ELECTRONS, INDIRECT EXCHANGE AND LOCALIZED MAGNONS IN MAGNETOACTIVE SEMICONDUCTORS

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The energy spectra of carriers and magnons in ferromagnetic and antiferromagnetic semiconductors are investigated for the case of very strong coupling between the spins of the magnetic atoms and the conduction electrons. An effective Hamiltonian acting in space of the smoothed spin-variable functions is set up and considerably simplifies the analysis. The carriers in ferromagnets are quasiparticles, which are called spinpolarons. At T = 0 their effective mass does not exceed by more than two times the effective mass of the band electron. With increasing temperature, it increases in accordance with a power law but at a rate which is slower than the decrease of the magnetization. Their mobility when scattered by magnons is estimated. A theory of indirect exchange between magnetic atoms via spinpolarons is developed, and is a limiting case opposite to that of the Ruderman-Kittel-Josida theory. Indirect exchange cannot be described here by an effective Heisenberg Hamiltonian. However a dispersion law of the usual type is obtained for the magnons, and the effective exchange integral is proportional to the width of the electron band. It is demonstrated that local magnon levels are formed near non-ionized donors. They are due to the dependence of the donor energy on the configuration in the vicinity of the donor and to indirect exchange via the donor electron. The employed magnetic adiabatic approximation can also be used to estimate the effective mass and decay of a single-spin magnetic polaron, the conditions of existence of which are investigated in the present paper.

THE question of the carrier states in magnetoactive semiconductors and of the influence of the carriers on the magnetic properties of these semiconductors is presently one of the least developed in solid-state theory. For a long time, the main emphasis in the investigation of such materials was on the interaction between the carriers and the optical lattice vibrations, as the result of which many papers were published on the theory of the small-radius polaron. The experimental results indicate, however, that in most cases such a model does not give an adequate description of the properties of such semiconductors[1]. We can therefore expect their distinguishing properties to be more due to strong carrier interaction with the magnetic subsystem than to interaction with the optical phonons.

We shall henceforth deal only with interactions between the carriers and the magnetic subsystem. Apparently, the first to advance the idea that autolocalized states of the conduction electron are energetically favored in antiferromagnetic semiconductors were Goodenough[2] and De Gennes[3]. Wolfram and Callaway[4] attempted to consider the autolocalized state of an electron in a ferromagnetic semiconductor in analogy with the small-radius polaron. It should be noted that the result of the present paper do not confirm the main conclusions of[4].

In an earlier paper[5] we proposed a model of autolocalized state of an electron in antiferromagnetic semiconductor, called the magnetic polaron. The violation of the magnetic order in this model reduces to a 180° rotation of the spins of a sufficiently large number of magnetic atoms from the same sublattice, so as to produce a microscopic region with ferromagnetic ordering, in which the electron is localized. It was shown in[6] that quasiparticles of a different type—quasioscillators—are possibly realized in antiferromagnetic semiconductors under certain conditions, besides the magnetic polarons. They are characterized by the fact that the degree of violation of the magnetic order increases with increasing distance from the electron to the center of the autolocalized state.

The present paper is devoted to an investigation of the energy spectra of carriers and magnons in ferromagnetic and antiferromagnetic semiconductors. We use the s-d model, and the s-d exchange integral is assumed to be very large. It is shown that under these conditions the carriers in the ferromagnetic semiconductors are quasiparticles, which henceforth will be called spinpolarons. There effective mass at T = 0 is not more than double the effective mass of the band electron. With increasing temperature, the mass increases in a power-law fashion, but more slowly than the decrease of the magnetization.

The spinpolarons tend to establish a ferromagnetic order in the system, and this leads to the appearance of indirect exchange between the magnetic atoms via the spinpolarons. Usually the indirect exchange is described with the aid of an effective Heisenberg Hamiltonian, derived by Ruderman, Kittel,[7] and Josida[8] for the case of small intraatomic exchange integrals. In this paper we developed a theory of indirect exchange for the opposite limiting case. It is shown that at very large s-d exchange integrals, the indirect exchange cannot be described with the aid of an effective Heisenberg Hamiltonian. For magnons, however, a dispersion law of the usual type is obtained, and the effective exchange integral is proportional to the width of the electron band.

Besides the free spinpolarons, interest attaches to the localized spinpolarons. These are, for example, states of the extra electrons of the donor centers. These

electrc s greatly influence the magnetization of the crystal ıear the defects, and produce in the magnon spectrum local or quasilocal levels of a different nature than those investigated in[9-11].

As indicated above, whereas the concentration of the carriers is insufficient to establish ferromagnetism of the crystal as a whole, in antiferromagnetic semiconductors each carrier tends to establish ferromagnetic ordering in a small fraction of the crystal and to be autolocalized in it. The procedure developed in this paper makes it possible to determine the effective mass and the damping of the magnetic polaron (or quasi-oscillator), something which was not done in the earlier paper[5,6]. The analysis is limited to the case when only one spin in the lattice is established antiparallel to the moment of its sublattice. Since the motion of the magnetic polaron is also accompanied by a displacement of the defect of the magnetic ordering, the effective mass of the magnetic polaron turns out to be larger.

In the Appendix are analyzed conditions for the existence of a magnetic polaron with one reversed spin (obviously, this model supplements the one considered in[5]). It is shown, in particular, that in the case of positive intraatomic exchange integral, such a single-spin polaron can be realized only at large values of the spin.

1. EFFECTIVE HAMILTONIAN OF THE SYSTEM IN THE CASE OF A STRONG INTRAATOMIC COUPLING

The analysis that follows is carried out formally within the framework of the s-d model, although it remains valid if the role of the conduction electron is played by the extra d-electron. It is assumed that the conduction electrons move only over the cations. The Hamiltonian of the system is written in the form

$$\mathcal{H} = \Sigma[E_f\delta_{\sigma\sigma'} - A(\mathbf{s}\mathbf{S}_f)_{\sigma\sigma'}]a_{f\sigma}^{*}a_{f\sigma'} + B\Sigma a_{f\sigma}^{*}a_{f+\Delta,\sigma} + \mathcal{H}_M, \quad (1)$$

$$\mathcal{H}_M = -\frac{I}{2}\Sigma(\mathbf{S}_f\mathbf{S}_{f+\Delta}). \quad (2)$$

Here $a_{f\sigma}^{*}$ and $a_{f\sigma}$ are the operators of creation and annihilation of the conduction electron with spin projection σ on the magnetic atom with number $f = (f_x, f_y, f_z)$, f and $\mathbf{S}_f$ are the spin operators of the conduction electron and of the f-th atom. Further, E_f is the "atomic" energy of the electron at the f, B is the Bloch integral, A is the s-d exchange integral, and I is the exchange integral between the magnetic atoms. The index Δ numbers the values of z of the nearest neighbors of the given atom. We have left out from the Hamiltonian (1), to simplify the notation, the terms describing the exchange of the conduction electron with the more remote atoms. It is assumed that the crystal lattice is primitive cubic.

Principal attention is paid in this paper to the situation typical of transition-element compounds, when the intraatomic exchange integral E, the width of the electron band 2zB, and the integral I of exchange between the magnetic atoms are related by

$$|A|S \gg z|B| \gg z|I|, \quad (3)$$

where S the spin the magnetic atom. Relation (3) indicates the orders of the quantities contained in them with respect to the overlap of the orbits of the neighboring atoms.

Under condition (3), the coupling between the spins of the electron and of the atom at which it is located is so strong, that they add up to form a total spin equal to $S + 1/2$ when $A > 0$ and $S - 1/2$ when $A < 0$. Thus, the crystal contains "irregular" spins; these differ by 1/2 from the regular ones, which move when the electrons move through the crystal. Since this motion causes the spin of each atom to become indeterminate, the choice of the spin variables describing the states of the magnetic atoms becomes difficult. To get around this difficulty, we shall construct later for the system an effective Hamiltonian that acts in the space of the functions of the smoothed spin variables, and which is invariant against the displacement of the conduction electrons.

If the system contains only one conduction electron, then its wave function at $A < 0$ is found in the form of an expansion in the basis functions

$$\chi_g(S_0^z) = \left[\sqrt{\frac{S + S_{g0}^z}{2S+1}}\, a_{g\downarrow}^{*} + \frac{a_{g\uparrow}^{*}S_g^{-}}{\sqrt{(2S+1)(S+S_{g0}^z)}}\right] \times \prod_f \delta(S_f^z, S_{f0}^z)|0\rangle_e, \quad S^{\pm} = S^x \pm iS^y. \quad (4)$$

Here $\delta(x, y)$ is the δ-function of discrete argument with values 1 or 0, and $|0\rangle_e$ is the vacuum electron function. The basis functions (4) describe the state of the system of spins, each of which has a fixed projection, when the total spin of the atom g, at which the electron is located, equals $S - \frac{1}{2}$. The Clebsch-Gordan coefficients in expression (4) differs from the usual ones, apart from the use of spin operators, also in that the projection of the total spin is expressed in terms of one of the possible values of the projection of the "proper" spin of the atom S_{g0}^z. Unlike the projection of the total spin of the atom, which can assume 2S values, S_{g0}^z can assume $2S + 1$ values. However, the additional degree of freedom is fictitious, for when $S_{g0}^z = -S$ the wave function (4) vanishes identically. Analogously, basis functions are constructed also for the case $A > 0$. Here, to the contrary, when using the projection of the "proper" spin of the atom instead of the projection of the summary spin, no account is taken of one of the allowed states. However, this is irrelevant when weakly-excited states of the system are considered and can be neglected at all temperatures if the spins are large.

The complete wave function of the system can be represented with the aid of the functions (4) in the form

$$\psi = \sum_{gS_0^z} c_g(S_0^z)\chi_g(S_0^z), \quad (5)$$

where the symbol S_0^z denotes the aggregate of variables S_{f0}^z for all the atoms. The coefficients $c_g(S_0^z)$ represent the envelope of the wave function of the system. It is important that the space of the parameters S_{f0}^z, on which they depend, remains invariant with respect to transitions of the electron from atom to atom.

Substitution of expression (5) in the Schrödinger equation with Hamiltonian (1) leads in the usual manner to a system of equations with respect to the coefficients $c_g(S_0^z)$. From the wave equation obtained in matrix form in this manner we can go over to an equation in operator form, by introducing the operators acting in the space of the functions of the parameters S_{f0}^z in the same manner as the corresponding operators in the space of

the functions of the true variables S_f^Z. In addition, to describe the spatial displacements of the electron, we introduce the parameters α_g^* and α_g. It should be emphasized that although these operators describe the creation or annihilation of an electron on an atom g, they are not operators of the free electron, but operators of the quasiparticles peculiar to the systems under consideration, namely the spinpolarons.

The use of relations of the type

$$S^+\delta(S^z,\ S_0{}^z) = S_0{}^-\delta(S^z,\ S_0{}^z)$$

makes it possible to obtain the effective Hamiltonian of the system, acting in the space of the envelopes of the wave functions $c_g(S^Z)$ (for simplicity, we shall leave out the index 0 of the smoothed spin variables S_{f0}^Z):

$$\tilde{\mathcal{H}} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_M; \tag{6}$$

$$\mathcal{H}_1 = \sum\left[E_g + \frac{A(S+1)}{2}\right]\alpha_g{}^*\alpha_g + \frac{B}{2S+1}\sum\sqrt{(S+S_g{}^z)(S+S_{g+\Delta}^z)}\,\alpha_g{}^*\alpha_{g+\Delta}, \quad A<0, \tag{7}$$

$$\mathcal{H}_1 = \sum\left[E_g - \frac{AS}{2}\right]\alpha_g{}^*\alpha_g + \frac{B}{2S+1}\sum\sqrt{(S+S_f{}^z+1)(S+S_{g+\Delta}^z+1)}\,\alpha_g{}^*\alpha_{g+\Delta}, \quad A>0;$$

$$\mathcal{H}_2 = \frac{B}{2S+1}\sum\frac{1}{\sqrt{S+S_f{}^z}}S_g{}^+S_{g+\Delta}^-\frac{1}{\sqrt{S+S_{g+\Delta}^z}}\alpha_g{}^*\alpha_{g+\Delta}, \quad A<0, \tag{8}$$

$$\mathcal{H}_2 = \frac{B}{2S+1}\sum\frac{1}{\sqrt{S+S_g{}^z+1}}S_g{}^-S_{g+\Delta}^+\frac{1}{\sqrt{S+S_{g+\Delta}^z+1}}\alpha_g{}^*\alpha_{g+\Delta}, \quad A>0;$$

$$\mathcal{H}_3 = \sum\left\{I(\mathbf{S_gS}_{g+\Delta}) - I_t\left[S_{g+\Delta}^z\left(S_g{}^z\pm\frac{1}{2}\right) + \frac{1}{2}S_{g+\Delta}^+S_g^-\sqrt{\frac{S+S_g{}^z\pm1}{S+S_g{}^z}} + \frac{1}{2}\sqrt{\frac{S+S_g{}^z\pm1}{S+S_g{}^z}}S_g{}^+S_{g+\Delta}^-\right]\right\}\alpha_g{}^*\alpha_g; \tag{9}$$

$$I_t = I\left[1\mp\frac{1}{2S+1}\right]. \tag{9'}$$

The upper sign in (9) and (9′) corresponds to the case $A > 0$, and the lower one to the opposite case.

It is easy to indicate the physical meaning of the different terms in the effective Hamiltonian (6). The Hamiltonian $\mathcal{H}_1$ reflects the main feature of the spinpolaron states: the total gain of the s-d exchange energy is ensured in these states. The Hamiltonian describes transitions of the spin polaron from atom to atom without a change of the smoothed spin projections, while the Hamiltonian $\mathcal{H}_2$ describes transitions in which the smoothed projections change. In both cases, the effective transition integrals depend essentially on the configuration of the smoothed spins. Finally, the Hamiltonian $\mathcal{H}_3$ represents a correction to the magnetic Hamiltonian $\mathcal{H}_M$, connected with the fact that the spin of the atom bearing the electron differs by one half from the spins of the remaining atoms. It can be shown that if we go over to variables $(S \pm \frac{1}{2})_g^Z$, the term proportional to I_t assumes the usual Heisenberg form.

2. INDIRECT EXCHANGE AND SPINPOLARONS IN FERROMAGNETIC SEMICONDUCTORS

As is well known, the presently existing theory of indirect exchange, formulated by Ruderman and Kittel[7] and by Josida[8], is based on the inequality $z|B| \gg |A|S$, which is well satisfied in rare-earth metals. We consider here indirect exchange in the opposite limiting case (3), which is typical of transition-element compounds. A large class of such compounds has carrier densities on the order of 10^{19}–10^{28} cm^{-3}, and exchange via the carriers can play an important role in them.

For simplicity we assume that there is no indirect exchange, described by the Hamiltonian $\mathcal{H}_M(2)$, between the magnetic atoms. Then, at the considered carrier densities, the ferromagnetic ordering of the spin system becomes energetically favored, since, according to (7), it ensures a maximum width of the spinpolaron band, and consequently the lowest position of its bottom. For concreteness we assume that the intraatomic exchange integral is negative. We introduce in the usual manner the operators of creation and annihilation of the spin deviations:

$$S_g{}^+ = b_g\sqrt{2S}, \quad S_g{}^- = b_g{}^*\sqrt{2S}, \quad S_g{}^z = S - b_g{}^*b_g, \tag{10}$$

which can be assumed to be connected by Bose commutation relations. In weakly excited states of the magnetic subsystem, the approximate equality $(b^*b)^n \approx b^*b$ is valid. This makes it possible to represent the Hamiltonian (8) in the form

$$\mathcal{H} = \frac{2SB}{2S+1}\sum\alpha_g{}^*\alpha_g + \frac{B}{2S+1}\sum\left\{b_{g+\Delta}^*b_g - 2S\left(1-\sqrt{1-\frac{1}{2S}}\right)(b_g{}^*b_g + b_{g+\Delta}^*b_{g+\Delta})\right\}\alpha_g{}^*\alpha_{g+\Delta} + \text{const}. \tag{11}$$

The first term in the right side of (11) is the Hamiltonian of the free spinpolarons. As seen from its structure, if $A < 0$, then the effective mass of the spin polaron is larger at T = 0 by a factor (1 + 1/2S) than the effective mass of the "bare" band electron. It is easy to verify that when $A > 0$ the effective mass of the spinpolaron coincides with the electron mass.

The analysis will now be continued for large spins, when the second term in (11) can be expanded in powers of 1/S. In the fundamental approximation in 1/S, the interaction with the magnons leads to renormalization of the Bloch integral

$$\bar{B} = B\left\{1 + \frac{1}{2S}\sum_q[\cos q_x a - 1]\,m_q\right\}, \tag{12}$$

where m_q is the average number of magnons with momentum q, and a is the lattice constant. It is seen from (12) that when the temperature is increased the effective Bloch integral decreases in proportion to $T^{5/2}$. Accordingly, the effective mass increases, the bottom of the spinpolaron band shifts, and this should become manifest in a shift of the intrinsic absorption edge of the semiconductor. We note that this shift occurs more slowly than the change of the magnetization, which is proportional to $T^{3/2}$. It should be emphasized that the values of the effective mass of the spinpolaron in a ferromagnet at T = 0 and the character of its temperature dependence remain unchanged if account is taken in the Hamiltonian (11) of the Hamiltonian $\mathcal{H}_M(2)$ of the magnetic subsystem. It is seen from the structure of the Hamiltonian (8) that a temperature shift of the bottom of the band should occur also in the paramagnetic state, but this shift is not estimated here. The results of[12] make it possible to obtain such an estimate in the limiting

case of a weak s-d coupling.

We now proceed to discuss the spectrum of the elementary excitations of the magnetic subsystem. In this case it is impossible to separate from the Hamiltonian (8) the term with the structure of the Heisenberg Hamiltonian, which would describe this exchange. Nonetheless, it follows directly from the Hamiltonian (11) that here, too, it is possible to introduce spin waves whose dispersion law is given in the fundamental approximation by

$$\omega_q = I_{eff}(z - \gamma_q),$$

$$I_{eff} = \frac{|B|}{2S}\sum_k n_k \cos k_x a, \quad \gamma_q = \sum_\Delta e^{-iq\Delta}, \tag{13}$$

where n_k is the distribution function for the spinpolarons with quasimomentum k (it is seen from the limiting transition $S \to \infty$ that the spinpolarons obey Fermi statistics).

As seen from expression (13), the effective exchange integral differs in this case strongly from the indirect exchange of the standard theory. It is proportional to the Bloch integral and to the total carrier density, if the latter is sufficiently small. Expression (13) has a structure typical of the nearest-neighbor approximation, and consequently the long-range action is immaterial here. At large carrier densities, the effective exchange integral can become negative, i.e., the ferromagnetic ordering turns out to be unstable. It should be noted that the ordinary theory of indirect exchange leads to stability of the ferromagnetic ordering at small carrier density[13].

It might seem from the form of the Hamiltonian (12) that a gap should appear in the magnon spectrum in the higher approximations in 1/S. A calculation made in the $1/S^2$ approximation has shown that there is no gap, at least in this approximation, for when $q \to 0$ the contribution of the terms diagonal in the spinpolaron indices (12), of order $1/S^2$, is compensated by the contribution of the nondiagonal terms of order 1/S:

$$\Delta\omega_q = \frac{|B|}{16S^2}\left\{\frac{1}{N}\sum\gamma_k n_k + \frac{P}{N^2}\sum\frac{[2\gamma_{r-q} - \gamma_r - \gamma_k]^2}{\gamma_r - \gamma_k} n_k(1 - n_r)\right\}$$

where P denotes the principal value. This approximation is insufficient to obtain the correct attenuation of the magnons, since their frequency is $\sim 1/S$.

The effective Hamiltonian (11) makes it possible to find not only the spectra of the elementary excitations, but also the kinetic coefficients. In particular, it is possible to obtain an estimate for the mobility of the spinpolarons when scattering by magnons predominates. If the spinpolaron gas is not degenerate, then it follows from the energy conservation laws that the momentum transfer from the spinpolaron to the magnons is of the order of $\sqrt{I/|B|}$ of its value (if the ferromagnetic order is due to indirect exchange, then I should be taken to mean here I_{eff}). Therefore the scattering of the spinpolarons by the magnons can be assumed in this case to be elastic. In the Born approximation, we obtain for the relaxation time the following expression:

$$\frac{1}{\tau_k} \approx \frac{1}{12}\frac{1}{(2\pi)^3 S^2}\frac{ka}{\hbar}|B|\left(\frac{k_B T}{I}\right)^{3/2}. \tag{14}$$

Using formula (4), we can show that at low temperatures the spinpolaron velocity is connected with its energy by the usual relation. The use of the relaxation time (14) leads to a temperature dependence of the spinpolaron mobility like $\sim T^{-4}$.

3. LOCAL AND QUASILOCAL MAGNON LEVELS

In this section we shall show that the presence of donor or acceptor impurities in ferromagnetic or antiferromagnetic semiconductors leads to the appearance of local or quasilocal levels in the magnon spectrum. At the present time, the local levels that result from the difference between the spin of the impurity atom or its exchange integral from the regular values of these quantities have been thoroughly investigated[9-11]. The local magnetic levels considered here have a different nature.

We shall investigate below the change, due to the donor atom, of the spectrum of the spin waves in a ferromagnetic semiconductor. We are considering the following model: One of the magnetic atoms is replaced by an impurity atom, which differs from the regular atoms by having an extra valence electron. This electron, however, cannot be regarded as localized on the impurity atom, for it is smeared out to a considerable degree over the neighboring magnetic atoms. In the considered materials, the radius of the localized electron state is as a rule small. It will be assumed that the electron can be located, apart from the impurity atom, only on magnetic atoms of the first coordination sphere of the defect. Accordingly, in the Hamiltonian (6) the "atomic" energy E_g is equal to E_0 for the central atom and to E_1 for its nearest neighbors.

It is natural to attempt to separate the fast and slow motions in the system under consideration, which is described by an effective Hamiltonian (6), by using the adiabatic approximation. In accordance with the condition (3), the spinpolaron should represent a rapid subsystem, and the smoothed spins a slow subsystem. The situation becomes more complicated, however, because the presence of the term $\mathcal{H}_2$ in the Hamiltonian (6) causes the rapid displacement of the spin polaron to be accompanied by rapid motions induced by them in the system of the smoothed spins. Therefore the adiabatic approximation can be used only at large spin values $S \gg 1$, when the motion frequencies corresponding to the Hamiltonian $\mathcal{H}_2$, in accordance with (8), are small in the weakly-excited states of the magnetic subsystem. It can be included in the Hamiltonian of the slow subsystem, which assumes the following form:

$$\mathcal{H}_{Sn} = \tilde{E}_n(S^z) + (\Phi_n|\mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_M|\Phi_n), \tag{15}$$

$$\Phi_n(S^z) = \sum_g c_g(S^z)\alpha_g^*|0\rangle_e, \tag{16}$$

where $\tilde{E}_n(S^z)$ and Φ_n are the eigenfunctions and the corresponding eigenvalues of the spinpolaron Hamiltonian $\mathcal{H}_1$ (7). The spinpolaron index n will henceforth be omitted, since we shall consider only the ground state of the donor atom.

Let us discuss the structure of the Hamiltonian $\mathcal{H}_S$. The Hamiltonians $\mathcal{H}_3$ and $\mathcal{H}_M$ averaged over the spinpolaron state describe the change, due to the presence of the extra electron of the direct exchange in the vicinity of the defect. The averaged Hamiltonian $\mathcal{H}_2$ describes the indirect exchange between the magnetic atoms via the spinpolaron. At small deviations of the spins from the equilibrium positions, the spinpolaron energy can be represented in the form of a series in these deviations:

$$\tilde{E}(S^z) \approx -D_0(S_0^z - S) - D_1 \sum_{\Delta} (S_\Delta^z - S). \quad (17)$$

Obviously, the quantities D_0 and D_1 can be interpreted as effective magnetic fields acting near the defect on the spins of the atoms. The diagonalization of the Hamiltonian $\mathcal{H}_1$ at small deviations of the spins from the equilibrium is trivial, and the results will not be presented here.

In certain cases, interest attaches to a situation in which the inequality $z|B| \gg |A|S$, which the opposite of (3), is satisfied. It is easy to verify that it is then possible to apply the usual adiabatic approximation to the Hamiltonian (1): the role of the rapid subsystem is played by the electron, and that of the slow one by the spins of the atoms. In fact, in the first approximation in A/B, the only contribution to the Hamiltonian of the slow subsystem is made by the electron-spin interaction term in (1), containing products of the z-components of the spins. On the other hand, the indirect exchange between the magnetic atoms via the conduction electron appears here only in the next higher approximation. The action of the electron on the spins of the atoms can also be expressed here in terms of the effective magnetic fields, in analogy with (17).

The determination of the magnon spectrum is greatly simplified if account is taken of the fact that the electron is located with the largest probability on the central atom. Therefore, in all the terms of the Hamiltonian (15), with the exception of $\langle\Phi|H_2|\Phi\rangle$, we can put $c_g(S^z) = \delta(g, 0)$, leaving the definition of the effective field (17) and the indirect-exchange operator unchanged. The inaccuracies connected with such a procedure are insignificant, for in this problem the effective fields and the indirect-exchange affect the state of the magnetic subsystem more strongly than the change of the spin and of the exchange integral due to the presence of the defect. After going over to the Bose operators with the aid of relations (10), the Hamiltonian (15) for $A > 0$ assumes in the harmonic approximation the form

$$\mathcal{H}_S = -IS\sum_{f\Delta}(b_f^* b_{f+\Delta} - b_f^* b_f) + D_0 b_0^* b_0 + D_1 \sum_{\Delta} b_\Delta^* b_\Delta + \sum_{\Delta}\{[I_t S' - IS] b_\Delta^* b_\Delta + S(I_t - I) b_0^* b_0 - [S(I_t\sqrt{S'/S} - I) - B'](b_0^* b_\Delta + b_\Delta^* b_0)\}, \quad (18)$$

where

$$S' = S + \frac{1}{2}, \qquad B' = \frac{B}{2S+1} c_0(0) c_\Delta(\Delta).$$

The symbols $c_0(0)$ and $c_\Delta(\Delta)$ denote the amplitudes $c_g(S^z)$ in (16) for the cases when all the spins in the electron localization region are directed upward, with the exception of the atom spin 0 or Δ respectively, the projection of which differs by unity from the maximum value. The form in which the Hamiltonian $\mathcal{H}_S$ is expressed in (18) is valid also for the limiting case $|A|S \ll z|B|$, if we put in it B' equal to 0 and I_t and S' are replaced by I and S, respectively.

The Hamiltonian (18) differs from the Hamiltonian of[9,10] in that it contains terms with an effective magnetic field and the indirect-exchange operator. These terms change neither the symmetry nor the rank of the localized perturbation, and therefore the calculation of the energy spectrum is carried out here in exactly the same manner as in the cited papers. The frequencies of the two s-type oscillations, of the three p-type oscillations, and of the two d-type oscillations are determined from transcendental equations that coincide formally with Eqs. (3.5)—(3.7) of[10], but the parameters contained in them have a different meaning:

$$\varepsilon = \frac{D_0}{6IS} + \frac{I_t}{I} - 1, \quad \rho = \frac{D_1}{IS} + \frac{I_t S'}{IS} - 1, \quad \gamma = \sqrt{\frac{S'}{S}\frac{I_t}{I}} - 1 - \frac{B'}{IS}. \quad (19)$$

In addition, the exchange integral I in[10] differs by a factor of 2 from the corresponding quantity in this paper.

An analysis of the indicated equations allows us to state that in the investigated case the relative positions of the levels, generally speaking, are different than in ferromagnetic dielectrics. In principle this makes it possible to distinguish between levels of the considered type and the Callaway-Izyumov levels from neutron-scattering results. In particular, if we can neglect the indirect exchange and the change of the exchange integral and of the spin of the central atom (for example, when $|A|S \ll z|B|$), then the spectrum of the localized magnons is determined by a single parameter ρ, which in accordance with (19) is practically equal to ϵ. Using the table of the numerical values of the Green's functions from[9], we can show that p- and d-type levels exist if ρ exceeds 4.7 and 5.4 respectively. The higher of the s-levels appears when ρ exceeds 0.6, and the lower when it exceeds 3.4. In the limit $D_0 \gg D_1 \gg 6IS$, it is also easy to obtain estimates for the frequencies of the localized magnons. The frequency of the highest s level is close to D_0, and the frequencies of the remaining levels are

$$\omega_i \approx \frac{IS}{2}(\rho + \sqrt{\rho^2 + 24\rho}) + \frac{K_i (IS)^2}{D_1},$$

where the parameter $K_i = 39$, 41, and 47 for the d, p, and s levels, respectively.

In the case of an antiferromagnet, the spin of the impurity atom can be either parallel or antiparallel to the angular momentum of its sublattice, depending on the relation between A, B, and $E_0 - E_1$. The second case, in general, is similar to the case just considered. In the first case, on the other hand, owing to the interaction between the spins and the electron, the amplitude of their oscillations in the vicinity of the donor increases. Therefore local oscillations with energies lying below the continuous spectrum are possible at sufficiently large magnetic anisotropy.

4. EFFECTIVE MASS AND DAMPING OF MAGNETIC POLARON

An adiabatic approximation of the type used in the preceding section makes it possible to determine the effective mass and the attenuation of the magnetic polaron. The latter is defined as an autolocalized state of the conduction electron in the antiferromagnetic semiconductor, such that one or several magnetic atoms reverse their direction, so that the electron is inside a microscopic region with ferromagnetic ordering. In the case under consideration, when relation (3) is satisfied, it would be more accurate to speak of an autolocalized state of the spin polaron.

In this paper we consider the case when the spin of only one atom is opposite to the angular momentum of its sublattice (single-spin magnetic polaron). The conditions for the existence of a single-spin polaron are analyzed in the Appendix. In particular, it is shown there that the single-spin polaron can be realized at small spins of the atoms only in crystals with negative intraatomic integral A. The analysis in this section is confined to this case only.

As follows directly from the form of the Hamiltonian (6), the energy of the magnetic polaron is determined mainly by the transitions of the spinpolaron from atom to atom without a change of the smoothed spin projections. In fact, transitions with a change of the spin projections inside the ferromagnetic microregion are impossible. They can occur only when the spinpolaron emerges from the ferromagnetic microregion, and the probability of this event is small. Therefore the effective frequency of such transitions is small compared with the frequency of transitions without a change of the spin projection, and this inequality increases with increasing spin of the atoms. By virtue of the foregoing, as before, $\mathcal{H}_1$ (7) plays the role of the Hamiltonian of the fast subsystem, and the Hamiltonian of the slow subsystem is given by expression (15).

The Schrödinger equation with Hamiltonian (15) describes both the motion of the magnetic polaron and the natural oscillations of the magnetic subsystem. A similar situation obtained in Pekar's investigation[14] of a polaron in an ionic crystal. It was shown there that the spectrum of the optical phonons is not changed by the presence of the polaron, but the positions of equilibrium of the lattice oscillators follow the displacement of the polaron. In the case of a magnetic polaron, the situation is more complicated. The autolocalization of the electron inside the magnetic-ordering defect produced by it greatly changes the magnon spectrum of the crystal: local magnon levels of the type considered in the preceding section appear, and the density of the states of the continuous spectrum changes. The amplitude of the zero-point oscillations of each spin depends on the position of the magnetic polaron and follows its displacements. As will be shown subsequently, the possibility of motion of the magnetic polaron over the crystal at T = 0 is due to the zero-point oscillations of the spin.

Let us consider the state of the system at T = 0, when the center of the magnetic polaron is located at the site f_1. The spins of the atoms execute small oscillations about the equilibrium positions, but the spin of the central atom takes practically no part in them: first, it is surrounded by atoms with the same spin direction and, second, the rotation of its spin would greatly increase the spinpolaron energy $\tilde{E}(S^Z)$. Therefore in the zeroth-approximation Hamiltonian $\mathcal{H}_{fl}$, which determines the states of the magnetic subsystem at the indicated position of the center of the polaron, it is possible to exclude the terms T_{fl}, which change the projection of the spin of the central atom:

$$\mathcal{H}_{f_1} = \mathcal{H}_S - T_{f_1},$$

$$T_{f_1} = \frac{I}{2} \sum_{\Delta,\, g\neq f_1,\, f_1+\Delta} c_g(S^z)\, S_{f_1}^+ S_{f_1+\Delta}^- c_g(S^z)$$

$$- \Big[\frac{I_t}{2} \sum_{\Delta} \Big\{ c_{f_1}(S^z)\, S_{f_1+\Delta}^+ S_{f_1}^- \sqrt{\frac{S + S_{f_1}^z - 1}{S + S_{f_1}^z}}\, c_{f_1}(S^z)$$

$$+ c_{f_1+\Delta}(S^z)\, S_{f_1}^+ S_{f_1+\Delta}^- \sqrt{\frac{S + S_{f_1+\Delta}^z - 1}{S + S_{f_1+\Delta}^z}}\, c_{f_1+\Delta}(S^z) \Big\}$$

$$+ \frac{B}{2S+1} \sum_{\Delta} \frac{c_{f_1}(S^z)}{\sqrt{S + S_{f_1}^z}}\, S_{f_1}^+ S_{f_1+\Delta}^- \frac{c_{f_1+\Delta}(S^z)}{\sqrt{S + S_{f_1+\Delta}^z}} + \text{c.c.} \qquad (20)$$

After the Hamiltonian (20) is linearized, the spin operators are replaced by Bose operators with the aid of the relations

$$S_f^x = \sqrt{\frac{S}{2}}\,(b_f + b_f^*), \quad S_f^y = i(-1)^{n_f}\sqrt{\frac{S}{2}}\,(b_f^* - b_f),$$

$$S_f^z = (-1)^{n_f}(S - b_f^* b_f),$$

where n_f is the number of the sublattice of the atom f.

Diagonalization of the Hamiltonian $\mathcal{H}_{fl}$ in the harmonic approximation is carried out, as usual[15], by intermixing the annihilation operators b_f for the atoms of one sublattice and the creation operators b_f^* for the other sublattice. We introduce the operator P_{fl}^*, with the aid of which the ground state of the magnetic subsystem is expressed when the center of the polaron is located at the site f_1, in terms of the vacuum state $|0\rangle$ for the spin deviations in the following manner:

$$|f_1\rangle = P_{f_1}^*(b_{f_1}^*)^{2S}|0\rangle$$

(in the state $|0\rangle$, all the spins of one sublattice are directed upward, and those of the other downward). This relation reflects the fact that the spin of the atom f_1 in the state $|f_1\rangle$ does not take part in the zero-point oscillations, remaining fixed in a direction opposite to the angular momentum of its sublattice. The operator $P_{f_1}^*$ can be represented in the form of a series in the operators b_f^* with $f \neq f_1$.

In order to take the damping into account, it is necessary to consider the excited states with the same spin projection. The most significant of them are the two-magnon states:

$$|f_1\nu_1\nu_2\rangle = \beta_{1\nu_1}^*(f_1)\,\beta_{2\nu_2}(f_1)\,|f_1\rangle,$$

where $\beta^*(f_1)$ is the operator for creation of a magnon when the polaron center is on the site f_1, and the index 1 or 2 denotes the sublattice to which the magnon is assigned.

To construct the Green's function of the moving polaron, it is convenient to generalize the theory of the damping effects[16] to the case of a degenerate zeroth-approximation ground state. To this end, the wave function of the system is written in the form

$$\Phi(t) = \sum_{h_1} G_{h_1}(t)\,\Big|\, h_1\Big\rangle + \sum_{h_1\nu_1\nu_2} G_{\nu_1\nu_2|h_1}(t)\,\Big|\, h_1\nu_1\nu_2\Big\rangle, \qquad (21)$$

and the initial conditions are chosen as follows:

$$G_{\nu_1\nu_2|h_1}(\pm 0) = G_{h_1}(-0) = 0, \quad \frac{d}{dt}G_{h_1}(0) = \frac{e^{ikh_1}}{\sqrt{N}}\,\delta(t) \qquad (22)$$

(N is the number of the atoms in the sublattice, k is the quasimomentum of the polaron).

Under the initial conditions (22), the quantity G_{h_1} in (21) can be regarded as the Green's function of the magnetic polaron. Writing the equation of motion for the coefficients G and going over to the Fourier representation in the energy and in the quasimomentum, we obtain the following expression for the sought Green's function in the main approximation in the zero-point oscillation

amplitude:

$$G_k(E)=\{E-T_k-M_k(E)\}^{-1},\quad T_k=\frac{1}{N}\sum_{f_1}e^{ik(f_1-h_1)}\langle h_1|T_{f_1}|f_1\rangle,$$

$$M_k(E)=\frac{1}{N}\sum_{f_1g_1\nu_1\nu_2}\frac{e^{ik(f_1-h_1)}\langle h_1|T_{g_1}|g_1\nu_1\nu_2\rangle\langle g_1\nu_1\nu_2|T_{f_1}|f_1\rangle}{E-\omega_{\nu_1}-\omega_{\nu_2}+i\eta}\quad(\eta\to 0) \tag{23}$$

(ω_ν is the magnon frequency, and the energy is reckoned from the center of the polaron band).

It is necessary first of all to verify that the attenuation of the polaron states is sufficiently small. If the magnon ν is not localized, then the difference between the operators $\beta^*_{i\nu}(f_1)$ and $\beta^*_{i\nu}(f_3)$ is of the order of $1/\sqrt{N}$. The ratio of the weights of the two partial states from the series for $P^*_{f_1}$ in the creation operators, which differ from each other in a pair of extra reversed spins, is of the order of $1/2zS$. Accordingly

$$\langle h_1|T_{g_1}|g_1\nu_1\nu_2\rangle\,/\,\langle h_1|T_{g_1}|g_1\rangle\sim 1/2SzN. \tag{24}$$

In accordance with the meaning of the problem, interest attaches only to the energies $E\ll|I|Sz$. For these energies, according to (23)—(24), the following estimate is valid:

$$\frac{\operatorname{Im}M(E)}{T(E)}\lesssim\frac{|I|Sz}{(2\pi)^6(2Sz)^2E}\int d^3p_1d^3p_2\delta(E-w|p_1|-w|p_2|)\sim\frac{1}{(2Sz)^2}\left(\frac{E}{w}\right)^4,$$

where $w\sim ISz$ (p is a dimensionless quantity). This estimate proves the statement made above concerning the attenuation at T = 0.

In order to estimate the width of the magnetic-polaron band at T = 0, it is convenient to use not the spin-wave approximation, but directly perturbation theory, which readily enables us to find the coefficients in the series of $P^*_{f_1}$ in the operators b^*_f. As the zeroth-approximation Hamiltonian it is necessary to choose the Ising part of the Hamiltonian $\mathcal{H}_{f_1}$ (20). The perturbation Hamiltonian thus consists of terms with the structure $S^+_gS^-_{g+\Delta}$. The practical equivalence of the spin-wave approximation to the elementary perturbation theory is the direct consequence of the fact that in both cases the role of the small parameter is played by the reciprocal coordination number.

As is well known, the use of standard perturbation theory for large systems ($N\to\infty$) raises difficulties in the normalization of the wave function. Here, however, there is no need to calculate the normalization constant; this constant cancels out when $\langle h_1|T_{f_1}|f_1\rangle$ is calculated in the fundamental approximation in z^{-1}. When $S=\frac{1}{2}$ we obtain for the energy of the magnetic polaron as a function of the quasimomentum k the following expression:

$$E(k)=\frac{1}{2}[E(f_1f_2f_3)-E(f_1)]+\frac{1}{2}\Big[Bc_{f_3}(f_1f_2f_3)c_{f_3}(f_1)+I\sum_{g\neq f_1f_3}c_g(f_1f_2f_3)c_g(f_3)\Big]^2\sum_\lambda e^{ik\lambda}. \tag{25}$$

here $E(f_1)$ and $E(f_1f_2f_3)$ are the eigenvalues of the Ising part of the Hamiltonian (20) for spin configurations differing from the ideal antiferromagnetic ordering in the reversal of the spins at the atom f_1 and at the triplet of atoms f_1, f_2, and f_3 respectively. The atom $f_3=f_2+\lambda$ is taken to mean one of the nearest neighbors of the atom f_1 in the same sublattice 1, while f_2 is the common nearest neighbor of the atoms f_1 and f_3 from the sublattice 2. The amplitudes $c_g(f_1)$ and $c_g(f_1f_2f_3)$ have a similar meaning. An estimate of all these quantities makes it possible to rewrite expression (25) in the following form:

$$E(k)\approx\frac{1}{400|I|}\left[B+\frac{7}{2\sqrt{2}}\right]^2\sum_\lambda e^{ik\lambda}.$$

it can be shown that in order for the single-spin polaron to be energetically favored over the multispin polaron it is necessary to satisfy the inequality $|B|<12|I|$. It is clear therefore that the width of the polaron band in the case under consideration is comparable with the limiting energy of the antiferromagnetic magnon, i.e., as assumed, the motion of the magnetic polaron is slower. When $S>\frac{1}{2}$, the polaron band should be even narrower. It follows from the structure of the series for $P^*_{f_1}$ that it decreases with increasing spin like z^{1-4S}.

As already mentioned, in addition to magnetic polarons, quasioscillators can possibly be realized in antiferromagnetic semiconductors[6]. However, if the conduction electron can go off from the center of the quasioscillator state only to the neighboring atoms, then such a quasioscillator is actually a particular case of the magnetic polaron. Therefore the result obtained above gives an estimate also for the width of the quasioscillator band.

APPENDIX

SINGLE-SPIN MAGNETIC POLARON

We consider a magnetic polaron at rest, when the spin of only one of the magnetic atoms is reversed relative to the angular momentum of its sublattice. The ratio of the quantities A and B in the Hamiltonian (1) is assumed arbitrary, but each of them greatly exceeds the exchange integral I. The single-spin polaron is energetically favored because of the possibility of resonant transitions between the atom with the reversed spin and its nearest neighbors.

In accordance with the statements made in Sec. 4, we can disregard here the zero-point oscillations of the spin and those transitions of the electron from atom to atom who are accompanied by a change of the spin configuration. The wave functions of the systems are sought in a form analogous to expression (3) and (4) of the second paper of[5], with allowance for the fact that only the spin of one atom is reversed. Substitution of these functions into the Hamiltonian (1) leads to systems of equations similar to (6) and (11) of[5], but only without the Taylor expansion of the coefficient $c_{g\sigma}$ from the expressions for the wave functions.

As discussed in[5], when $A<0$, the aggregate of the coefficients $c_{g\sigma}$ can be regarded as a bispinor wave function. The corresponding eigenvalue problem is actually the bispinor Schrödinger equation with localized perturbation, connected with the spin flip at the center of the magnetic polaron. This equation, like Eq. (6) of[5], is solved by eliminating three components of the bispinor wave function, as the result of which a scalar equation of the following type is obtained:

$$\left[-\frac{AS}{2}+K(E')\delta(f_1,0)-E'\right]c_{f_1\uparrow}-B^2R(E')\sum_{\Delta\Delta'}c_{f_1+\Delta+\Delta',\uparrow}=0, \tag{A.1}$$

where

$$K(E')=\frac{AS}{2}\left\{2+A\left[\frac{A(S-1)}{2}+zS|I|+E'\right]^{-1}\right\},$$
$$R(E')=\frac{AS}{2}-E'+\frac{A^2S}{2}\left[E'+\frac{A(S-1)}{2}-zS|I|\right]^{-1},$$
$$E'=E-2zS^2|I| \quad (E_g=0).$$

Since the coefficients of the Hamiltonian in (A.1) themselves depend on the sought energy, the problem of its determination is essentially self-consistent. The quantity $K\delta(f_1, 0)$ in (A.1) will be regarded as the perturbation operator. It is easy to find the "energy spectrum" of the unperturbed Hamiltonian:

$$E(k,E')=-\frac{AS}{2}-B^2R(E')\gamma_k^2. \qquad (A.2)$$

The eigenfunctions of the unperturbed Hamiltonian are, obviously, plane waves (k—wave vector). Of course, unlike[17], the "energy spectrum" (A.2) cannot be interpreted as the spectrum of the system in the absence of the perturbation, since it depends on the sought energy. Using these eigenfunctions and eigenvalues of the unperturbed Hamiltonian, it is easy to construct for (A.1) a Green's function leading to the following equation for the determination of the energy of the magnetopolaron state:

$$1=\frac{K(E')}{\pi^3}\int_0^{\pi}\frac{d^3k}{B^2R(E')\gamma_k^2+AS/2+E'}. \qquad (A.3)$$

For the case $A > 0$ this energy is determined from the equation

$$1=\frac{M(E')}{\pi^3}\int_0^{\pi}\frac{d^3k}{R(E')+B^2N(E')\gamma_k^2},$$
$$N(E')=[AS/2+E']^{-1}, \quad M(E')=R(E')+E'+AS/2. \qquad (A.4)$$

It is easy to verify that when $|6B| \gg |AS|$ Eqs. (A.3) and (A.4) have no solutions, i.e., the single-spin polaron cannot exist. In the opposite limiting, (A.3) leads to the following result:

$$E\approx\frac{A}{2}(S+1)-\frac{2S}{2S+1}\sqrt{6}B+\frac{6|I|S^2(4S+1)}{2S+1}.$$

Equation (A.4), neglecting the exchange integral I in first order in B/A, reduces to a transcendental equation of somewhat simpler structure:

$$1+\frac{1}{2S}=\frac{\varepsilon}{\pi^3}\int_0^{\pi}\frac{d^3k}{\varepsilon-\cos k_x-\cos k_y-\cos k_z},$$

$$\varepsilon=\left|E'+\frac{AS}{2}\right|\frac{\sqrt{2S+1}}{2|B|}. \qquad (A.5)$$

An analysis of Eq. (A.5) shows that the discrete level is possible only for $S > 1$. It must be borne in mind that it should be sufficiently deep to offset the loss of magnetic energy when the spin of the central atom of the magnetic polaron is reversed. We can therefore expect that in fact when $A > 0$ the single-spin polaron can be realized only at even higher values of the spin. In the limiting case of large spins, the energy of the magnetic polaron has the same magnitude as when $A < 0$. A qualitative explanation of the difference in the tendency to autolocalization for positive and negative integrals A was given in[5].

Note added in proof (7 February 1969). Expression (13) for the effective indirect-exchange integral was obtained under the condition that the intra-atomic exchange energy AS is large compared with the width of the electron band. It can be shown, however, that in expression (13) can be used also under a much less stringent condition: this energy should be larger than the Fermi energy of the carriers.

In a recent paper (A. Yanase and T. Kasuya, J. Phys. Soc. Japan, 25, 1025, 1968) an attempt was made to consider the state of a magnetic defect of the same type as in Sec. 3 of this paper. However, Yanase and Kasuya lost sight of the need for seperating the fast and slow motions in the system. As the result, the form of the magnetic Hamiltonian (9) postulated by them is in error, and the results of the entire paper can not be regarded as correct.

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