ELECTRIC FIELD MECHANISM OF RELAXATION OF PARAMAGNETIC CENTERS INTERACTING WITH CARRIERS

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A new mechanism is proposed for spin relaxation of paramagnetic impurity centers in semiconductors. This mechanism is due to the possibility of electric-dipole excitation of the spin transitions of the impurities located in sites of a crystal without an inversion center. A comparison is made with the known mechanisms of spin relaxation due to interaction of the paramagnetic centers with the carriers.

The question of the role of the carriers in paramagnetic relaxation was first considered theoretically in [1, 2]. It turned out here that the mechanism of the interaction of the conduction electrons with the paramagnetic center (PC), proposed in [2], lead to too large relaxation times and are not realistic. The exchange-scattering mechanism [1] has made it possible to explain in a number of cases the available experimental data, but generally speaking its effectiveness is limited by the phenomenon of polarization of the band electrons, which sets in at high PC concentrations and at large spin-lattice relaxation times of the conduction electrons.

In crystals in which the local symmetry of the PC has no inversion center, the spin-Hamiltonian contains terms linear in the electric field (the electric-field effect [3]). In this case the interaction of the PC with the electric field of the carriers can also give rise to paramagnetic relaxation. The possible effectiveness of such a mechanism is evidenced by the correlation, noted in [4, 5], between the relaxation times of a number of PC and the values of their electric-field constants. It was assumed in these references that the paramagnetic relaxation is due to the interaction of the PC with the fluctuating electric field of the carrier plasma. Such an analysis corresponds to allowance for only the electron-gas field-fluctuation components with the extremely long wavelengths.

In crystals with small amounts of PC (absence of impurity conductivity), the spin-plasmon relaxation mechanism takes place in a limited range of electron concentrations, when $\omega_{ab} \sim \Omega$ or $\omega_{ab} \ll \Omega$ for one- and two-plasmon processes, respectively (ω_{ab} is the transition frequency and Ω is the Langmuir frequency). The indicated limitation does not appear when account is taken of the short-wave components of the electric-field fluctuations, which are connected with motion of electrons that do not interact with one another. At the same time, owing to the direct coupling of the PC spin with the translational motion of the band electrons, the polarization of the latter, which is inherent in the mechanism of [1], does not take place in this case. The present communication is devoted to these questions.

The Hamiltonian of the interaction of a PC with the electric field of the carriers is given by

$$V = \sum_{ijk} [\beta_{ijk} H_j \hat{S}_k + \alpha_{ijk} \{\hat{S}_j \hat{S}_k\}] \hat{E}_i \equiv \sum_i \hat{D}_i \hat{E}_i \,. \tag{1}$$

Here α_{ijk} and β_{ijk} are the components of the tensors of the electric-field effect, $\vec{H}$ is the external magnetic field, $\hat{\vec{E}}$ is the operator of the electric field produced by the conduction electrons at the location of the PC; $\{S_jS_k\} = S_jS_k + S_kS_j$.

The probability W_{ab} of the transition of the PC from the state a to the state b is equal to

$$W_{ab} = \frac{1}{\hbar^2} \sum_{ij} (D_i)_{ab} (D_j)^*_{ab} \frac{1}{(2\pi)^3} \int dk \, \langle E_i E_j \rangle_{k\omega_{ab}} \tag{2}$$

$\langle E_iE_j\rangle k\omega_{ab}$ is the space-time Fourier transform of the correlator of the electron-gas electric field [6]. In the considered case of large k (ka > 1, a is the Debye radius), in the absence of degeneracy and at $k_BT \gg \hbar\omega_{ab}$ we can represent $\langle E_iE_j\rangle k\omega_{ab}$ in the form

$$\langle E_i E_j \rangle^0_{k\omega_{ab}} = 16\sqrt{6}\pi^{3/2} \frac{e^2 n}{\epsilon^2 s} \frac{k_i k_j}{k^5} e^{-\frac{3\omega_{ab}^2}{2k^2 s^2} - \frac{\hbar^2 k^2}{8 m^* k_B T}}. \quad (3)$$

$\langle E_i E_j \rangle^0_{k\omega}$ is the Fourier transform of the correlator of an ideal electron gas. Equation (3) does not vanish only if $a^2 > 8m^* k_B T/\hbar^2$, which is satisfied if $\omega_{ab} \sim \Omega$. In this case the transition probability is expressed in terms of the MacDonald function $K_0(\hbar\omega_{ab}/2k_BT)$ and at $\hbar\omega_{ab} \ll k_BT$ it is approximately equal to

$$W_{ab} = 8\left(\frac{2\pi}{3}\right)^{1/2} \frac{e^2 n}{\hbar^2 \epsilon^2 s} |D_{ab}|^2 \ln \frac{2{,}2 k_B T}{\hbar\omega_{ab}}. \quad (4)$$

n is the concentration of the conduction electrons, m* is their effective mass, ε is the dielectric constant of the crystal, and s is the thermal velocity of the electrons.

For a bipolar se-iconductor it is obvious that

$$W_{ab} = W_{ab}^{(n)} + W_{ab}^{(p)}$$

$W_{ab}^{(n)}$ and $W_{ab}^{(p)}$ are the probabilities of the relaxation transitions when the PC interacts with electrons and holes, respectively.

The weak dependence of $W_{ab}/|\vec{D}_{ab}|^2$ on the value of the magnetic field is connected with the nonresonant transfer of the Zeeman energy to the band electron during the course of its scattering by the PC. However, when the magnetic field is turned off, one can expect an appreciable decrease of the relaxation time[1] for centers with S > 1/2. Unlike the phonon Raman scattering process, the temperature dependence of the transition probability is proportional here to $T^{-1/2}$ when $n \neq n(T)$. On the other hand, if the appearance of the electrons in the band is connected with thermal influx of electrons, then a rapid (nearly exponential) dependence of the transition probability on the temperature is observed.

Comparison of (4) with the corresponding expressions in [1, 4] shows that the relaxation mechanism considered here is more effective than the one-plasmon mechanism and is no less effective (at electric-field constant values typical of deep donors) than the exchange-scattering mechanism. Consequently, formula (4) can serve as an alternative for an explanation of the experimental relaxation of deep donors (e.g., those cited in [4]).

At large carrier densities ($\omega_{ab} \ll \Omega$), the short-wave Fourier components of the electric field $\langle E_i E_j \rangle^0_{k\omega_{ab}}$ become negligibly small. It was assumed in [5] that in this range of concentration the transition probability is determined by two-plasmon processes and in a higher order of perturbation theory. Estimates, however, show that in the screened nonresonant interaction of the electrons with the PC (connected with Fourier components $\langle E_i E_j \rangle_{k\omega_{ab}}$, for which $1/a \gtrsim k > \omega_{ab}/s$) is much narrower in the lowest order of perturbation theory, and its contribution to W_{ab} is comparable with the contribution of the two-plasmon processes.

In [7], we have experimentally investigated the dependence of the time of paramagnetic relaxation of phosphorus and silicon on the concentration of the conduction electrons, which result from optical illumination. The concentrations of the band electrons did not exceed 10^8 cm^{-3}. The spin-plasmon mechanism does not operate in this case, since $\omega_{ab} \gg \Omega$. The mechanism considered here, to the contrary, is effective also at such carrier densities. Unfortunately, a quantitative comparison with experiment is still impossible, since the values of the electric-field components are not known for these impurities.

[1] At $\vec{H} = 0$ we have $\omega_{ab} \neq 0$, so that the spin Hamiltonian contains crystal-field terms.

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CONTRIBUTION TO THE THEORY OF BAND FERROMAGNETISM

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A model is investigated with a single-electron spectrum that is unstable against electron-hole pairing. Conditions are found under which the singlet and triplet electron-hole pairing can coexist and lead to a ferromagnetic state of collectivized conduction electrons.

It is known that in the approximation of a simple parabolic electron band $\varepsilon(\vec{p}) = p^2/2m$ the ferromagnetic state resulting from Coulomb interaction between conduction electrons is possible only if the interaction is strong enough [1, 2].

The magnetization of the electron gas, say in the x direction, can be expressed in the form

$$M_x \sim -i \sum_\sigma \int \frac{d^3q}{(2\pi)^3} G_{-\sigma,\sigma}(q, -0), \tag{1}$$

where

$$G_{-\sigma,\sigma} = -i \langle Ta_{-\sigma}(t) a_\sigma^+(0) \rangle .$$

We shall discuss below substances having a one-electron spectrum with singular properties, namely either semimetals with almost coinciding electron and hole Fermi surfaces [3], or semiconductors with a forbidden band much smaller than the exciton binding energy [4], or else metals with narrow allowed bands, when the condition $\varepsilon(\vec{p}) = -\varepsilon(\vec{p} + \vec{q})$ is approximately satisfied [5].

We shall show that a transition to the ferromagnetic semiconducting state is possible for such systems even in the case of weak interaction. The ferromagnetism is brought about here by the collectivized electrons of the conduction band of the doped semiconductor.

In all the listed cases, at an arbitrarily weak interelectron in-eraction, the system is unstable against electron-hole pairing in either the singlet state, characterized by an anomalous Green's function $G^{21}_{\sigma\sigma} = -i\langle Ta_{2\sigma} a^+_{1\sigma}\rangle$ and a binding potential V_C, or in a triplet state characterized by an anomalous function $G^{21}_{-\sigma\sigma} = -i\langle Ta_{2-\sigma} a^+_{1\sigma}\rangle$ with a potential V_T (the subscripts 1 and 2 characterize here either the numbers of the bands [3, 4] or two states that differ by the vector $\vec{q}$).

It will be shown below that singlet and triplet electron-hole pairings can exist in the case of a doped semimetal [3] or semiconductor [4], or else in the case of a metal in which the number of electrons per unit cell differs slightly from unity. But in this case the functions $G^{11}_{-\sigma\sigma}$ and $G^{22}_{-\sigma\sigma}$, which characterize the magnetization in accordance with (1), also turn out to be different from zero at arbitrarily weak interaction, as can be easily seen from the following formal equation: