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Magnetic polarons (ferrons) of complicated structure

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So far magnetic polarons in antiferromagnetic semiconductors (ferrons) were assumed to arise due to the charge carrier self-trapping by a ferromagnetic or canted antiferromagnetic region. If the ferron size is not very large, they should have much more complicated structure: the magnetically deformed region consists not only of a magnetized core, which traps the electron, but also of its surrounding with the oppositely directed total moment, which repulses the trapped electron. The compensating moment of the surrounding oscillates with a period of doubled lattice constant and with the amplitude diminishing very smoothly, on increase in the distance from the core.

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The energy of a charge carrier in a magnetic semiconductor depends strongly on the type of the magnetic ordering, being minimal for the ferromagnetic ordering. For this reason the carriers tend to establish the latter. In nondegenerate antiferromagnetic semiconductors they can do that only inside a small region since a single electron cannot establish a ferromagnetic ordering in a crystal of macroscopic size. The idea that a mobile electron or hole creates a magnetized region inside of an antiferromagnetic semiconductor which traps it was advanced more than 30 years ago [1 – 4], and after this was repeated by many other authors (see, e.g., the review article [5]). In its simplest version a conduction electron creates a ferromagnetic region and stabilizes it by its localization inside this region.

Certainly, this state belongs to the self-trapped ones and in this respect resembles the well-known lattice polaron. This justifies, to some extent, the use of the name "magnetic polaron" for it adopted in the pioneering papers [1, 2]. But, in reality, this name is inaccurate as creation of this quasiparticle means change of the crystal magnetization. Meanwhile, the term "magnetic polarization" is never used for the crystal magnetization. For this reason the present author proposed later the name "ferron" for the quasiparticle invented by him in Refs. [1,2]. The ferrons were observed experimentally in EuSe and EuTe [6].

In addition to the free ferrons, the bound ferrons were considered in Refs. [2, 7, 8]. They correspond to the electrons (holes) captured by the donor (acceptor) impurities in an antiferromagnetic semiconductors. These electrons (holes) produce magnetized regions in the vicinity of the donors which attract the electrons to the donors jointly with the Coulomb potential of the donor atom. The central portion of the magnetized region may be completely ferromagnetic, and the rest – canted antiferromagnetic. Perhaps, the former may be absent at all. The bound ferrons influence the electrical and magnetic properties of the crystals strongly. In particular, due to their large magnetic moments, bound ferrons can change the sign of the paramagnetic Curie temperature from negative to positive. They also lead to a jump in the crystal magnetization in weak magnetic fields [9].

These results were obtained for the ferron sizes large as compared with the lattice constant. In the present paper the ferrons of the size comparable with the lattice constant will be considered. It will be shown that in some cases its structure may be considerably more complicated than that of the large ferrons. Especially nontrivial is the fact that the local magnetization can be with alternating signs. This means that the core of the magnetically deformed region attracts the electron and the region outside the core repulses it. The coordinate dependence of the compensating moment of the surrounding should be rather complicated for small magnetic anisotropy: it should oscillate with a period of doubled lattice constant and with the amplitude diminishing very smoothly, on increase in the distance from the core.

Such a complex ferron can be considered to be a manifestation of a real magnetic polaron. In fact, the magnetic polarization corresponds to the appearance of a magnetic moment inside a certain region of the crystal and of a compensating moment with the opposite direction elsewhere. A close analogy with the lattice polarization is obvious. The latter leads to the appearance of the local charge density but the total crystal charge

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remains zero. But one should point out that the total magnetization remains nonzero in the case considered since the magnetic compensation is incomplete. Hence the quasiparticle discussed is intermediate between the ferron and the true magnetic polaron.

A simple model will be used to corroborate the idea of the true magnetopolaronic effect. The self-trapped state which is found in this model corresponds to a state intermediate between the bound ferron and the bound true magnetic polaron. Due to an impurity potential, the electron is located at magnetic atoms closest to the impurity. The localization region acquires a core magnetic moment, becoming ferromagnetic or canted antiferromagnetic.

It will be shown that the region surrounding the localization region also acquires a magnetic moment. Its total value is opposite to the direction of the ferron moment and compensates the former up to more than 70 % in the one-dimensional case and up to 40 % in the three-dimensional case. The compensating moment displays oscillations with a period equal to the doubled lattice constant. If the magnetic anisotropy is small, the oscillation amplitude tends to zero very smoothly. For larger anisotropy, the tendency toward the oscillation attenuation increases strongly. The magnetization compensation decreases the energy of the self-trapped state very strongly, e.g., more than twice in the one-dimensional case.

First an one-dimensional chain of the magnetic atoms will be considered within the framework of the $s-d$-model, to which the uniaxial magnetic anisotropy term is added since it determines the direction of the magnetic moments.

The atomic chain is directed along the y-axis, and the magnetic easy axis is x. The undisturbed magnetic structure is represented by two sublattices with the spins up ($S^x = S$) for the even atomic numbers $g = 2n$ and spins down ($S^x = -S$) for the odd atomic numbers $g = 2n + 1$. Under influence of the trapped electron the d-spins can be deviated from the x-axis. The donor impurity is assumed to be located symmetrically with respect to these two atoms belonging to different magnetic sublattices. Then the Hamiltonian of the system can be represented in the form

$$H_{sd} = -t[a^*_{-1,\sigma}a_{0,\sigma} + a^*_{0,\sigma}a_{-1,\sigma}] - \\ - A\sum_{g=-1,0}(\mathbf{s}\mathbf{S}_g)_{\sigma\sigma'}\, a^*_{g,\sigma}a_{g,\sigma'} - \\ - I\sum \mathbf{S}_g\mathbf{S}_{g+1} - K\sum (S^x_g)^2, \qquad (1)$$

where $a^*_{g,\sigma}, a_{g,\sigma}$ are the s-electron operators corresponding to the conduction electrons or holes at atom g with the spin projection σ, $\mathbf{s}$ is the s-electron spin operator, $\mathbf{S}_g$ that of the d-spin of atom g, the d-spin magnitude being S. The form (1) assumes that the $d-d$-exchange integral I corresponding to the antiferromagnetic ordering is negative. The constant K of the uniaxial anisotropy is positive. The energy of the Coulomb interaction between the impurity and the electron is an additive constant and for this reason is omitted here.

Only the lowest bound state of the conduction electron will be considered. It corresponds to the s-electron shared with equal probabilities between atoms with the numbers $g = -1$ and $g = 0$. Perhaps, such a situation is possible also for the self-trapped electron in a perfect crystal but for very special parameter values.

We begin with the case of the magnetic anisotropy tending to zero. The electron effect on the d-spins is tantamount to an effective magnetic field directed along the z-axis. Hence in the vicinity of the impurity the d-spins lie inside the $z-x$ plane. By the symmetry of the system with $K \to 0$, $S^z_{2n} = S^z_{-2n-1}$ with the magnetized core formed by atoms –1 and 0, and the S^x- components are of the opposite signs for the two sublattices.

In what follows, the inequality $AS \ll W$ will be first used where W is the charge carrier bandwidth. In this case it is convenient to carry out the canonical transformation of the s-electron operators which diagonalizes the first term in Eq. (1):

$$a_{\pm,\sigma} = 2^{-1/2}[a_{-1,\sigma} \pm a_{0,\sigma}]. \qquad (2)$$

Then the standard procedure of constructing the magnetic Hamiltonian from the $s-d$-Hamiltonian (1) can be used: the latter is averaged over the s-electronic state. Taking its spin to be aligned with the z-axis, one puts $< a^*_{g,1/2}a_{g,1/2} >= 1$ for $g = 0$ or –1 and all other averaged pairs of operators equal to zero.

Introducing the polar angles for the d-spins, one can represent the magnetic energy in the form

$$E = J\sum_g \cos(\theta_g + \theta_{g+1}) - \\ - L[\cos\theta_{-1} + \cos\theta_0] - K/2\sum_g \sin^2\theta_g + E_0, \qquad (3)$$

$$J = -IS^2, \qquad L = AS/4,$$

where E_0 is the energy of the antiferromagnetic ordering.

Minimizing Eq. (3) with respect to the polar angles of the spins θ_g, one obtains the following set of the nonlinear equations:

$$J\sin(\theta_g + \theta_{g+1}) + J\sin(\theta_g + \theta_{g-1}) - \\ - L\sin\theta_g[\delta_{g,-1} + \delta_{g,0}] + K\sin(2\theta_g) = 0. \qquad (4)$$

Obviously,

$$\theta_{\pm\infty} = \pi/2. \tag{5}$$

This boundary condition remains in force if $KS^2N \gg L$ where N is the number of atoms in the chain.

Further, the solution should satisfy the equality

$$\theta_{-(g+1)} = \theta_g \tag{6}$$

with $g \geq 0$. Hence, it is sufficient to consider nonnegative g.

The corresponding treatment for the double exchange case $W \ll AS$ can be carried out using the De Gennes [10] expression for the effective hopping integral between atoms 0 and (-1)

$$t_{\text{eff}} = t \cos\left(\frac{\theta_{-1} + \theta_0}{2}\right). \tag{7}$$

This Eq. ensures a high accuracy if the total angle between the spins is not very large [9]. Using Eqs. (1), (6), (7), one again arrives to the set of equations (4) but with $L = t$ in this case.

To solve the set of equations (4), one should make it finite, putting

$$\theta_f = \pi/2 \tag{5a}$$

for some value f of the number of the atom. The cutt-off of the set of equations (4) at a finite f is tantamount to using a variational procedure. Hence, it exaggerated the energy the more the less is f.

The case of $f = 1$ corresponds to the purely ferron state when the magnetized region coincides with the core region, inside which the electron is localized (atoms 0 and –1). But already accounting for the deviation of the next spin ($f = 2$) will lead to appearance of the magnetic polarization.

For $f = 2$ one can obtain solutions analytically if $L \ll 1$ or $L \gg 1$ (L in the J units). In the former case, putting $\theta_0 = \pi/2 - \alpha$ and $\theta_1 = \pi/2 - \beta$, one finds for $KS^2 \ll Ł, J$: $\alpha = 2L/5$ and $\beta = -\alpha/2$. As the angles α and β are proportional to the core moment and to the compensating moment, respectively, one sees that the latter amounts to one half of the former. In the limiting case of large L, when $\theta_0 \ll 1$, one finds θ_1 to be $3\pi/4$. Hence, the compensating moment depends on L very weakly and amounts to 71 % of the core moment.

Now let us discuss the intermediate L values. Results for them are obtained by numerical calculations based on the set of Eqs.(4). For example, for $L = 3$ the ground-state energies of the system in the J units are –2.72 for $f = 1$, –4.78 for $f = 2$ and –5.846 for $f = 16$. Hence even the simplest version of accounting for the compensating moment $f = 2$ decreases the bound ferron energy by a factor of 1.76, and for $f = 16$ by a factor of 2.15.

For $f = 1$ the ferron magnetic moment coinciding with that of the core (0 and –1 atoms) is $1.62S$. For $f = 2$ the core moment is $1.796S$, i.e., higher than for $f = 1$. But, instead, the compensating magnetic moment appears at atoms $g = 1$ and –2. Its total value is -1.058S, which amounts to 59 % of the core moment. Hence the total magnetic moment of the system is $0.738S$, which amounts only to 41 % of the core magnetic moment.

But a more detailed investigation with $f = 16$ shows that the compensating magnetization is not concentrated in the closest vicinity to the core. It is long-range and has a rather complicated structure. To begin with, the total core moment is $1.992S$ for $L = 3$. Moreover, the same sublattice, to which the atom $g = 0$ belongs, is magnetized in the same direction as this atom, and the magnetization of the lattice falls down very smoothly, so that the magnetization of the atom $g = 14$ amounts to 18 % of that of atom $g = 0$. But the other sublattice is magnetized still more strongly in the opposite direction, and this magnetization falls off very smoothly, too. Hence the magnetization outside the ferron oscillates with a period of the doubled lattice constant and with the amplitude decreasing with increasing distance from the ferron (Figure). The total compensating moment outside the electron localization region up to atom $g = 15$ is –1.044S.

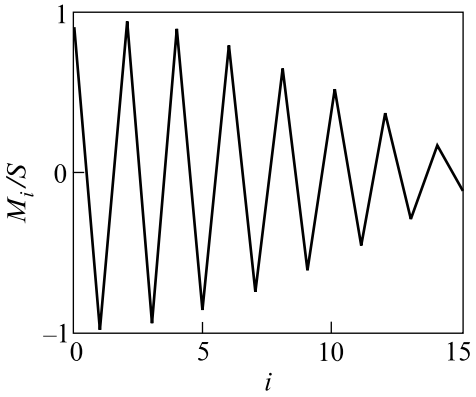

Fig. 1. Dependence of the moment M_i on the number i of the magnetic atom (one-dimensional system)

The oscillation length decreases with increasing magnetic anisotropy. The fact that the anisotropy suppresses the magnetization oscillations is obvious from the limiting case of the Ising model $K \to \infty$. Then the magnetization is aligned not with the z-axis but with the x-axis. This problem is solved exactly: for $L < 2J$ the ferron does not exist, and for $L > 2J$ it corresponds

to the spins of atoms 0 and (–1) parallel to each other. Outside this core the d-spins form the ideal antiferromagnetic ordering with the spins of atoms bordering the core being antiparallel to the core spin. Thus, the oscillations of the magnetic moment outside the core are really absent in the limit of a giant magnetic anisotropy.

The lower bound for the compensating moment up amounts to 52 % of the core moment. It should be noted that,though the energies for $f = 2$ and $f = 16$ differ only by 22%, qualitatively, the behavior of the magnetic polarization in these cases differs drastically.

Nevertheless, in both these cases the real contribution of such bound ferrons to the magnetization of the crystal in a magnetic field is considerably less than it was assumed initially. Hence accounting for the magnetic polarization is necessary for an accurate calculation of the magnetic properties of the ferron-containing crystals.

The ferron contribution will be evaluated in the three-dimensional case. It will be assumed that the crystal structure is simple cubic, and that the antiferromagnetic ordering is staggered. The donor atom is located at the center of an unit cell, and the donor electron is spread over 8 atoms located at the angles of the unit cell. Only the magnetic moments of these atoms and of 24 atoms adjacent to them will be taken into account. The former atoms form the moment of the ferron core M_C and the latter the compensating moment of the surrounding M_S.

Using the three-dimensional analogue of the Hamiltonian (1) and again introducing the angles θ_0 and θ_1 for the moments of the former and of the latter, respectively, after minimizing the total energy with respect to the angles, one arrives to the following equations:

$$\begin{gathered} -3J\sin(2\theta_0) - 3J\sin(\theta_0 + \theta_1) + L\sin\theta_0 = 0, \\ \sin(\theta_0 + \theta_1) + \sin(2\theta_1) + 3\cos\theta_1 = 0. \end{gathered} \tag{8}$$

Solution of the Eqs.(8) shows that the ratio M_S/M_C depends on L very weakly and is close to 40%. For the same reasons as in the one-dimensional case, the oscillations of the magnetic moment should also exist in the three-dimensional case. Again the bound ferromagnetic polaron energy is of the order hundred percent lower than the ferron energy. This energy lowering is very large and points to a considerably smaller density of the thermally delocalized charge carriers than in the usual ferron scenario.

The energy of the ready-made noninteracting magnetic moments directed normally to the sublattice does not change by their reversing. Hence these moments can lead a very rapid growth of the magnetization of the crystal in very small magnetic fields, with which they are aligned. It dominates for noticeable impurity densities as the magnetization of the antiferromagnetic portion of the crystal is still very small in such fields. In the limiting case $T \to 0$ an arbitrarily small field should cause the complete ordering of the bound ferron moments, which manifests itself as a steep jump in the magnetization of the crystal. As follows from results presented above this jump may turn out to be considerably less than it was assumed previously. At finite temperatures, if the field is too small to change the magnetic moments of the bound ferrons, the ferron contribution to the crystal magnetization is described by the Langevin function. Respectively, the growth of this contribution with the field turns out to be considerably more smooth.

It should be specially noted that the present investigation refutes the point of view expressed by some physicists according to which the bound electrons can produce the canted antiferromagnetic ordering in antiferromagnetic semiconductors.

Concerning the actuality of the present investigation, it should be noted that the problem of the magnetic polarons (ferrons) remains actual for more than 30 years beginning from their theoretical prediction [1]. This is related to the fact that just ferrons determine the electrical and magnetic properties of many magnetic materials, including the manganites which attract much attention due to their giant magnetoresistance now. Now the bound ferron existence is beyond any doubts. Experimental data on them are presented in the review article [11]. A refined theory of the ferron states developed here which is based on the partial compensation of the magnetic moment of the ferromagnetic region by its surrounding will make it possible to treat phenomena in these materials more consistently.

The theoretical prediction about the magnetization oscillations made above did not find an experimental confirmation yet, but this theoretical result is quite new and possibly will stimulate new experimental investigations. At least, this effect seems to be interesting from the physical point of view.

The small magnetic polarons investigated here are typical of slightly doped manganites. But they differ from the lattice small polarons drastically. The latter are characterized by the fact that in the zeroth approximation the electrons are localized at certain sites and due to this cannot establish the ferromagnetic ordering even in very small crystal regions (for this aim the electron transitions between the magnetic atoms are necessary). The small magnetic polarons considered here allow the electron transitions between the magnetic atoms and hence the local ferromagnetic ordering.

It should be stressed that the small lattice polarons are hypothetical quasiparticles, and their existence is not proved yet. According to the estimates of the present author, the standard polarization small polarons are impossible in the magnetic semiconductors and manganites as they are only partially polar crystals [11, 12]. On the other hand, in such Jahn-Teller materials as the manganites the coupling strength between the holes and Jahn-Teller phonons is unknown. Meanwhile, this coupling should be very strong for the formation of the small Jahn-Teller polarons, the reasons for which are quite not obvious.

The most decisive argument agains small lattice polarons of both types in the manganites is their metallic state at sufficiently high doping levels. Really, in materials where they exist the Mott delocalization of the donor electrons or acceptor holes, i.e. the metallic state, is impossible because of giant effective masses of the charge carriers in the small polaron states [11]. The same, the similarity of the basic properties of the manganites with the properties of the standard non-Jahn-Teller and only partially polar magnetic semiconductors makes it possible to conclude that the small lattice polarons do not exist in the manganites.

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