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The energy spectrum and g-factor of the bound magnetic polaron in a cubic Fe-based diluted magnetic semiconductor

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Abstract. The bound magnetic polaron (BMP) in a cubic Fe-based diluted magnetic semiconductor is considered. It is shown that the energy spectrum of the BMP is identical to that of the three-dimensional harmonic oscillator with a spin-orbital coupling. The polaron effect manifests itself in the existence of a splitting in the energies of the excited states of the polaron. It is shown also that the g-factor of the BMP is smaller than the g-factor of a free electron calculated in the molecular-field approximation.

1. Introduction

The bound magnetic polaron (BMP) is one of the most interesting physical phenomena arising in diluted magnetic (also referred to as semimagnetic) semiconductors (DMSs) due to the strong exchange interaction of a carrier with magnetic ions. In the family of Mn-based DMSs the polaron effect for electrons bound on shallow donors has now been thoroughly examined both theoretically and experimentally (see, for a review, [1]). In particular, it is found that there exists a zero-magnetic-field spin splitting of the electron level in a non-vanishing random exchange field of all the ions in the electronic orbit [2].

There is now a growing interest in the study of physical properties of DMSs based on the other transition-metal ions. The specific feature of wide-gap Fe-based $A_{1-x}^{II} Fe_x B^{VI}$ DMSs lies in the fact that the 3d⁶ multiplet of the Fe²⁺ ion (total orbital momentum L = 2 and spin S = 2) is split in the host crystal by both the crystal field and spin-orbital coupling, giving a non-degenerate ground state with $\langle S \rangle = \langle L \rangle = 0$; an applied magnetic field mixes this non-magnetic ground state and the excited states of the ion, so that Fe ions exhibit the Van Vleck paramagnetism [3], Correspondingly, the ground state of the system consisting of a donor-bound electron and a number of Fe^{2+} ions within its orbit is doubly degenerate, and no spin splitting exists in the absence of an applied magnetic field (this follows from the Kramers theorem) [4]. In this case the exchange interaction between the electron spin and those of the ions results only in a decrease of the energy of the system. However, it was shown in [5] and [6] that if the spin splitting of the BMP ground state in an applied magnetic field becomes close to an excitation energy of the Fe ion, the exchange interaction manifests itself in the effect of anticrossing of the Raman lines, which correspond to the electron spin-flip and excitation of an Fe ion. Note that there is an analogy between the Van Vleck BMP and the usual concept of the polaron arising due to the coupling between the electron and optical phonons: in both cases the electron interacts with elementary excitations

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of the crystal, which have approximately constant energy of excitation. The difference is in the form of the interaction, which results in some specific features of the Van Vleck BMP. The possibility of autolocalization of a free carrier in the Van Vleck DMS was considered in [7] (see also [8]).

In this paper we consider the Van Vleck BMP in a DMS with cubic symmetry. The case of a cubic crystal is of interest, in particular, due to the recent discovery of persistent shallow-donor electrons, arising in CdTe-based alloys after illumination [9]. Our main goal is to show that a very simple description of the polaron state exists, and the energy spectrum of the Van Vleck BMP is identical to the energy spectrum of a three-dimensional harmonic oscillator with spin-orbital coupling. This approach gives the possibility of extending the analysis of [6] and calculating the BMP wave functions and the energy spectrum in the case of a polaron with an arbitrary coupling. We will also find the effective g-factor of the BMP in the low-magnetic-field region, where the Zeeman terms can be considered as a perturbation.

2. The Van Vleck BMP in the zero-magnetic-field case

We consider a localized electron in the state with wave function $\psi(r)$ and Fe ions in the electronic orbit. In the absence of an applied magnetic field the Hamiltonian of the system reads [6]

$$H_0 = \sum_n H_n + \frac{1}{2}\sigma \cdot M \qquad M = \sum_n c_n S_n \tag{1}$$

where H_n and S_n are the Hamiltonian and the spin of the *n*th Fe²⁺ ion, σ_x , σ_y , and σ_z are the Pauli matrices for the electron spin, and

$$c_n = -\alpha |\psi(R_n)|^2 \tag{2}$$

is the exchange-interaction energy of the electron spin with the *n*th magnetic ion (R_n is the position of this ion and α is an exchange constant). The spectrum of Fe²⁺ in a cubic crystal was calculated in [10]. The lowest-lying states of the Fe ion are a singlet ground state A₁ (in the following we put its energy equal to zero) and a triplet T₁ with energy ε_0 . To a very good approximation the spin operator S_n connects the ground state $|n, 0\rangle$ of the *n*th ion only with the states belonging to the triplet T₁, and it is convenient to write the triplet state as

$$|n,i\rangle = (1/\sqrt{2})S_n^i|n,0\rangle$$
 $i = x, y, z.$ (3)

(The normalization factor $1/\sqrt{2}$ can be found simply noting that $\sum_i |\langle n, i|S_n^i|n, 0\rangle|^2 = S(S+1) = 6$.) The wave functions $S_n^j|n, i\rangle$ consist not only of the ground and lowestlying triplet states, but also of some other, higher-lying states of the ion. However, in the following we shall assume the interaction energies c_n to be much smaller than energy spacing ε_0 ,

$$c_n \ll \varepsilon_0.$$
 (4)

Then, in the polaron ground state the admixture of triplet states is small for each Fe^{2+} ion (of the order of c_n/ε_0), the admixture of highest-lying states (which is of the order of $(c_n/\varepsilon_0)^2$) can be neglected, and it is enough to consider only the ground and first excited

triplet states for each ion. The inequality (4) holds very well, e.g., for a shallow-donor electron.

Even in the case (4), when the exchange interaction of an electron with one ion is small, the polaron effect can be large because the electron interacts with a great number of magnetic ions. Indeed, in the second order of the perturbation theory with respect to the last term of the Hamiltonian (1) the polaron energy can be found using (3) to be (cf [5])

$$\mathcal{E}_{\text{pol}}^{(2)} = -3\eta\varepsilon_0 \qquad \eta = \frac{1}{2\varepsilon_0^2} \sum_n c_n^2. \tag{5}$$

So, the value of the polaron effect is governed by the parameter η , which is of the order of $(c_n/\varepsilon_0)^2 N$, where $N \gg 1$ is the typical number of magnetic ions in the electronic orbit. This parameter increases linearly with the concentration of magnetic ions x and may be large. Our aim is to consider the Van Vleck BMP for arbitrary values of coupling constant η without using any kind of perturbation theory.

The specific feature of the polaron Hamiltonian (1) lies in the fact that the interaction term is just a product of the electron spin operator σ and an additive operator acting on the states of all the magnetic ions (see (1)). This results, as was pointed out in [6], in a great reduction of the possible set of N-ion wave functions that can enter the expression for the BMP wave function. For example, among a great number of possible wave functions corresponding to the state of the system with an ion excited to the triplet state, only combinations

$$\frac{1}{2\varepsilon_0\sqrt{\eta}}\sum_n c_n S_n^i |\text{vac}\rangle \qquad i = x, y, z \tag{6}$$

can appear (|vac) denotes the state in which all the ions are in their ground states). In the general case we consider the states of the magnetic ion subsystem with k, l, m ions in $|x\rangle$, $|y\rangle$, and $|z\rangle$ states respectively:

$$|k,l,m\rangle = F(k,l,m) \sum_{\{n\}} c_{n_1} c_{n_2} \dots c_{n_{k+l+m}} S_{n_1}^x \dots S_{n_k}^x S_{n_{k+1}}^y \dots S_{n_{k+l}}^y S_{n_{k+l+1}}^z \dots S_{n_{k+l+m}}^z |\text{vac}\rangle$$
(7)

where the sum is taken over all sets $\{n_1, n_2, \ldots, n_{k+l+m}\}$ with $n_1 \neq n_2 \neq \ldots \neq n_{k+l+m}$. In the case when only a small number of magnetic ions is excited, $k + l + m \ll N$, the normalization coefficient reads

$$F(k,l,m) = \sqrt{k!l!m!} / (2\varepsilon_0 \sqrt{\eta})^{k+l+m}.$$
(8)

The matrix elements of collective operator M entering the interaction term of Hamiltonian (1) in the basis of wave functions (7) are

$$\langle k', l', m'|M^{x}|k, l, m \rangle = 2\varepsilon_{0}\sqrt{\eta} \left(\delta_{k+1,k'}\sqrt{k+1} + \delta_{k-1,k'}\sqrt{k} \right) \delta_{l,l'}\delta_{m,m'} + ST$$

$$\langle k', l', m'|M^{y}|k, l, m \rangle = 2\varepsilon_{0}\sqrt{\eta} \left(\delta_{l+1,l'}\sqrt{l+1} + \delta_{l-1,l'}\sqrt{l} \right) \delta_{k,k'}\delta_{m,m'} + ST$$

$$\langle k', l', m'|M^{z}|k, l, m \rangle = 2\varepsilon_{0}\sqrt{\eta} \left(\delta_{m+1,m'}\sqrt{m+1} + \delta_{m-1,m'}\sqrt{m} \right) \delta_{k,k'}\delta_{l,l'} + ST.$$
(9)

ST is stated in (9) in order to denote the terms much smaller than $\varepsilon_0 \eta^{1/2}$. For example, due to the fact that $\langle n, x | S_n^y | n, z \rangle \neq 0$ the matrix element $\langle 1, 0, 0 | M^y | 0, 0, 1 \rangle$ is also non-zero,

but has the order of magnitude of $(\varepsilon_0^2 \eta)^{-1} \sum_n c_n^3$, i.e. $\varepsilon_0 (\eta/N)^{1/2}$. Neglecting such small terms we arrive at the conclusion that there is a direct correspondence between the basis (7) and that of eigenfunctions of a three-dimensional harmonic oscillator (namely, the state $|k, l, m\rangle$ corresponds to the state of an oscillator with k, l, and m of x-, y-, and z-phonons, respectively), and the operator M corresponds to $2\varepsilon_0\eta^{1/2}(a+a^+)$, where a^+ and a are the creation and annihilation operators for this oscillator. Taking into account that the state with k+l+m excited ions has the energy $(k+l+m)\varepsilon_0$, we can rewrite the polaron Hamiltonian (1) as

$$H_0 = \varepsilon_0 [a^+ \cdot a + \sqrt{\eta} \sigma \cdot (a + a^+)]. \tag{10}$$

In such a way the many-particle Hamiltonian of the BMP is reduced to the Hamiltonian of the 3D harmonic oscillator with spin-orbital coupling proportional to $\sigma \cdot p$, where p is the momentum operator. The inequality (4) allowed us to linearize the response of the subsystem of magnetic ions and describe the action of a great number of magnetic ions on the electron spin with field operators a^+ and a.

The problem of diagonalization of the Hamiltonian (10), as well as of the initial Hamiltonian (1), is connected with the non-commutation of the Pauli matrices, but now it is possible to use the methods developed for the harmonic oscillator and the properties of its wave functions. The solution for arbitrary values of coupling constant η is given in the appendix. The polaron Hamiltonian commutes with operator

$$J = \frac{1}{2}\sigma + i(a \times a^{+}) \tag{11}$$

which has the meaning of the operator of the total angular momentum for the 3D oscillator (10), and the polaron states can be classified by values of $J^2 = j(j+1)$, where $j = \frac{1}{2}, \frac{3}{2}, \ldots$, and $J_z = -j, \ldots, j$. According to the appendix, the energy spectrum of the polaron is given by the solutions of the equation

$$-E/\varepsilon_0 + j - \frac{1}{2} = A_1 \eta / \{B_1 - A_2 \eta / [B_2 - A_3 \eta / (B_3 - \ldots)]\}$$
(12)

where

$$B_k = -\frac{E}{\varepsilon_0} + k + j - \frac{1}{2} \qquad A_k = \begin{cases} k & \text{if } k \text{ is even} \\ k + 2j + 1 & \text{if } k \text{ is odd} \end{cases}$$
(13)

Along with j we will also characterize the polaron states by the principal quantum number n = 1, 2, ..., in such a way as $E_{1j} < E_{2j} < ...$ The ground state of the polaron is that with n = 1, $j = \frac{1}{2}$. The polaron ground-state energy $E_{pol} = E_{1,j=1/2}$ decreases monotonically with increasing coupling constant and has the following asymptotic behaviour:

$$E_{\text{pol}} = -3\eta\varepsilon_0$$
 $(\eta \ll 1)$ $E_{\text{pol}} = -(1+\eta)\varepsilon_0$ $(\eta \gg 1).$ (14)

For the intermediate values of η the ground-state energy and the energies of a few lowestlying excited states were found from equation (12) numerically. The excitation energies of the polaron $\Delta E_{n,j} = E_{n,j} - E_{1,1/2}$ as functions of the coupling constant are shown in figure 1.

At the end of this section we will discuss the physical meaning of the quantum number j of the BMP states. It follows from the commutational relations for the spin operator of the Fe ion and (3) that the matrix elements of S_n between the T_1 states are

$$\langle n, i | S_n^j | n, k \rangle = \frac{1}{2} i \varepsilon_{ijk} \tag{15}$$





Figure 1. The dependences of the excitation energies of the BMP $\Delta E_{n,j} = E_{n,j} - E_{1,1/2}$ (in units of ε_0) on coupling constant η . The quantum numbers of excited states of the polaron are shown in parentheses: (n, j).

Figure 2. The ratio of the BMP g-factor g_p to the g-factor of a free electron g^* as a function of coupling constant η .

where *i*, *j*, k = x, *y*, *z*, and ε_{ijk} is the totally antisymmetric tensor of the third rank. (15) implies that the matrices (3×3) of the ion spin operator between functions $2^{-1/2}|n, x - iy\rangle$, $|n, z\rangle$, $2^{-1/2}|n, x + iy\rangle$ differ from those of angular momentum *l* only by a factor of $\frac{1}{2}$. In our approximation (4), when the interaction of the electron spin with each ion is taken into account only in the first order, one can consider the collective states (7) with one excited ion $(|1, 0, 0\rangle, |0, 1, 0\rangle$, and $|0, 0, 1\rangle$) as the states with angular momentum l = 1; the states with two excited ions as superpositions of states with l = 0, 1, and 2; etc. This is the physical meaning of the orbital momentum $i(a \times a^+)$ of the 3D oscillator (10) and the reason for the existence of the quantum number $j = l \pm \frac{1}{2}$ for the BMP. Note also that the existence of splitting in the energies of the excited states of the polaron is in agreement with the energy spectrum of the system consisting of one electron and one Fe ion, which was calculated in [6].

3. The g-factor of the polaron

In an applied magnetic field it is necessary to add the term

$$H' = \frac{1}{2} g_{\mathsf{e}} \mu B \cdot \sigma + g_{\mathsf{Fe}} \mu B \cdot \sum_{n} S_{n}$$
⁽¹⁶⁾

to Hamiltonian (1) (g_e and g_{Fe} are the band g-factor of an electron and that of an Fe ion, respectively). In the following we will neglect the interaction of the orbital momentum of the Fe ion with the magnetic field because the crystal-field splitting of the Fe levels is much greater than the spin-orbital one [6]. In the collective basis (7) the Hamiltonian H' takes the form

$$H' = \frac{1}{2}g_{e}\mu B \cdot \sigma + \frac{1}{2}ig_{Fe}\mu B \cdot (a \times a^{+}) + \varepsilon_{0}b \cdot (a + a^{+})$$
(17)

$$b = \frac{g_{\rm Fe}\mu B}{\varepsilon_0^2 \sqrt{\eta}} \sum_n c_n.$$
(18)

The second term in (17) arises due to relation (15). Performing the unitary transformation

$$e^{b \cdot (a^+ - a)} (H_0 + H') e^{b \cdot (a - a^+)}$$
(19)

with b given by (18) we can write the BMP Hamiltonian in a magnetic field as

$$H = H_0 + \frac{1}{2}g^*\mu B \cdot \sigma + \frac{1}{2}ig_{\text{Fe}}\mu B \cdot (a \times a^+) - \varepsilon_0 b^2$$
⁽²⁰⁾

where

$$g^* = g_e - \frac{4g_{Fe}}{\varepsilon_0} \sum_n c_n = g_e + \frac{4N_0 \alpha x}{\varepsilon_0} g_{Fe}$$
(21)

is the g-factor of the free electron in a cubic Fe-based DMS (see, e.g., [11]). In (21) we have used the definition (2) and replaced the summation over magnetic ions by the integration over their positions. N_0 is the number of cations per unit volume.

Hamiltonian (20) describes properly the energy spectrum of the BMP in the region of moderate magnetic fields, $g_{Fe}\mu B \ll \varepsilon_0$, where magnetic-field-induced mixing of the states of the Fe ion is small. For a typical value of $\varepsilon_0 \simeq 2$ meV it corresponds to B < 5 T. The third term of (20) describes the splitting linear in B of the excited triplet states of the ions, and the last term, which is proportional to the number of ions in the electronic orbit and does not depend on the exchange constant α , gives the decrease in energy of the system due to the Van Vleck paramagnetism. Due to the strong enhancement of the electron g-factor g^* the second term of (20) becomes comparable to ε_0 in the region of moderate magnetic fields. The anticrossing of the split-up ground level of the polaron with split-down excited ones in the region $g^*\mu B \simeq \varepsilon_0$ [5] can be easily considered on the basis of Hamiltonian (20) for the case of weak coupling, when the second term of H_0 (10) can be regarded as a perturbation. It should be noted also that the present theory cannot be directly extended to the region of high magnetic fields, $g_{Fe}\mu B \ge \varepsilon_0$, because the relations (3) for the ground and lowest-lying excited states of the Fe ion do not hold in this case.

In this section we will pay attention to the region of small magnetic fields, where $g^*\mu B \ll \varepsilon_0$ and the splitting of the ground state of the BMP can be written as $g_p\mu B$. The g-factor of the BMP g_p , as well as the g-factor of a free carrier in a DMS, is strongly enhanced due to the exchange interaction with magnetic impurities: $g_p, g^* \gg 1$. However, due to the fact that the polaron ground state is not a state with well defined projection of the electron spin (see (A7)) the polaron g-factor turns out to be smaller than the g-factor of a free electron in the molecular-field approximation (21). The higher the coupling constant is, the smaller g_p is. The dependence $g_p(\eta)$, which was found by consideration of the second term in (20) as a perturbation and with use of (A7) for the wave function of the ground state of the polaron, is shown in figure 2 (the third term in (20), which gives the correction to g_p of the order of $g_{Fe} \simeq 2 \ll g^*$, was neglected in this calculation). The polaron g-factor decreases rapidly with the coupling constant (for the weak-coupling case $g_p \simeq (1-4\eta)g^*$) and tends to the limit $\frac{1}{3}g^*$ at $\eta \gg 1$.

4. Numerical estimations and conclusion

It was shown in sections 2 and 3 that, in the case when the exchange interaction of the electron spin with one Fe ion is small (4), all physical properties of the BMP are determined by one parameter, the coupling constant η given by (5). Substitution of (2) into (5) and

replacement of the summation over n by integration over the positions of magnetic ions gives

$$\eta = x \frac{N_0 \alpha^2}{2\varepsilon_0^2} \int |\Psi(\mathbf{R})|^4 \,\mathrm{d}\mathbf{R}.$$
(22)

We perform the numerical estimations for $Zn_{1-x}Fe_xSe$. Because the Fe-Fe exchange interaction has not been taken into account and the present theory is valid only for small enough x, we will put in the following x = 0.05. For the H-like wave function and using the values $N_0\alpha = 250$ meV, $\varepsilon_0 = 1.8$ meV [12] we find $\eta = 19.2/(N_0a_B^3)$. The basic assumptions of the present theory, i.e. (i) the interaction of the electron with one ion is small and (ii) there is a large number of ions in the electronic orbit, imply $(N_0\alpha/\pi\varepsilon_0(N_0a_B^3))^2 \ll 1$ and $8\pi x(N_0a_B^3) \gg 1$. The maximum value of the coupling constant compatible with these conditions is $\eta_{max} = 0.16$ (the localization radius $a_B \simeq 20$ Å). It is seen from figures 1 and 2 that this value of η_{max} corresponds to the case of intermediate coupling.

For the electron bound on a shallow donor $(a_{\rm B} = 40 \text{ Å})$ we obtain $\eta \simeq 0.02$. Correspondingly, the splitting between two lowest-lying excited states of the polaron, which are the coupled states of the electron spin and collective states of ions with one ion excited to the triplet $(n = 2, j = \frac{1}{2} \text{ and } n = 1, j = \frac{3}{2}$, see figure 1), is about 2 cm⁻¹. Such a splitting can lead to the existence of two satellite lines in the Raman spectra, which are placed around the line with $\hbar\Delta\omega = \varepsilon_0$ corresponding to the excitation of a lone Fe ion. The possibility of experimental observation of optical transitions of the polaron from the ground to excited states is connected with the problem of preparation of high-quality Febased semimagnetic alloys. It follows also from the results of section 3 that the ratio of the polaron g-factor to the g-factor of a free electron for $Zn_{0.95}Fe_{0.05}Se$ is $g_p/g^* = 0.93$. This correction to the g-factor should be taken into account if the s-d exchange constant is determined from the spin splitting of donor-bound electrons.

In conclusion, it is shown in this paper that in a cubic Fe-based DMS the exchange coupling of the spin of a donor-bound electron with a great number of Fe ions can be treated as a coupling of the spin with a three-dimensional harmonic oscillator. This makes it possible to calculate the energy spectrum of the BMP for arbitrary values of the coupling constant. The polaron effect can lead to the existence of additional lines in the Raman spectra, which correspond to the transitions of the polaron from the ground state to excited states. It is shown also that the polaron effect results in a decrease of the giant spin splitting of the electron level: the g-factor of the BMP is smaller than the g-factor of a free electron.

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Appendix

It is convenient to solve the Schrödinger equation

$$H|\Phi\rangle = E|\Phi\rangle$$

(A1)

with Hamiltonian (10) in the representation of the coherent states. Namely, multiplying (A1) on $|q\rangle = \exp\{q \cdot a - q^* \cdot a^+\}|0\rangle$, where $q = q_1 + iq_2$ and $|0\rangle$ is the vacuum state $(a|0\rangle = 0)$, we obtain

$$q \cdot (\mathrm{d}\varphi/\mathrm{d}q) - \sqrt{\eta}\sigma \cdot (\mathrm{d}\varphi/\mathrm{d}q) - \sqrt{\eta}\sigma \cdot q\varphi(q) = (E/\varepsilon_0)\varphi(q) \tag{A2}$$

$$\varphi(q) = \exp\{\frac{1}{2}|q|^2\}\langle q|\Phi\rangle. \tag{A3}$$

The eigenfunction with the total angular momentum j and its projection on the z-axis $J_z = j$ (see the paragraph after (11)) can be written in the form

$$\varphi_j(q) = (q_x + \mathrm{i}q_y)^{j-1/2} \sum_{k=0}^{\infty} u_k (\sigma \cdot q)^k \uparrow .$$
(A4)

(The expressions for the states with $J_z \neq j$ can be found from (A4) by the rotation of the coordinate system.) Substitution of wave function (A4) into (A2) gives

$$\sqrt{\eta}u_{k-1} - B_k u_k + \sqrt{\eta}A_{k+1}u_{k+1} = 0 \qquad (u_{-1} \equiv 0)$$
(A5)

where coefficients A_k and B_k are given by (13). The trinomial recurrence relations for u_k can be analysed in the same way as in the theory of the Mathieu functions. We introduce $F_k = u_k/u_{k-1}$, and obtain from (A5) the expressions for them in the form of continued fractions

$$F_k = \sqrt{\eta} / (B_k - \sqrt{\eta} A_{k+1} F_{k+1}) \qquad k = 1, 2, \dots$$
 (A6)

The spectral equation is found from relation (A5) at k = 0 to be $B_0 = \sqrt{\eta}A_1F_1$, which leads to the expression (12). The wave function is then written as

$$\varphi_j(q) = u_0(q_x + \mathrm{i}q_y)^{j-1/2} \left\{ 1 + \sum_{k=1}^{\infty} F_1 F_2 \dots F_k(\sigma \cdot q)^k \right\} \uparrow$$
(A7)

where the constant u_0 should be found from the normalization condition. It can be shown that in the case $j = \frac{1}{2}$:

$$u_0^{-2} = 1 + \sum_{k=1}^{\infty} A_1 A_2 \dots A_k (F_1 F_2 \dots F_k)^2.$$
 (A8)

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