

<sup>1</sup>Although the mapping of Fig. 3d has sections with  $|d\Phi/d\varphi| < 1$ , this mapping becomes uniformly stretching if the metric is properly chosen.

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## Indirect multispin exchange

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If the indirect exchange between  $f$ -spins is effected by  $s$ -electrons with nonzero total spin, then it is of essentially non-Heisenberg character. For the particular case of indirect exchange via a donor electron in a magnetic semiconductor it is possible to construct an equivalent magnetic Hamiltonian having the form of the square root of an expression bilinear in the  $f$ -spins. The Ruderman-Kittel term is a small correction to it. The constructed Hamiltonian accounts for the spectrum of the system accurately, but the average values of the spin operators can be expressed in terms of its eigenfunctions only in a manner that is, generally speaking, different from the manner accepted in quantum mechanics. The Hamiltonian contains all the spin invariants possible for isotropic systems: multispin, biquadratic, etc. The spin-spin interaction is noncentral. With the aid of this Hamiltonian the localized magnons in a ferromagnetic semiconductor are investigated.

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### 1. INTRODUCTION

As is well known, the isotropic exchange interaction is accurately described by the Heisenberg Hamiltonian only in the case of a system consisting of two spin- $\frac{1}{2}$  magnetic atoms. If the spin,  $S$ , of these atoms exceeds  $\frac{1}{2}$ , the exchange between them is described by a Hamiltonian that is a polynomial of degree  $2S$  in the scalar product,  $S_1 \cdot S_2$ , of the spins.<sup>[1]</sup> Even more complex is the situation in the case of a large number of atoms, when into the exchange Hamiltonian enter multi-spin terms of the type  $S_g \cdot S_f \dots S_k \cdot S_n$ . Although for many physical systems the Heisenberg term in the magnetic Hamiltonian is the dominant term, in certain cases the non-Heisenberg terms are not small. Of the non-

Heisenberg Hamiltonians only those that are linear combinations of quadratic and biquadratic terms (i.e.,  $S_g \cdot S_f$  and  $(S_g \cdot S_f)^2$ ; see, for example, Refs. 2-4) have been investigated in detail. Hamiltonians with four-spin terms,  $(S_g \cdot S_f)(S_k \cdot S_n)$ , added to the Heisenberg terms have also been investigated.<sup>[5]</sup>

In the present paper we shall show that in certain physical systems the isotropic exchange interaction is described by a Hamiltonian of a type entirely different from the type indicated above.<sup>[1]</sup> These are systems in which the indirect exchange between the localized  $f$ -spins is effected by mobile  $s$ -electrons that are completely polarized with respect to spin. Such a situation differs sharply from the indirect exchange in systems

described by the Ruderman-Kittel-Kasuya-Yoshida (RKKY) theory, in which the  $s$ -electrons in the zeroth approximation in the  $s$ - $f$  exchange are not polarized. In the RKKY theory the  $s$ - $f$  exchange contributes to the  $s$ -electron energy only in second-order perturbation theory. Since the  $s$ - $f$  exchange Hamiltonian is linear in the  $f$ -spins, in the indicated order the correction to the  $s$ -electron energy should be quadratic in these spins. Therefore, the magnetic Hamiltonian, which by implication is a correction, expressed in terms of the  $f$ -spins, to the  $s$ -electron energy, has in the RKKY theory the structure of the Heisenberg Hamiltonian.

If, on the other hand, the  $s$ -electrons are completely polarized with respect to spin, then the correction to the electron energy is nonzero even in first-order perturbation theory in terms of the  $s$ - $f$  exchange. This is precisely the situation in the case of a singly-charged donor in a magnetic semiconductor (e.g., the  $Gd^{3+}$  ion in  $EuO$ ): its single  $s$ -electron effects the indirect exchange between the  $f$ -spins in the vicinity of the defect. The spin of the  $s$ -electron is always parallel or antiparallel to the resultant moment of the atoms in the neighborhood of the defect, irrespective of how the moment is aligned in space. Therefore, the correction to the energy is proportional to the moment,

$$M = \left| \sum_{\mathbf{f}} \mathbf{S}_{\mathbf{f}} \right|,$$

of the region in which the  $s$ -electron is localized. Thus, the Hamiltonian should have a structure of the type  $[\sum \mathbf{S}_{\mathbf{g}} \times \mathbf{S}_{\mathbf{f}}]^{1/2}$ .

There also exist other systems in which the indirect exchange is effected by  $s$ -electrons polarized with respect to spin. Examples of these systems are degenerate ferromagnetic semiconductors. In them, as a rule, the distance,  $AS$ , between the subbands of the conduction band, split by the  $s$ - $f$  exchange, is, at  $T=0$ , high compared to the Fermi energy of the  $s$ -electrons.<sup>[6]</sup> However, the polarization vanishes at sufficiently high temperatures. Thus, the situation here is much more complex than in the case of indirect exchange in the vicinity of donors: the properties of degenerate semiconductors are described by a Hamiltonian of the root type only at low temperatures. The RKKY theory is applicable to them at temperatures much higher than the Curie point.<sup>[6]</sup> It has not as yet been possible to construct for degenerate semiconductors a magnetic Hamiltonian that is valid at all temperatures.

In the present paper a root Hamiltonian constructed for a singly-charged donor is used to investigate the influence of such defects on the magnon spectrum of a ferromagnetic semiconductor. This problem is similar to the problem of the Heisenberg defect in a ferromagnet<sup>[7,8]</sup>: a non-Heisenberg defect also causes the appearance of localized or quasi-localized levels in the magnon spectrum. But the situation here differs from the situation considered in Refs. 7 and 8 not only quantitatively, but also qualitatively. First, the effective exchange here is essentially noncentral. Secondly, in contrast to the situation considered in Refs. 7 and 8, the

range of the magnetic disturbance introduced by the electron of the donor can significantly exceed the lattice constant. It will be shown that, other conditions being equal, the appearance of localized magnons becomes more difficult as the radius of the electron orbit increases. Finally, besides the localized excitations corresponding to the small deviation from the ferromagnetic order, there also exist such high-energy localized excitations whose presence leads to the replacement of the ferromagnetic order in the vicinity of the defect by an antiferromagnetic order.

## 2. THE MAGNETIC HAMILTONIAN OF THE DONOR

We consider a positively-charged defect in a ferromagnetic semiconductor in the case when the defect retains a mobile electron near it. The Hamiltonian for the  $s$ - $f$  model in this case has the form

$$\mathcal{H} = \sum_{\mathbf{g}, \mathbf{h}} B(\mathbf{h}) a_{\mathbf{g}\sigma}^{\dagger} a_{\mathbf{g}+\mathbf{h},\sigma} + \sum_{\mathbf{g}} U(\mathbf{g}) a_{\mathbf{g}\sigma}^{\dagger} a_{\mathbf{g}\sigma} - A \sum_{\mathbf{g}} (\mathbf{S}_{\mathbf{g}} \mathbf{s})_{\sigma\sigma'} a_{\mathbf{g}\sigma}^{\dagger} a_{\mathbf{g}\sigma'}, \quad (1)$$

where  $a_{\mathbf{g}\sigma}^{\dagger}$  and  $a_{\mathbf{g}\sigma}$  are the creation and annihilation operators for an  $s$ -electron with spin  $\sigma$  on a magnetic atom with number  $\mathbf{g}$ ,  $\mathbf{S}_{\mathbf{g}}$  and  $\mathbf{s}$  are the spin operators of this atom and a conduction electron respectively, the quantity  $B(\mathbf{h})$  is the integral of transfer of an electron from the atom  $\mathbf{g}+\mathbf{h}$  to the atom  $\mathbf{g}$ , and  $U(\mathbf{g})$  is the potential of the defect. Below the third term in the Hamiltonian (1)—the term describing the  $s$ - $f$  exchange—will be treated as a perturbation. Therefore, as a preliminary, it is advisable to perform the canonical transformation that diagonalizes the  $s$ -electron part of the Hamiltonian (1), i.e., its first two terms:

$$a_{\mathbf{g}\sigma} = \sum_n \psi_n(\mathbf{g}) a_{n\sigma}, \quad \sum_{\mathbf{g}} \psi_n^*(\mathbf{g}) \psi_m(\mathbf{g}) = \delta_{mn}. \quad (2)$$

As a result of the transformation, the Hamiltonian (1) assumes the form:

$$\mathcal{H} = \sum_n \epsilon_n a_{n\sigma}^{\dagger} a_{n\sigma} - A \sum_{\mathbf{g}} \psi_n^*(\mathbf{g}) \psi_m(\mathbf{g}) (\mathbf{S}_{\mathbf{g}} \mathbf{s})_{\sigma\sigma'} a_{n\sigma}^{\dagger} a_{m\sigma'}. \quad (1a)$$

Bearing in mind the fact that we want to construct the magnetic Hamiltonian up to the terms  $\sim A^2$  inclusively, let us carry out that canonical transformation of the Hamiltonian (1a) which eliminates the terms  $\sim A$ , which are nondiagonal in the index,  $n$ , of the orbital state of the electron of the donor (the actual parameter of smallness will be indicated below). Using the well-known rules for the permutation of the spin operators in the terms  $\sim A^2$ , we obtain the following expression for the transformed Hamiltonian (the off-diagonal terms  $\sim A^2$  have been omitted):

$$\begin{aligned} \mathcal{H} &= \sum_n (\epsilon_n + \mathcal{H}_{2n}) a_{n\sigma}^{\dagger} a_{n\sigma} - \sum_n (\mathbf{K}_n \mathbf{s})_{\sigma\sigma'} a_{n\sigma}^{\dagger} a_{n\sigma'}, \\ \mathcal{H}_{2n} &= \sum_{\mathbf{g}, \mathbf{f}} \mathcal{B}_n(\mathbf{g}, \mathbf{f}) \mathbf{S}_{\mathbf{g}} \mathbf{S}_{\mathbf{f}}, \\ \mathcal{B}_n(\mathbf{g}, \mathbf{f}) &= \frac{A^2}{4} \sum_{m \neq n} \frac{\psi_n^*(\mathbf{g}) \psi_m(\mathbf{g}) \psi_m^*(\mathbf{f}) \psi_n(\mathbf{f})}{\epsilon_n - \epsilon_m}, \end{aligned} \quad (3)$$

$$\mathbf{K}_n = \sum_{\mathbf{g}} \mathcal{A}_n(\mathbf{g}) \mathbf{S}_{\mathbf{g}}, \quad \mathcal{A}_n(\mathbf{g}) = A W_{\mathbf{g}} - 2 \mathcal{B}_n(\mathbf{g}, \mathbf{g}), \quad W_{\mathbf{g}} = |\psi(\mathbf{g})|^2.$$

The eigenfunction of the Hamiltonian (3) is sought in the form

$$\Phi_n(S^+) = [\varphi_n(S^+) a_{n\uparrow}^+ + \chi_n(S^+) a_{n\downarrow}^+] |0\rangle_e, \quad (4)$$

where  $|0\rangle_e$  is the electron vacuum function,  $(\varphi, \chi)$  is the two-component wave function of the  $f$ -spins. The use of the formulas (3) and (4) allows the representation of the Schrödinger equation in the following form:

$$\begin{aligned} \frac{1}{2}K^+\varphi + (E - \frac{1}{2}K^2 - \mathcal{H}_2)\chi &= 0, \\ (E + \frac{1}{2}K^2 - \mathcal{H}_2)\varphi + \frac{1}{2}K^-\chi &= 0, \quad K^\pm = K^x \pm iK^y. \end{aligned} \quad (5)$$

Let us first consider the case of equivalent atoms, when the electron can be found with equal probability on any one of the  $\mathcal{N}$  atoms in the neighborhood of the defect, i.e.,  $W_g = 1/\mathcal{N}$ . Using the relation  $S^-F(S^+) = F(S^+ + 1)S^-$ , which follows from the definition of the operator  $S^-$ , and taking into account the fact that the Hamiltonian  $\mathcal{H}_2$  conserves the total spin,  $\mathcal{M}$ , of the system of  $\mathcal{N}$  atoms, to which the operator  $\mathbf{K}$  is proportional in the present case, we obtain from (5):

$$\begin{aligned} \left( E - \mathcal{H}_2 + \frac{\bar{A}}{2\mathcal{N}}\mathcal{M} \right) \varphi = \frac{\bar{A}^2}{4\mathcal{N}^2} \left[ E - \mathcal{H}_2 - \frac{\bar{A}}{2\mathcal{N}}(\mathcal{M} + 1) \right]^{-1} \mathcal{M}^{-1} \mathcal{M}^+ \varphi, \\ \bar{A} \approx A + \frac{A^2}{4} \sum_{n \neq m} \frac{|\psi_m(0)|^2}{\epsilon_n - \epsilon_m}. \end{aligned} \quad (6)$$

With allowance for the equality

$$\mathcal{M}^{-1}\mathcal{M}^+ = \mathcal{M}^2 - \mathcal{M}(\mathcal{M} + 1)$$

it follows from the formula (6) that

$$[E - \mathcal{H}_2 + \bar{A}/4\mathcal{N}]^2 \varphi = (\bar{A}/2\mathcal{N})^2 [\mathcal{M}^2 + \frac{1}{4}] \varphi.$$

Thus, we can introduce the magnetic Hamiltonian

$$\mathcal{H}_M = -\frac{\bar{A}}{2\mathcal{N}} \left[ \frac{1}{2} \pm \sqrt{\mathcal{M}^2 + \frac{1}{4}} \right] + \mathcal{H}_2, \quad (7)$$

whose eigenvalues furnish the energy spectrum of the system in question.

If we do not assume that the atoms are equivalent, then the construction of the magnetic Hamiltonian is possible only when the terms  $\sim W_g$ , i.e.,  $1/\mathcal{N}$ , are negligible compared to  $2S$ , where  $S$  is the spin of the magnetic atom (they arise upon the commutation of  $K^-$  and  $(E - K)^{-1}$  after the elimination of  $\chi$  from the second equation of the system (5)). The magnetic Hamiltonian is then given by the expression

$$\mathcal{H}_{Mn} = \mathcal{H}_{1n} + \mathcal{H}_{2n} + O(A^3), \quad \mathcal{H}_{1n} = \pm \left[ \sum_{g, f} \mathcal{A}_n(g) \mathcal{A}_n(f) S_g S_f \right]^{1/2}, \quad (7a)$$

a particular case of which is, with the specified degree of accuracy in  $1/2S\mathcal{N}$ , the Hamiltonian (7).

Evidently, the leading term,  $\mathcal{H}_1$ , in the Hamiltonian (7a) is an essentially non-Heisenberg Hamiltonian. The Heisenberg term,  $\mathcal{H}_2$ , which corresponds to the Ruderman-Kittel approximation, is a correction of the next order in  $A$ . Let us list the properties of the Hamiltonian  $\mathcal{H}_1$ .

1) The eigenfunction,  $\varphi$ , of the Hamiltonian  $\mathcal{H}_1$  is only one component of the two-component wave function,  $(\varphi, \chi)$ , of the magnetic sub-system (4). Therefore, in the subspace defined by the eigenfunctions,  $\varphi$ , of the Hamiltonian  $\mathcal{H}_1$ , the mean value of any spin operator  $\hat{L}$  should be computed with allowance for (5) from the formula

$$\bar{L} = \langle \varphi | \hat{D} \cdot \hat{L} | \varphi \rangle / \langle \varphi | \hat{D} \cdot 1 | \varphi \rangle, \quad (8)$$

where the deformation operator  $\hat{D}$  converts the operator  $\hat{L}$  into

$$\hat{D} \cdot \hat{L} = \hat{L} + \frac{1}{2} K^- (\mathcal{H}_M - \frac{1}{2} K^2)^{-1} \hat{L} (\mathcal{H}_M - \frac{1}{2} K^2)^{-1} K^+$$

(account has been taken of the fact that  $\chi$  and, consequently,  $K^+\varphi$  satisfy the same wave equation with the Hamiltonian (7a) as  $\varphi$ ).

In a state with a fixed energy, the squares of the moduli of the eigenfunctions,  $\varphi$  and  $\chi$ , of the Hamiltonian  $\mathcal{H}_1$ , (7a), do not individually have the meaning of the probability density of a definite spin configuration; this density is given by the sum  $|\varphi|^2 + |\chi|^2$ , where  $\varphi$  and  $\chi$  should be connected with each other by the relation (5). Therefore, in the general case the Hamiltonian  $\mathcal{H}_1$  should be treated not as the true, but as an equivalent, Hamiltonian. But in certain cases (as for example, in the spin-wave approximation (see below)) it is sufficient to consider only one of the components of the magnetic wave function  $(\varphi, \chi)$ , and then the Hamiltonian  $\mathcal{H}_1$  can be regarded as the true Hamiltonian.

2) The double sign of the Hamiltonian  $\mathcal{H}_1$  is a consequence of the fact that the Hamiltonian, by its physical meaning, is the energy of the coupling of the  $s$ -electron spin and the total spin,  $\mathcal{M}$ , of the magnetic  $f$ -atoms into a single spin equal either to  $\mathcal{M} + \frac{1}{2}$  or  $\mathcal{M} - \frac{1}{2}$ . Thus, to the two signs in the Hamiltonian  $\mathcal{H}_1$  correspond two possibilities: parallel or antiparallel orientation of the spin of the electron relative to the angular momentum  $\mathcal{M}$ .

3) The energy of the  $s$ - $f$  coupling depends on the magnitude of the angular momentum  $\mathcal{M}$ , but does not depend on its direction in space. Therefore, the Hamiltonian  $\mathcal{H}_1$  is invariant under a simultaneous rotation of the spins of all the magnetic atoms through one and the same angle (a property which must be possessed by any magnetic Hamiltonian in isotropic space).

4) The reality of the eigenvalues of the Hamiltonian  $\mathcal{H}_1 \sim \sqrt{K^2}$  is evident from the structure of the Hamiltonian.

5) Into the Hamiltonian  $\mathcal{H}_1$  enter all the invariants,  $(S_1 \cdot S_2)^{n_1} (S_3 \cdot S_4)^{n_3} (S_5 \cdot S_6)^{n_5} \dots$  ( $0 \leq n_i \leq 2S$ ), that are possible in an isotropic system. This follows from the definition of the algebraic function of the operator as the corresponding series

$$\begin{aligned} \left[ \sum_{g, f} \mathcal{A}_n(g) \mathcal{A}_n(f) S_g S_f \right]^{1/2} = (R+Q)^{1/2} = R^{1/2} \left[ 1 + \frac{Q}{2R} - \frac{Q^2}{8R^2} + \dots \right], \\ R = \sum_g \mathcal{A}_n^2(g) S(S+1), \quad Q = \sum_{g \neq f} \mathcal{A}_n(g) \mathcal{A}_n(f) S_g S_f. \end{aligned} \quad (9)$$

6) The interaction between the various atoms is not central, but multiplicative ( $\sim \alpha_n(\mathbf{g})\alpha_n(\mathbf{l})$ ). If the electron of the donor is in the ground orbital state, when  $\psi(\mathbf{g})$  has the point-group symmetry of the crystal, then the spin of the atom  $\mathbf{g}$  interacts equally intensely with all the atoms located symmetrically about the positively charged defect, irrespective of the distance between them and the atom  $\mathbf{g}$ .

7) Since the radicand in the Hamiltonian  $\mathcal{H}_1$  has its maximum value when all the spins are directed in like manner, the indirect exchange described by the Hamiltonian  $\mathcal{H}_1$  strives, in the case of the energetically advantageous direction of the spin of the  $s$ -electron relative to the angular momentum  $\mathfrak{M}$  moment, to establish a ferromagnetic order.

Let us now find out the conditions of applicability of the expression (7a). It is not difficult to verify that in the case of a complete ferromagnetic order (all  $S_g = S$ ) the terms of first order in  $A$  in (7a) differ from the exact result for the  $s$ - $f$  exchange energy only by a quantity  $\sim 1/2S\mathfrak{M}$ , while the terms  $\sim A^2$  with allowance for the orthonormality condition, (2), for the functions  $\psi_n(\mathbf{g})$  vanish. When, however, the ferromagnetic order is completely destroyed, the terms  $\sim A$  predominate if the inequalities  $AS \ll 10\mathfrak{M}^{1/2}(\epsilon_{2s} - \epsilon_{1s})$ ,  $w\mathfrak{M}^{-1/2}$ , where  $w$  is the conduction-band width, are fulfilled. Thus, the condition  $\epsilon_{2s} - \epsilon_{1s} \gg AS$  is sufficient, but not necessary for the applicability of the above-obtained results.

### 3. THE MAGNON SPECTRUM IN THE CASE OF RADICAL EXCHANGE

In order to understand better the characteristics of the indirect exchange described by the Hamiltonian  $\mathcal{H}_1$ , (7a), it is advisable to investigate, as a preliminary, the magnon spectrum of the model system in which there are no other exchange-interaction mechanisms. Going over in (7a) from the  $f$ -spin operators to the magnon operators

$$\begin{aligned} S_i^z &= S - b_i^+ b_i, & S_i^+ &= S_i^z + iS_i^y = (2S)^{1/2} b_i, \\ S_i^- &= S_i^z - iS_i^y = (2S)^{1/2} b_i^+ \end{aligned} \quad (10)$$

and limiting ourselves to the first approximation in  $1/S$ , we obtain the magnon Hamiltonian in the form (we consider only the ground state of the electron)

$$\mathcal{H}_{md} = \frac{A}{2} \sum_{\mathbf{g}, \mathbf{l}} W_{\mathbf{g}\mathbf{l}} (b_{\mathbf{g}}^+ b_{\mathbf{l}} - b_{\mathbf{l}}^+ b_{\mathbf{g}}). \quad (11)$$

In (11), the  $s$ - $f$  exchange integral is, for definiteness, assumed to be positive. Representing the wave function in the form

$$\varphi_{\epsilon} = \sum_{\mathbf{g}} c_{\mathbf{g}} b_{\mathbf{g}}^+ |0\rangle, \quad (12)$$

where  $|0\rangle$  is the vacuum magnon function, we obtain a system of equations for the determination of the coefficients  $c_{\mathbf{g}}$ :

$$(W_{\mathbf{g}} - \epsilon)c_{\mathbf{g}} - W_{\mathbf{g}} \sum_{\mathbf{h}} W_{\mathbf{h}\mathbf{g}} c_{\mathbf{h}} = 0, \quad \epsilon = \frac{2\omega}{A}. \quad (13)$$

The system of equations (13) can be solved easily. From it follows the equality

$$\left[ 1 - \sum_{\mathbf{g}} \frac{W_{\mathbf{g}}^2}{W_{\mathbf{g}} - \epsilon} \right] \sum_{\mathbf{h}} W_{\mathbf{h}\mathbf{g}} c_{\mathbf{h}} = 0. \quad (14)$$

The first factor in (14) can vanish only when  $\epsilon = 0$ . The corresponding solution has the form  $c_{\mathbf{h}} = \text{const}$ . For  $\epsilon > 0$  the second factor should vanish, i.e., the magnon spectrum is given by the relation

$$\epsilon = W_{\mathbf{g}}, \quad (15)$$

the coefficients  $c_{\mathbf{h}}$  in (12) being nonzero for those atoms with which the electrons remain with a probability equal to  $W_{\mathbf{g}}$ .

As an example, let us consider a defect in a simple cubic crystal, assuming that the electron wave function is nonzero only on the six nearest neighbors of the defect (for them  $W_{\mathbf{g}} = 1/6$ ). Then the system has a magnon level with  $\epsilon = 0$  and a completely symmetric wave function

$$\varphi_{\epsilon} \sim \sum_{\Delta} (b_{\Delta}^+ + b_{-\Delta}^+) |0\rangle,$$

where  $|0\rangle$  is the vacuum function and the index  $\Delta$  runs through the values  $\mathbf{x} = (a, 0, 0)$ ,  $\mathbf{y} = (0, a, 0)$ , and  $\mathbf{z} = (0, 0, a)$ . Besides the indicated level, there also exists a fivefold degenerate level with  $\epsilon = 1/6$ ; to it corresponds the three  $p$ -type wave functions:  $\varphi_p \sim (b_{\Delta}^+ - b_{-\Delta}^+) |0\rangle$ , and the two  $d$ -type wave functions:  $\varphi_d \sim (b_{\Delta}^+ + b_{-\Delta}^+ - b_{\Delta'}^+ - b_{-\Delta'}^+) |0\rangle$ . Notice that in the case of an exponential law of decrease at infinity ( $\psi(\mathbf{g}) \sim \exp(-\alpha|\mathbf{g}|)$ ), the magnon spectrum contains arbitrarily low frequencies, and the frequency  $\epsilon = 0$  is the limiting point of a set of frequencies.

The existence of a fully symmetric magnon with zero frequency is not connected with the specific character of the interaction between the magnetic atoms, but is, according to a theorem on broken symmetry, a universal property of isotropic systems. In the case under consideration this property can be substantiated in the following manner. As is well known, the creation of a fully symmetric magnon implies that the total angular momentum,  $\mathfrak{M}$ , of the  $f$ -spins can be rotated without changing its magnitude (this can be easily verified with the aid of the formulas (10) and (12)). On the other hand, in the situation under consideration the spin of the  $s$ -electron and the angular momentum  $\mathfrak{M}$  are always collinear, and therefore the former, when it is rotated, should be rotated through the same angle. But this implies a rotation of the total moment of the " $f$ -spins +  $s$ -electron" system without a change in its magnitude  $\mathfrak{M} \pm \frac{1}{2}$ . It is clear that in isotropic space the energy of the system does not change in such a rotation.

The spectrum of the not fully symmetric magnons, (15), formally coincides with the change,  $W_{\mathbf{g}}$ , in the energy of an isolated magnetic atom in a magnetic field when the projection of its spin decreases by unity. However, it does not at all follow from this that the effect of the  $s$ - $f$  exchange on the magnetic-atom state described by the Hamiltonian  $\mathcal{H}_1$ , (7a), amounts to the

action of some effective magnetic field on the system of mutually noninteracting  $f$ -spins. In the first place, such a field would make the space anisotropic, and in it the frequency of the fully symmetric magnon would turn out to be nonzero. Secondly, the energy (15) is actually connected not with the declination of the spin localized on one atom, but with a collective declination that is passed on from atom to atom, i.e., the interaction between the atoms plays a fundamental role. Indeed, if the magnon were localized on the atom  $g$  (i.e., if  $c_g = \delta_{g0}$ ), then according to (13) the energy  $\epsilon$  would be equal to  $W_g - W_g^2$ . The addition of  $-W_g^2$  to the energy  $W_g$  is due to the fact that the deflection of the spin  $S_g$  led to a change in the direction of the total spin of the atoms and, consequently, in the direction of the spin of the donor's electron, which spin is always directed along the total moment. Of course, such a localized state cannot be an eigenstate, since its wave function is not orthogonal to the wave function of the magnon with  $\epsilon = 0$  (when  $c_g = \delta_{g0}$  the equality

$$\sum_{\mathbf{q}} c_{\mathbf{q}} = 0,$$

which expresses the condition for their orthogonality, is not fulfilled).

The complete description of the magnon state will be given if, besides  $\varphi$ , we also find the second component,  $\chi$ , of the magnetic wave function (4), which corresponds to the opposite orientation of the spin of the  $s$ -electron. Using the formulas (5), (10), and (12), we obtain:

$$\chi = \frac{1}{(2S)^{1/2}} \sum_{\mathbf{q}} W_{\mathbf{q}} c_{\mathbf{q}} |0\rangle. \quad (15a)$$

According to (15a), for the fully symmetric magnon state  $\varphi_s$  the component  $\chi_s$  is nonzero, but it makes only a small contribution ( $\sim 1/2S\mathfrak{N}$ ) to the operator averages (8). That  $\chi_s$  is nonzero follows from the fact that the spin of the  $s$ -electron rotates together with the total moment of the system, so that its component along the original direction should decrease. The smallness of  $\chi_s$  is a consequence of the smallness of the angle through which the moment ( $\sim \mathfrak{N}S \gg 1$ ) should rotate in order for its projection to change by unity.

For the not fully symmetric magnons the component  $\chi$ , (15a), in accordance with the formula (14), vanishes exactly. This result follows from the fact that the creation of a not fully symmetric magnon, in contrast to the creation of a fully symmetric one, implies the decrease of the total moment of the  $f$ -atoms by unity. Since its projection decreases simultaneously by unity, its direction in space does not change. Therefore, the direction of the spin of the  $s$ -electron does not also change.

The above-obtained results, according to which in the spin-wave approximation the component  $\chi$  of the wave function is either equal to zero, or small compared to  $\varphi$ , imply that in this approximation the equivalent Hamiltonian  $\mathcal{H}_1$ , (7a), can be regarded as the true Hamiltonian.

#### 4. THE MAGNON SPECTRUM OF A FERROMAGNET WITH SINGLY CHARGED DONORS

In this section we shall investigate the effect of singly-charged donors on the magnon spectrum of ferromagnetic semiconductors. In the Hamiltonian (7a) we shall consider only the radical term  $\mathcal{H}_1$ , which is linear in  $A$ . We shall first of all consider the simplest model of a donor with a small radius: the  $s$ -electron is, in the main, localized on  $z$  nearest neighbors of the defect, which replaces the normal atom with index zero. Besides the indirect exchange via the donor's electron, there also exists a direct ferromagnetic exchange between the magnetic atoms. The introduction of the defect changes it too. The effect of this circumstance on the magnon spectrum is investigated in detail in Ref. 8, and will not be considered here.

Performing a Fourier transformation of the  $f$ -magnon operators in the Hamiltonian (11):

$$b_{\mathbf{q}} = N^{-1/2} \sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}} b_{\mathbf{r}},$$

we can write the Hamiltonian of a ferromagnet with a defect in the form

$$\mathcal{H} = \sum_{\mathbf{q}} \omega_{\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} \mp \frac{|A|}{2N} \sum_{\mathbf{q}\mathbf{q}'} (\gamma_{\mathbf{q}-\mathbf{q}'} - \gamma_{\mathbf{q}} \gamma_{\mathbf{q}'}) b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}'} \pm \frac{AS}{2}, \quad (16)$$

$$\omega_{\mathbf{q}} = J(1 - \gamma_{\mathbf{q}}), \quad \gamma_{\mathbf{q}} = \frac{1}{z} \sum_{\Delta} e^{i\Delta\cdot\mathbf{q}},$$

where  $\Delta$  is the vector joining an atom with its nearest neighbors.

The first term in (16) describes the  $f$ -magnons in a perfect crystal; the second, the effect of the defect. Its structure differs essentially from the structure of the Hamiltonian investigated in Refs. 7 and 8 by the non-central nature of the interaction of the spins of the various atoms. In the expression (16), as in (7a), both signs are retained. We shall assume, for definiteness, that  $A > 0$ ; then the upper sign corresponds to the energetically disadvantageous orientation of the spin of the  $s$ -electron relative to the local moment. In this state the indirect exchange tends to destroy the ferromagnetic order in the vicinity of the defect, but in the case of a sufficiently strong direct exchange the ferromagnetic order can still turn out to be stable. A change in the direction of the  $s$ -spin constitutes the creation of a high-frequency localized  $s$ -magnon with energy  $AS$ , that does not modify the local ferromagnetic order. However, its appearance softens the localized  $f$ -magnons (the distinctive  $s$ - $f$ -magnon-magnon interaction). If, however, the direct exchange is weak, and some of the  $f$ -magnon frequencies become negative upon the excitation of the high-frequency  $s$ -magnon, then the local ferromagnetic order should be replaced by an anti-ferromagnetic order.

To find the  $f$ -magnon spectrum, we construct the equal-time Green function

$$G_{\mathbf{k}\mathbf{q}} = -i \langle 0 | b_{\mathbf{k}} b_{\mathbf{q}}^{\dagger} - b_{\mathbf{q}}^{\dagger} b_{\mathbf{k}} | 0 \rangle \theta(t).$$

After the Fourier transformation of the equation of motion for  $G_{\mathbf{k}\mathbf{q}}$ , found with the aid of the Hamiltonian (16), we obtain the following integral equation for the determination of the function (the magnetic-atom lattice is assumed to be a simple-cubic lattice with constant  $a$ ):

$$(\omega - \omega_{\mathbf{k}})G_{\mathbf{k}\mathbf{q}} = \delta_{\mathbf{k}\mathbf{q}} \mp \frac{A}{6N} \sum_{\mathbf{p}} \left[ \sum_{i=x,y,z} (\cos k_i a \cos p_i a + \sin k_i a \sin p_i a) - \frac{1}{3} \sum_{i,j=x,y,z} \cos k_i a \cos p_j a \right] G_{\mathbf{p}\mathbf{q}}. \quad (17)$$

Since the nucleus of Eq. (17) is degenerate, it is not difficult to obtain its exact solution:

$$G_{\mathbf{k}\mathbf{q}} = \frac{\delta_{\mathbf{k}\mathbf{q}}}{\omega - \omega_{\mathbf{q}}} \mp \frac{A}{2N(\omega - \omega_{\mathbf{k}})(\omega - \omega_{\mathbf{q}})} \left\{ \frac{1}{1 \pm AR} \times \left[ \frac{1}{3} \sum_{\mathbf{r}} \cos k_i a \cos q_i a - \gamma_{\mathbf{k}\mathbf{q}} \right] + \frac{1}{3(1 \pm AP)} \sum_{\mathbf{r}} \sin k_i a \sin q_i a \right\}, \quad (18)$$

where we have adopted the notation

$$P = \frac{1}{6N} \sum_{\mathbf{k}} \frac{\sin^2 k_x a}{\omega - \omega_{\mathbf{k}}},$$

$$R = \frac{1}{6N} \sum_{\mathbf{k}} \frac{\cos^2 k_x a - \cos k_x a \cos k_y a}{\omega - \omega_{\mathbf{k}}}.$$

As can be seen from the formula (18), the Green function has, besides the poles corresponding to a perfect crystal, poles at the points determined by the equalities

$$1 \pm AP = 0, \quad 1 \pm AR = 0. \quad (19)$$

From Eqs. (19), it is not difficult to find the conditions for the existence of localized  $f$ -magnon levels. Using the numerical values of the integrals  $P$  and  $R$  given in Ref. 8, we find that localized magnon levels are possible if the Curie temperature of the crystal,  $T_c \approx J(S+1)/3$ , is less than  $0.03A(S+1)$ . For  $AS \sim 0.5$  eV, this implies that localized magnons exist when  $T_c < 150$  K. Naturally, if the spin of the  $s$ -electron is oriented along the moment of the crystal, then the localized levels lie above the magnon band, i.e., they have frequencies higher than  $2J$ . In the case when the  $s$ -electron spin is oriented along the opposite direction the frequencies of the localized  $f$ -magnons turn out to be negative. If, on the other hand, the conditions for the existence of localized levels above the magnon band are not fulfilled and the defect creates only virtual levels near the top of the band, then the inversion of the spin of the electron leads only to the appearance of virtual magnon levels near the bottom of the band. However, the frequencies of the  $f$ -magnons remain positive, i.e., the ferromagnetic order is maintained in the event of the creation of an  $s$ -magnon. In the  $J \rightarrow 0$  limit Eqs. (19) yield the results of the preceding section.

Let us now consider the other limiting case: a donor of large radius. Substituting the wave function (12) into the Hamiltonian (11) with the direct-exchange Hamiltonian added to it, we obtain

$$JS \sum_{\Delta} (c_{\mathbf{k}} - c_{\mathbf{k}+\Delta}) \mp \frac{A}{2} \sum_{\mathbf{r}} W_{\mathbf{r}} W_{\mathbf{r}} (c_{\mathbf{k}} - c_{\mathbf{r}}) = \omega c_{\mathbf{k}}. \quad (20)$$

We shall be interested in the magnon levels near the top of the magnon band. The dominant contribution to their wave functions is made by the wave vectors close to  $\Pi = (\pi/a, \pi/a, \pi/a)$ , at which the frequency  $\omega_{\mathbf{k}}$  attains its maximum value. Therefore, in analogy to the effective-mass method in the theory of electron levels in semiconductors, the coefficients  $c_{\mathbf{g}}$  can be represented in the form

$$c_{\mathbf{g}} = \varphi_{\mathbf{g}} e^{i\Pi\mathbf{g}}, \quad (21)$$

where  $\varphi_{\mathbf{g}}$  is a slowly varying function of the coordinate,  $\mathbf{g}$ , of the magnetic atom. Expanding formally the quantity  $\varphi_{\mathbf{g}+\Delta}$  in a power series in the vector  $\Delta$ , we obtain from (20) and (21) (in (20) we take only the lower sign) the equation

$$JSa^2 \Delta \varphi_{\mathbf{g}} + \frac{A}{2} W_{\mathbf{g}} \varphi_{\mathbf{g}} - \frac{A}{2} W_{\mathbf{g}} e^{i\Pi\mathbf{g}} \sum_{\mathbf{r}} W_{\mathbf{r}} e^{i\Pi\mathbf{r}} \varphi_{\mathbf{r}} = (\omega - 2J) \varphi_{\mathbf{g}}. \quad (22)$$

The last term on the left-hand side of (22) is small, since in the sum over  $\mathbf{r}$  the slowly varying function of the coordinates,  $W_{\mathbf{r}} \varphi_{\mathbf{r}}$ , is multiplied by a rapidly oscillating factor. Therefore, this term will be neglected in the subsequent analysis.

In the hydrogen-like donor model the quantity  $W_{\mathbf{g}}$  is given by the expression

$$W_{\mathbf{g}} = \frac{1}{\pi} \left( \frac{a}{R} \right)^3 \exp \left( -\frac{2|g|}{R} \right), \quad (23)$$

where  $R$  is the radius of the orbit. If in Eq. (22) we replace the signs of all the terms by the opposite signs, then the corresponding problem becomes identical with the problem of the capture of a particle with mass  $M = (2JSa^2)^{-1}$  by the exponential potential well  $V_0 \exp(-r/r_0)$ . Its solution is well known (see Calogero's book).<sup>[9]</sup> In particular, the first level in the well appears when  $2Mr_0^2 V_0 > 1.44$ . Interpreting this condition to fit the case (22), (23) under consideration here, we obtain the condition for the existence of localized magnons above the top of the magnon band:

$$A(S+1)/T_c > 18R/a. \quad (24)$$

Thus, for a given  $AS$  value and a given Curie temperature  $T_c$ , the larger the radius of the orbit of the donor is, the more difficult it is to satisfy the condition for the existence of localized magnon levels in the vicinity of the donor defect. Physically, this is due to the fact that, as  $R$  increases, the height of the potential hump for the magnon in the vicinity of the defect decreases like  $R^{-3}$ , and the effect due to the decrease of the height of the hump exceeds the effect due to the increase in its radius. For  $AS \sim 0.5$  eV and  $R \approx 4a$ , localized magnons are possible only in semiconductors with  $T_c$  lower than 70 K. This condition is satisfied by almost all the rare-earth semiconductors.

For a second bound state to appear in the exponential well, it is necessary that the inequality  $2Mr_0^2 V_0 > 6.1$  be satisfied,<sup>[9]</sup> i.e., the condition

$$A(S+1)/T_c > 76R/a. \quad (25)$$

should be fulfilled.

For the above-indicated values of the parameters a second localized magnon level is possible only when  $T_c < 20$  K. Hence, we can, in particular, conclude that the donor states of large radius that arise upon the doping of EuO ( $T_c = 69$  K) with gadolinium can lead to the appearance of only one localized magnon on each donor atom.

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<sup>1)</sup>This Hamiltonian should be interpreted not as a true Hamiltonian, but as an equivalent one, since its eigenvalues give the correct spectrum, but the probability density of a definite spin configuration is, generally speaking, given not by the square of the modulus of its eigenfunction, but by a more

complicated expression.

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## Spin waves in amorphous and finely divided ferromagnets with allowance for dipole-dipole interaction

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Complex dispersion relations are obtained, in the long-wave approximation, for spin waves in a ferromagnet with parameters that fluctuate randomly in space (for the corresponding results without allowance for dipole-dipole interaction, see V. A. Ignatchenko and R. S. Iskhakov, [Sov. Phys. JETP **45**, 526 (1977)]). When the exchange constant  $\alpha$  fluctuates, allowance for dipole-dipole interaction shifts the break in the modified dispersion law toward longer waves and leads to a change in the damping law for long spin waves: when  $k$  is less than a certain critical value  $k_c$ , the damping  $\omega'' \sim k^7$ ; for  $k > k_c$ , we get  $\omega'' \sim k^5$ . When the axis of magnetic anisotropy fluctuates, allowance for dipole-dipole interaction modifies both channels of interaction of the random inhomogeneity function  $\rho(\mathbf{r})$  with the spin wave  $\mathbf{m}(\mathbf{r}, t)$  and leads to the appearance of a new channel of interaction of  $\mathbf{m}$  with  $\rho$  via the stochastic magnetostatic fields produced by the stochastic magnetic structure. The dispersion law now contains a characteristic wave number for dipole-dipole interaction,  $k_M = (4\pi/\alpha)^{1/2}$ .

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### INTRODUCTION

In an earlier paper,<sup>[1]</sup> we calculated the modification of the dispersion relation and of the damping for long ( $ka \ll 1$ , where  $a$  is the lattice parameter) spin waves in a medium with either isotropic or anisotropic inhomogeneities, having an arbitrary correlation radius  $r_0$ . The phenomenological theory of spin waves developed in Ref. 1 is correct both for finely divided and for amorphous ferromagnets. Because there is at present no systematic theory of amorphous magnetism, the correlation radius  $r_0$  of the fluctuations of the corresponding parameter (the exchange parameter or the amount and direction of the anisotropy) cannot so far be calculated theoretically and occurs in the theory as a phenomenological constant. Therefore particular interest attaches to results that predict from what experimental observations  $r_0$  can be determined. In particular, a possible basis for determination of  $r_0$  might be the experimental observation of the characteristic break in the dispersion curve  $\omega(k^2)$  at  $k = \frac{1}{2}r_0$ , which was ob-

tained in Ref. 1 for the case of fluctuations of the exchange constant  $\alpha$ .

When there are spatial fluctuations of the axis of magnetic anisotropy (this phenomenon may be characteristic of certain classes of amorphous magnets), there must occur in the material a stochastic static magnetic structure whose correlation properties are determined by a magnetic-field-dependent correlation radius  $r_H = (\alpha M/H)^{1/2}$ . This leads to the result that the inhomogeneities interact with the spin waves by two paths: directly, and through the stochastic magnetic structure. In the dispersion relation both characteristic radii,  $r_0$  and  $r_H$ , occur.

Both in Ref. 1 and in all works known to us on this topic, dipole-dipole interaction was neglected. Allowance for this interaction is the purpose of the present paper.<sup>[1]</sup> Formally, the problem reduces to supplementing the effective magnetic field of Ref. 1 with a field  $\mathbf{H}_m(\mathbf{r}, t)$  determined by the equations of magnetostatics (we neglect effects of propagation of electro-