Ferron-polaron states of carriers in antiferromagnetic semiconductors

V. D. Lakhno and E. L. Nagaev
(Submitted March 4, 1976; resubmitted July 2, 1976)
Fiz. Tverd Tela (Leningrad) 18, 3429-3432 (November 1976)

It is shown that ferron states in antiferromagnetic semiconductors are stabilized by the electron-induced polarization of the ionic crystal lattice. An electron captured by a ferromagnetic microregion polarizes the lattice much more strongly than a free electron and the process reduces the electron energy. A calculation is reported of the maximum Neel temperature at which a ferron is still stable at $\boldsymbol{T} = 0$. It is found that in the case of europium chalcogenides this temperature is twice as high as in the absence of such polarization. The ferron dissociation temperature also increases by a factor of 2.
PACS numbers: 75.50.Ee, 71.85.Ce

Nagaev [1,2] proposed a special conduction-electron state in a semiconductor crystal: An electron creates a micro-region of a new phase, which is normally unstable, and stabilizes this phase by becoming localized inside it. The condition for the existence of such a quasiparticle is a lower position of the bottom of the conduction band in the unstable phase than in the stable phase. The formation of such quasiparticles can be expected particularly in anti-ferromagnetic semiconductors in which a conduction electron is self-localized in a ferromagnetic microregion (ferron states). This prediction has been confirmed experimentally for antiferromagnetic semiconductors EuSe and EuTe (ref. 3). The results indicated that ferrons are more stable than expected on the basis of the initial estimate [1, 2].

We shall show that the ferron stability increases considerably as a result of polarization of the ionic lattice of a crystal by these quasiparticles. The constant α representing the coupling of electrons to optical phonons in ionic crystals is small (~ 2-3), i.e., the interaction can be described by the perturbation theory (as shown in ref. 4, where this theory applies up to $\alpha < 6$). Thus, a free electron polarizes weakly the lattice and the polaron shift of its energy $\alpha\omega$ (ω is the optical phonon frequency) is small (~ 0.01 eV). However, an electron captured by a ferromagnetic microregion polarizes the lattice much more strongly: The polaron shift is of the order of $\boldsymbol{e}^2/\boldsymbol{e}^*\mathbf{R}$, where **R** is the radius of the ferromagnetic microregion, e is the electron charge, $1/\varepsilon^* = 1/\varepsilon_\infty - 1/\varepsilon_0$, ε_0 and ε_∞ are the static and high-frequency permittivities of the crystal. Under typical conditions this quantity is 0.2-0.3 eV, i.e., it is close to the difference between the minimum energies of an electron in the ferromagnetic and antiferromagnetic states. Thus, a state of this kind represents essentially a mixed ferron-polaron state.

We shall consider a ferron in a polarizable crystal with a conduction band whose width W is large compared with the **s-f** exchange energy $\mathbf{As}/2$. We shall use a method similar to that employed by Pekar in the case of a tight-binding polaron.[5] The functional of the total energy of the

$$F = \frac{\hbar^2}{2m}\int (\nabla\psi)^2 d^3r - \frac{As}{2}\int_0^R \psi^2 d^3r - \frac{1}{2}\int \mathrm{pD}d^3r + E_d\,, \tag{1}$$

where $\boldsymbol{\psi}$ is the wave function of an electron; Ed is the formation energy of a ferromagnetic microregion; **p** is the polarization; **D** is the induction vector; ***m*** is the effective mass of a band electron. The energy of the polarization of the lattice by an electron $\boldsymbol{E_p}$ is

$$E_p = -\frac{1}{2}\int \mathrm{pD}d^3r\,. \tag{2}$$

The minimum of the functional **F** gives the total energy of the system. We shall solve the problem by applying the variational principle, in which it is assumed that a ferromagnetic order is established inside a sphere of radius ***R***. The radius ***R*** is the variational parameter. A test electron wave function $\boldsymbol{\psi}$ describes the ground state of the electron in a spherically symmetric well of radius ***R*** and of constant depth $\boldsymbol{As}/2$.

Thus, the electron wave function is selected in the form

$$\left.\begin{aligned} \psi &= c_1 \frac{\sin kr}{r}, \qquad k = \sqrt{2m\left(\frac{As}{2} - |E_e|\right)}, \qquad r < R, \\ \psi &= c_2 \frac{e^{-\aleph r}}{r}, \qquad \aleph = \sqrt{2m|E_e|}, \qquad r > R \end{aligned}\right\} \tag{3}$$

where $\boldsymbol{E_e}$ is the electron energy in the well: c_1 and c_2 are the constants found from the matching condition at the boundary (at $\mathbf{r} = \boldsymbol{R}$) and from the normalization of wave functions. It follows from Eqs. (2) and (3) that in the case of a deep level in a well, $\aleph \boldsymbol{R} >> 1$, the energy of the polarization of the lattice by an electron has the absolute value

$$|E_p| \approx 0.9 \frac{e^2}{\varepsilon^* R}. \tag{4}$$

Since $|\boldsymbol{E_p}|$ increases with decreasing $\boldsymbol{R}$, inclusion of $\boldsymbol{E_p}$ in Eq. (1) reduces the radius $\boldsymbol{R}$ at which the total energy of the system has its minimum value, i.e., the lattice polarization tends to reduce the ferron moment.

The conditions of stability of such a localized state can be found by discussing a situation in which the electron level is close to the top of the well. Then, the radius of the electron state $\aleph^{-1}$ is large compared with the radius of the ferromagnetic region $\boldsymbol{R}$. If we retain only the terms which are linear in $\aleph \boldsymbol{R}$, we find that Eqs. (2) and (3) give the following expression for the polarization energy:

$$E_p \approx -\frac{e^2 \aleph}{\varepsilon^*} 2(1 - \ln 2). \tag{5}$$

Thus, on the basis of Eq. (5), the functional **F** which includes the relationship between $\aleph$ and $\boldsymbol{k}$ given by Eq. (3) has the following form in the limit $\aleph \boldsymbol{R} << 1$:

$$E \approx \frac{\hbar^2 k^2}{2m} - \frac{As}{2} - 2(1 - \ln 2)\frac{e^2}{\varepsilon^*}\sqrt{\frac{mAs}{\hbar^2} - k^2} + \frac{4\pi}{3}|J|s\left(\frac{R}{a}\right)^3, \tag{6}$$

where $\boldsymbol{J}$ - **ZIs**; I is the Integral of the exchange between neighbors; Z is the coordination number; s is the spin of a magnetic atom; a is the lattice constant.

The parameter $\boldsymbol{k}$ is defined by the equation

$$\sin kR = \frac{2}{\pi} kR_m, \qquad \frac{\pi}{2} < kR < \pi, \qquad R_m^{-1} = \frac{2}{\pi\hbar}\sqrt{mAs}.$$

The maximum value of $\boldsymbol{J}$ can be found by equating to zero both the energy $\boldsymbol{E}$ of Eq. (6) and its derivative with respect to the well radius:

$$E = 0, \qquad \frac{\partial E}{\partial R} = 0. \tag{7}$$

We shall now report a numerical calculation for europium chalcogenides with the following parameters: $\boldsymbol{e}_O \approx 9.4$, $\boldsymbol{e}_\infty = 5$, $\boldsymbol{As} = 0.6$ eV, $\boldsymbol{W} = 5$ eV, $\boldsymbol{a} = 3 \cdot 10^{-8}$ cm, $\omega = 0.01$ eV, $\alpha = 4$. The effective mass of an electron is assumed to be approximately equal to the free-electron mass [6]. In this case we can assume that $|E_p| >> \alpha\omega$, i.e., we can ignore the energy of the polarization of the lattice by a free electron. The maximum Neel temperature T_{NC} of a crystal in which a ferron should still be stable at $T << T_N$; is

T_{NC}≈21°K. In the absence of the electron-phonon interaction, the maximum Neel temperature is T^{max}_{N}≈9°K, i.e., this interaction doubles the ferron stability.

We can easily also find the highest temperature at which a ferron state is stable if its Neel point $\mathbf{T_N}$ is less than the maximum value $\mathbf{T_{NC}}$ representing the upper limit of the ferron stability. If $\mathbf{T > T_N}$, the free energy of the system under discussion differs from Eq. (6) because the quantity | $\boldsymbol{J}$ |s in the last term is replaced with the gain in the free energy D(T) resulting from the creation of a ferromagnetic microregion in an antiferromagnetic semiconductor at a nonzero temperature (this energy is calculated per atom).

We can find F using the well-known thermodynamic relationship between the free energy and the average energy E(T), as given in ref. 7:

$$E = T^2 \frac{\partial}{\partial T}\left(\frac{F}{T}\right). \tag{8}$$

Integrating this expression between $\boldsymbol{T}$ and ∞ and bearing in mind that $\boldsymbol{F}\rightarrow\boldsymbol{-T}\ln(2\boldsymbol{s}-1)$ in the limit $\boldsymbol{T}\rightarrow\infty$, we find that

$$F(T) = -T\ln(2s+1) + T\int_T^\infty \frac{E(\tau)}{\tau^2}d\tau \tag{9}$$

If the magnetic interaction is described by the Heisen berg Hamiltonian, we find that in the nearest-neighbor approximation the energy per atom is

$$E(T) = -\frac{J}{2s}\langle s_0 s_\Delta\rangle \tag{10}$$

where $\langle s_0 s_\Delta\rangle$ is the correlation function of the nearest-neighbor spins. It follows from Eqs. (9) and (10) that if we know the temperature dependence of $\langle s_0 s_\Delta\rangle$, we can then find the free energy of an antiferromagnet. This energy can be estimated by the method of high-temperature expansions:

$$\langle s_0 s_\Delta\rangle = \frac{Tr\left\{\left(s_0 s_\Delta\right)\cdot e^{-\frac{Hm}{T}}\right\}}{Tr\left\{e^{-\frac{Hm}{T}}\right\}} = \frac{Js(s+1)}{ZT} \tag{11}$$

The quantity | J |s in Eq. (6) should be replaced with the difference between the energy of ferromagnetic ordering and the free energy of antiferromagnetic ordering at $T \geq T_N$ in accordance with Eqs. (9) - (ll):

$$D(T) = -\frac{Js}{2} - F(T) = T\ln(2s+1) + \frac{J^2(s+1)^2}{4ZT} - \frac{Js}{2} \tag{12}$$

Hence, it follows that the ferron state should be destroyed at a temperature T_F ~ 20°K, i.e., the stabilization of a ferron by the lattice polarization is double (for the parameters given above) that in the unpolarized lattice case.

These estimates are in agreement with the experimental results reported in ref. 3, which demonstrate that a ferron may exist in a paramagnetic phase at temperatures considerably lower than T_N.

REFERENCES

1. E. L. Nagaev,Pis`ma Zh. Eksp. Teor.Fiz. 6, 485 (1967) [JETP Lett.6, 18 (1967)]
2. .E. L. Nagaev, Zh. Eksp. Teor. Fiz. 54, 228 (1968) [Sov. Phys. JETP27, 122 (1968)].
3. P. Wachter, CRC Crit. Rev. Solid Stare Sci. 3, 189 (1972).
4. Yu. A. Firsov (ed.), Polarons [in Russian], Nauka, Moscow (1975).
5. I. Pekar, Investigations in Electron Theory of Crystals [in Russian], Gostekhizdat, Moscow (1951).
6. E. L Nagaev, in: Rare Earth Semiconductors (ed. by V. P. Zhuze) [in Russian], Nauka, Leningrad (1977).
7. L. Landau and E. Lifshitz, Statistical Physics, Pergamon Press, London (1958).