms^2}{E_0 + U} \right)^3 \left(\frac{E_0 + U}{2(2ms^2 E_0)^{1/2}} \right)^2 \frac{1}{l}, \quad l = \frac{\pi \rho s^2 \hbar^4}{E^2 m^2 kT} \quad (21)$$

(the notation is defined after formula (1)). Lax's result for the total recombination cross section can be obtained with the aid of (21) if it is assumed that the main contribution to the cross section is made by electrons of energy $E_0 \sim kT$ when they are captured by a binding level with $U \sim kT$, the sticking function on which is on the order of unity:

$$\sigma \approx 2^2 \left(\frac{e^2 Z}{\chi ms^2} \right)^3 \left(\frac{ms^2}{2kT} \right)^4 \frac{E_c^2 m^2}{\rho \hbar^4} \approx \frac{4^3}{6} \left(\frac{ms^2}{2kT} \right)^4 \sigma_1. \quad (22)$$

By a rigorous analysis, Lax obtains for the total recombination cross section the formula

$$\sigma = \frac{\sigma_1}{\gamma^2} \int_0^\infty \frac{dP(\eta)}{d\eta} d\eta \int_\eta^\infty d\eta' \int_0^\infty \frac{e^{-\nu/2(\xi+\eta')^2} d\xi}{\{[1+(\xi+\eta')/4]^2 - \xi\}^2 \{1 - \exp[-(\xi+\eta')/\gamma]\}} \quad (23)$$

$$\gamma = 2kT/ms^2, \quad \eta = 2U/ms^2, \quad \xi = 2E_0/ms^2.$$

Substituting in this formula the expression $dP(\eta)/d\eta$ (the calculation of which will be discussed later on), it arrives at a cross section that differs from (22) by a factor on the order of unity (see Eq. (3.13) of [1]). This mistaken result was obtained by Lax from the correct formula (23) because he used an incorrect expression for $dP(\eta)/d\eta$.

We proceed to construct the sticking function. Accepting Lax's assumption that $P(U)$ depends only on the binding energy U and does not depend on how the electron has fallen into a state with such a binding energy, we obtain for the sticking function the equation

$$P(\eta) = \int_{\eta+\mu=0} K(\eta, \mu) P(\eta+\mu) d\mu, \quad \mu = \frac{\pm 2\hbar\omega}{ms^2}, \quad \eta = \frac{2U}{ms^2}, \quad (24)$$

where $K(\eta, \mu)$ is the probability of the transition from the binding level η to the binding level $\eta + \mu$ ($\mu > 0$ when phonons are emitted, $\mu < 0$ in the case of phonon absorption). The kernel $K(\eta, \mu)$ is chosen by Lax to be the standard expression for the transition probability of an electron of fixed kinetic energy colliding with an acoustic phonon. The kinetic energy of the electron on the binding orbit is assumed by Lax to be constant and equal to its mean value, i. e., $\epsilon = U$ (in accordance with the virial theorem). On the other hand, in the case of the rigorous approach the transition probability must be averaged over a microcanonical distribution. The mathematical error in Lax's calculations lies in the fact that in place of $F(\epsilon)$, the mean value of a function of the kinetic energy—he uses $F(\langle\epsilon\rangle)$, i. e., a function of the mean value of the kinetic energy. This leads to an incorrect result. The error in the average manifests itself particularly strongly in the region of low binding energies. Thus, according to Lax, if the electron is on a binding level with $U < ms^2$, it cannot emit an acoustic phonon (accordingly $P(U) = 0$ at $U < ms^2$). Actually, however, there are regions of phase space in which the kinetic energy is $\epsilon > ms^2$ even at these values of the total energy, and consequently the electrons can still emit acoustic phonons (and accordingly $P(U) \neq 0$, albeit small, at $U < ms^2$). It turns out that the contribution of this energy region to the total cross section is significant, whereas Lax simply sets it equal to zero.

In the region of large η ($\eta \gg 1$), Lax goes over from the integral equation (24) to a differential equation of the Fokker-Planck type

$$P''(\eta)/P'(\eta) = -2\langle\mu\rangle/\langle\mu^2\rangle, \quad (25)$$

$$\langle\mu^2\rangle = \int_{\eta+\mu=0} K(\eta, \mu) \mu^2 d\mu. \quad (26)$$

The boundary conditions follow from the meaning of the sticking function:

$$P(0) = 0, \quad P(\eta) \rightarrow 1 \quad (\eta \rightarrow +\infty). \quad (27)$$

The first condition means that only bound electrons "stick," and the second means that the electrons that land on levels with binding energy $U \gg kT$ stick completely. Solving (25) and (27), and using the moments (26) calculated with his kernel, Lax obtained

$$dP(\eta)/d\eta \approx \eta^2 e^{-\eta/\gamma} / 2\gamma^2. \quad (28)$$

Substituting (28) in (23) he verifies that the main contribution to the total recombination cross section is made by allowance for the capture of an electron with energy $E_0 \sim kT$ on binding levels $U \sim kT$. This way Lax justifies the replacement of the integral equation by a differential equation (inasmuch as the condition $\eta \sim \gamma \gg 1$ is satisfied at $U \sim kT$). Integrating, he ultimately arrives at a formula for the total recombination cross section in the form (22) which contains, apart from an insignificant numerical factor of the order of unity, a

superfluous factor ($ms^2/2kT$) in comparison with the formula (1) obtained by us.

The correct expression for the kernel $K(\eta, \mu)$ is the following:

$$K(\eta, \mu) d\mu = [\tau(\eta, \mu)]^{-1} d\mu / [\tau(\eta)]^{-1}, \quad (29)$$

where $[\tau(\eta)]^{-1}$ is the total number of transitions per unit time from the binding level η as a result of interaction with acoustic phonons, averaged over the microcanonical distribution:

$$f(p, r) \sim \delta(U + p^2/2m - e^2 Z/\kappa r),$$

and $[\tau(\eta, \mu)]^{-1} d\mu$ is the number of transitions from the level upon emission ($\mu > 0$) or absorption ($\mu < 0$) of phonons with energy from the internal ($\mu, \mu + d\mu$), averaged with the same function $f(p, r)$. Using the standard expressions for the transition probability we obtain, in contrast to Lax, the kernel in the form

$$K(\eta, \mu) = \frac{|\mu|}{[(4+\mu)^2 + 16\eta]^2 [1 - e^{-\mu/\gamma}] N(\eta)}, \quad -\eta \leq \mu \leq \infty, \quad (30)$$

$$N(\eta) = \int_{-\infty}^{+\infty} \frac{|\mu| d\mu}{[(4+\mu)^2 + 16\eta]^2 (1 - e^{-\mu/\gamma})}.$$

If we now solve (25) and (27), calculating the moments (26) with the kernel (30), then we get

$$dP(\eta)/d\eta \approx \eta e^{-\eta/\gamma} / \gamma^2, \quad P(\eta) = 1 - (1 + \eta/\gamma) e^{-\eta/\gamma}. \quad (31)$$

Substituting this expression in (23), we verify that now the main contribution to the total recombination cross section is made by allowance for the capture of electrons with energy $E_0 \sim ms^2$ on binding levels with $U \sim ms^2$. Integrating, we obtain a total cross section $4\sigma/3$, where σ is given by (1). This result can be qualitatively obtained as a consequence of formula (21) with the aid of simple arguments, similar to those that have led to formula (22), if it is assumed that the main contribution to the total cross section is made by capture of an electron with energy $\sim ms^2$ on a binding level with $U \sim ms^2$, the sticking function for which is determined by (31). The fact that the cross sections calculated by the methods of Pitaevskii and Lax do not agree, is caused by the incorrect replacement of the integral equation for the sticking function by a differential equation in the region of small η , which makes the principal contribution to the cross section. The first correct integral equation for the sticking function $P(\eta)$ is the region of small η is given in^[3], where this equation was solved numerically and where plots were constructed for $P(\eta)$ at $\gamma = 2, 10, \text{ and } 50$, and for the recombination cross section σ at a temperature $3.5^\circ \text{K} < T < 10^\circ \text{K}$. The numerical values of the recombination cross section, calculated in accordance with our formula (1), lie exactly on the curve obtained by computer calculation in^[3]. It should be noted that no analytic expressions were obtained in that reference at all, for either the sticking function or the total recombination cross section.

CONCLUSION

It is shown in this paper that the total cross section for the capture of an electron by a positive center, with transfer of the excess energy to acoustic phonons, under conditions when the quasi-classical analysis is valid, is described by formula (1). It differs from Lax's well known expression for the cross section in the fact that it is larger than the latter by a factor $2kT/ms^2$ and is inversely proportional to the third power of the temperature and not to the fourth power as in Lax's formula.

We have shown that the total recombination cross section is much easier to calculate by Pitaevskii's method than by Lax's method (supplemented by the requirement that the probability of the electronic transitions be correctly averaged). However, Lax's method has the advantage that it makes it easy to obtain the cross section for capture of an electron with a fixed energy E_0 by a recombination center, if the sticking function is known

$$\begin{aligned} \sigma(\xi) &= \int_0^{\infty} \sigma(\xi, \eta) P(\eta) d\eta \\ &= \frac{\sigma_1}{\xi} \int_0^{\infty} \frac{P(\eta) (\xi + \eta)^2 d\eta}{\{[1 + (\xi + \eta)/4]^2 - \xi^2\} \{1 - \exp[-(\xi + \eta)/\gamma]\}}, \end{aligned} \quad (32)$$

where ξ , η and γ are defined in (23). For $\xi \gg 1$ the main contribution to the cross section $\sigma(\xi)$ is made by captures on levels with dimensionless binding energy $\eta \sim \xi \gg 1$, so that expression (31) can be used for the sticking function and we get

$$\begin{aligned} \sigma(\xi) &= \frac{2}{3} \frac{4^4 \sigma_1}{\xi^3 \gamma}, \quad 1 \ll \xi \ll \gamma, \\ \sigma(\xi) &= \frac{4^4}{3} \frac{\sigma_1}{\xi^4}, \quad \xi \gg \gamma. \end{aligned} \quad (33)$$

It is seen from this formula that the differential capture cross section increases rapidly with decreasing ξ , and this explains the decisive role played by capture of electrons with low energies in the recombination process.

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New effect in electron-nuclear double resonance with distant nuclei

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Anomalously intense ENDOR signals due to polarization of the lattice nuclei ("Larmor" ENDOR) are observed in LiF crystals with relatively low F-center concentrations ($n \sim 2 \times 10^{17} \text{ cm}^{-3}$) at the Larmor frequency of the lattice nuclei. The dependences of the ENDOR signal intensity on the microwave field strength and on the mismatch ($H - H_0$) (H_0 is the magnetic field strength corresponding to the center of the ESR line) are studied at various temperatures and concentrations of the paramagnetic centers. It is found that the dynamic behavior of the Larmor ENDOR is the opposite of that of the "distant" ENDOR described in the literature, in which signals at the Larmor frequency of the lattice nuclei are also observed. Larmor ENDOR is observed in samples with relatively low concentrations of paramagnetic centers and is maximal at the center of the ESR line. The ENDOR mechanism is not the same for distant and near nuclei. It is shown that lattice nuclear polarization required for the observation of Larmor ENDOR is not connected with dipole-dipole pool effects and can be ascribed to relaxation processes or to saturation of forbidden microwave transitions. It is important that Larmor ENDOR is observed in the presence of regions with nonequilibrium nuclear polarization in the sample even if there is no net polarization. It is found that the existence of Larmor ENDOR may be regarded as experimental proof that the lattice nuclei relax via a paramagnetic impurity.

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The study of the dynamic laws and mechanisms governing the production of electron-nuclear double resonance (ENDOR) signals yields not only detailed information of the physical processes occurring in bound electron-nuclear systems, but also contributes to a wider and more successful application of this method. The

most investigated ENDOR mechanisms are the mechanism connected with the effective decrease $T_1 - T_1^{\text{eff}}$ of the time of the spin-lattice relaxation by a radio-frequency (RF) field,^[1,2] and the method of "distant" ENDOR.^[2-4] Mechanisms of "negative" ENDOR,^[5,6] ENDOR due to shift of the ESR line,^[7] and due to re-