

Theory of antiferromagnetic impurities in magnets

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Local levels of a weakly coupled antiferromagnetic impurity with spin S' in a Heisenberg ferromagnet or ferrimagnet are considered. A method for treating such problems is proposed and is valid for all temperatures T for which the spin-wave approximation is applicable for the matrix. All the $2S'+1$ impurity levels are found and turn out to be nonequidistant. The nonequidistance of the levels leads to oscillations of the specific heat and susceptibility on variation of the magnetic field H , and also to a distinctive dependence of the ferromagnetic resonance lineshape on T and H . The possibilities of experimental observation of the effects considered are discussed.

1. INTRODUCTION

The effect of magnetic impurities on the properties of magnetically ordered crystals has been studied in a number of works; cf., e.g., [1-3]. The introduction of an impurity alters the spectrum of the magnetic excitations, leading, in particular, to the appearance of local or quasi-local magnon levels. The experimental study of these levels gives valuable information about the magnetic interactions [4]. The presence of impurities can also have an appreciable effect on the thermodynamics of the magnet, leading to unusual dependences of the magnetization and specific heat on the field H and temperature T [1,3,5].

The spectra of antiferromagnetic impurities in a ferromagnet have been discussed earlier; cf. [1,3,6,7]. Figure 1 illustrates features of the spectrum of these impurities. The antiferromagnetic character of the interaction of the impurity with a spin of the matrix (the exchange integral $J' > 0$) leads to the result that an antiparallel "Néel" alignment of the impurity spin S' with respect to the matrix spins S is energetically favorable. Therefore, the energy of the impurity states should increase with increase of the projection m' of the impurity spin along the direction of the magnetization (for more detail, see below). Then, if we neglect relativistic interactions that do not conserve the total moment M_Z , for low T all $2S'+1$ impurity spin levels will be stable: decay of any of them, with a transition to a level of smaller projection m' and emission of a spin wave with $\Delta m_Z = -1$, is (like the inverse absorption process) forbidden by the energy and momentum conservation laws. Thus, at $T=0$ all the states under consideration are strictly localized, irrespective of the magnitude of the interaction constants, whereas, e.g., for a weakly coupled ferromagnetic impurity the levels are quasi-local and have width even at $T=0$ [1,2].

The problem of the spectrum of the one-magnon excitations of a Heisenberg ferromagnet with an isolated antiferromagnetic impurity was solved exactly by Wang and Callen [3], but only for the impurity-spin value $S' = 1/2$. In the case of the one-dimensional Heisenberg model, two-magnon excitations have also been studied [6,7]. Izyumov and Medvedev [1] have considered the spectrum and the thermodynamic contribution of antiferromagnetic impurities of low concentration for all S' . However, they used the spin-wave approximation (the lowest approximation of the Holstein-Primakoff method), and so their results are valid only for small deviations from the above-mentioned "Néel" state. In particular, this limitation prevents us from studying the energies of higher excited states of the impurity ($2S'$

+ 1)-multiplet, and also from treating quantitatively the effects associated with appreciable change (under the influence of T or H) in the occupation of these states. At the same time, the characteristic thermodynamic effects associated with the given impurities appear in precisely that region of T and H in which the state of the impurities begins to deviate appreciably from the Néel state [1,3]. It is therefore of fundamental interest to go beyond the framework of the spin-wave approximation here.

The present work has the aim of elucidating the qualitative features of the spectrum of all the $2S'+1$ local levels of an antiferromagnetic impurity in a magnet, and their effect on the thermodynamics for a low concentration of impurities, when their mutual interaction can be neglected. We shall use the Heisenberg model with nearest-neighbor interaction, and for simplicity shall consider principally the case of a weakly coupled impurity $J' \ll J$, where J' and $-J$ are the exchange integrals for the impurity and the spins of the matrix respectively. The temperature will be assumed to be low compared with the Curie point of the pure substance: $T \ll T_C \sim J$, but the relation between T and J' can be arbitrary. Under these conditions, the deviations of the matrix spins from saturation are small, and for them, as in the spin-wave approximation, we can use the Maleev-Dyson transformation [8] to Bose operators. Then, since for the local levels of interest to us these deviations $\delta S = S - S_z$ are usually, even for large J'/J , not large (cf. [1] and below), the approximation used in practice and the results for the spectrum are evidently also valid for $J' \gtrsim J$. On the other hand, the deviations of the impurity state from the Néel state (for which $m' = -S'$) can be large, so that the states of the impurity must be described exactly. As a result, we arrive at the problem of a localized spin interacting with a boson field of magnons, and to solve this problem we can use the usual methods of perturbation theory.

The principal result obtained when this interaction is taken into account is the unequal spacing of the $2S'+1$ impurity energy levels: as we move further away from the ground state, the spacing between the levels increases. This leads to a number of unusual features in the behavior of an impurity magnet in a magnetic field. In particular, oscillations in the specific heat and susceptibility of such a system with variation of the field H , and also a distinctive dependence of the ferromagnetic



FIG. 1

resonance shape on H and T, should be observed. The temperature dependences of the magnetization and specific heat are also considered.

In Sec. 5, the results are generalized to the case of a ferromagnet. It is shown that, in accordance with the remarks in [1], the principal results remain valid if the spin of the impurity in the ground state is oriented against the total magnetization of the crystal. Otherwise, as in the case of weakly coupled impurities of other types (a ferromagnetic impurity in a ferromagnet, or impurities in an antiferromagnet), the levels become quasi-local and there are no oscillations of the susceptibility and specific heat in a magnetic field, although temperature anomalies are still possible. In the conclusion, we briefly discuss the correspondence with more realistic models of magnets and also the possibilities of experimental observation of the effects considered.

2. LOCAL LEVELS OF AN ANTIFERROMAGNETIC IMPURITY IN A FERROMAGNET

We shall consider a Heisenberg ferromagnet with spin S and nearest-neighbor interaction J. Let there be a substitutional impurity with spin S' at the point r=0. Then the Hamiltonian has the form

$$\mathcal{H} = \frac{1}{2} \sum_{\mathbf{r}} \sum_{\mathbf{r}'} J_{\mathbf{r}\mathbf{r}'} (S_{\mathbf{r}}^+ S_{\mathbf{r}'}^+ + S_{\mathbf{r}}^- S_{\mathbf{r}'}^-) - H \sum_{\mathbf{r}} g_{\mathbf{r}} S_{\mathbf{r}}^z. \quad (1)$$

Here $S_{\mathbf{r}}^{\pm} = S_{\mathbf{r}}^x \pm iS_{\mathbf{r}}^y$, and the coordinates \mathbf{r} and \mathbf{r}' label the lattice sites. The exchange integral $J_{\mathbf{r}\mathbf{r}'}$ is non-zero only when \mathbf{r} and \mathbf{r}' are nearest neighbors, and equals $-J$ for $\mathbf{r}, \mathbf{r}' \neq 0$ and J' when \mathbf{r} or \mathbf{r}' equals zero; $g_{\mathbf{r}}$ denotes the gyromagnetic ratio, equal to g for the spins of the matrix and g' for the impurity. For definiteness, we shall assume the lattice to be a cubic Bravais (simple, bcc or fcc) and for simplicity in the formulas we shall first make the factors g and g' equal: $g' = g$.

As noted in [1-3], the eigenstates of the system are characterized by a well-defined value of the z-component M^z of the total spin, taking the values $M_{\max}^z, M_{\max}^z - 1, M_{\max}^z - 2, \dots$, where $M_{\max}^z = (N-1)S + S'$, N being the total number of spins. We shall denote the eigenfunctions corresponding to the component $M^z = M_{\max}^z - m$ by ψ_m ($m = 0, 1, 2, \dots$), and the energy levels by E_m . Then for the function ψ_0 describing the completely aligned state, we have

$$S_{\mathbf{r}}^+ \psi_0 = S_{\mathbf{r}} \psi_0, \quad S_{\mathbf{r}}^+ \psi_0 = 0, \quad (2)$$

where the magnitude of $S_{\mathbf{r}}$ equals S for $\mathbf{r} \neq 0$ and S' for $\mathbf{r} = 0$.

The state ψ_1 , for which $M^z = M_{\max}^z - 1$, was considered in detail in [1-3]; we shall call this state a one-particle state. The wave function of this state can be written in the form

$$\psi_1 = \sum_{\mathbf{r}} \varphi_{\mathbf{r}} S_{\mathbf{r}}^- \psi_0. \quad (3)$$

Then the normalization condition has the form

$$\sum_{\mathbf{r}} 2S_{\mathbf{r}} |\varphi_{\mathbf{r}}|^2 = 1. \quad (4)$$

Substituting (2) into the Schrödinger equation, we obtain an equation for the function $\varphi_{\mathbf{r}}$:

$$(\epsilon_1 - gH) \varphi_{\mathbf{r}} = - \sum_{\mathbf{r}'} S_{\mathbf{r}} J_{\mathbf{r}\mathbf{r}'} (\varphi_{\mathbf{r}} - \varphi_{\mathbf{r}'}), \quad (5)$$

where $\epsilon_1 = E_1 - E_0$, $E_0 = -\frac{1}{2} zJS^2(N-2) + zJ'SS' - gH[(N-1)S + S']$, and z is the number of nearest neighbors, equal to 6, 8, and 12 for the simple cubic, bcc and fcc lattices respectively. From (5) it is easy to obtain an equation for the energy ϵ_1 [1-3]:

$$\frac{J'}{J} + \rho \frac{\epsilon_1 - gH}{zSJ} - (\epsilon_1 - gH) \left[\frac{J'}{J} \left(1 - \frac{S'}{S} \right) + \rho \frac{\epsilon_1 - gH}{zSJ} \right] \sum_{\mathbf{k}} \frac{1}{\epsilon_1 - gH - \omega_{\mathbf{k}}} = 0, \quad (6)$$

where

$$\rho = 1 + \frac{J'S'}{JS}, \quad \omega_{\mathbf{k}} = zSJ \left(1 - \frac{1}{z} \sum_{\Delta} e^{i\mathbf{k}\Delta} \right),$$

and Δ are the coordinate differences $\mathbf{r}' - \mathbf{r}$, determined by the lattice structure, for the z nearest neighbors of the site \mathbf{r} .

As noted in [1-3], for an antiferromagnetic impurity $J' > 0$ there are local levels with $\epsilon_1 < 0$ (for $H=0$) in the system. We shall give expressions for ϵ_1 and the function $\varphi_{\mathbf{r}}$ in the limiting cases of weakly and strongly coupled impurities for the lowest, most symmetric, so-called s-state [1-3]:

For $J' \ll J$,

$$\epsilon_1 = gH - zSJ' - zS \frac{(J')^2}{J} (I_W - 1); \quad (7a)$$

$$\varphi_0^2 = \frac{1}{2S'}, \quad \varphi_{\Delta} \approx -\varphi_0 \frac{J'S'}{JS} (I_W - 1);$$

For $J' \gg J$,

$$\epsilon_1 = gH - zSJ' \left(1 + \frac{S'}{zS} \right); \quad (7b)$$

$$\varphi_0^2 = \frac{1}{2S'(1 + S'/zS)}, \quad \varphi_{\Delta} = -\varphi_0 \frac{S'}{zS}.$$

Here φ_{Δ} are the values of $\varphi_{\mathbf{r}}$ at the sites adjacent to the impurity, and I_W is the Watson integral, equal to 1.516, 1.393 and 1.345 for the simple cubic, bcc and fcc lattices respectively.

We now consider the many-particle states ψ_m and, in accordance with what has been said above, we shall be interested in the lowest local levels with $m \leq 2S'$. We write the wavefunction ψ_m in a form analogous to (3):

$$\psi_m = \sum_{\mathbf{r}_1, \dots, \mathbf{r}_m} \varphi_{\mathbf{r}_1, \dots, \mathbf{r}_m} S_{\mathbf{r}_1}^- \dots S_{\mathbf{r}_m}^- \psi_0. \quad (8)$$

From the Schrödinger equation we obtain the following equation for $\varphi_{\mathbf{r}_1, \dots, \mathbf{r}_m} \equiv \varphi_{1, \dots, m}$:

$$(\epsilon_m - mgH) \varphi_{1, \dots, m} = - \sum_{\mathbf{r}} S_{\mathbf{r}} [J_{\mathbf{r}1} (\varphi_{12, \dots, m} - \varphi_{2, \dots, m}) + \dots \quad (9)$$

$$+ J_{\mathbf{r}m} (\varphi_{1, \dots, m-1, m} - \varphi_{1, \dots, m-1, \mathbf{r}})] - \frac{1}{2} \{ J_{12} [\varphi_{113, \dots, m} + \varphi_{223, \dots, m} - 2\varphi_{123, \dots, m}] + J_{13} [\varphi_{121, \dots, m} + \varphi_{223, \dots, m} - 2\varphi_{123, \dots, m}] + \dots + J_{23} [\varphi_{122, \dots, m} + \varphi_{133, \dots, m} - 2\varphi_{123, \dots, m}] + \dots + J_{m-1, m} [\varphi_{1, \dots, m-1, m-1, m} + \varphi_{1, \dots, m-1, m} - 2\varphi_{123, \dots, m}] \}.$$

Here $J_{\mathbf{r}1} = J_{\mathbf{r}\mathbf{r}_1}$ and $J_{\mathbf{r}k} = J_{\mathbf{r}\mathbf{r}_k}$. The term in the curly brackets in (9) describes the magnon-magnon interaction. In fact, with neglect of this term, Eq. (9) has the solution

$$\varphi_{\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_m} = \varphi_{\mathbf{r}_1} \varphi_{\mathbf{r}_2} \dots \varphi_{\mathbf{r}_m}, \quad \epsilon_m^{(0)} = m\epsilon_1, \quad (10)$$

where $\varphi_{\mathbf{r}}$ and ϵ_1 are solutions of the one-particle problem (5), (6). In this approximation the energy levels ϵ_m are equidistant.

We shall assume the magnon-magnon interaction to be small and take it into account by perturbation theory; the

appropriate conditions of applicability will be elucidated below. Then, to first order, we have for the correction $\epsilon_m^{(1)}$

$$\epsilon_m^{(1)} = -m(m-1) \sum_{r,r'} S_r S_{r'} J_{rr'} \varphi_r \varphi_{r'} (\varphi_r - \varphi_{r'})^2. \quad (11)$$

From (7), (10) and (11) we obtain for the energy levels $\epsilon_m = \epsilon_m^{(0)} + \epsilon_m^{(1)}$ in both the limiting cases considered in (7) an expression of the form

$$\epsilon_m = m(g'H - \gamma) + 1/2 m(m-1)\alpha, \quad (12)$$

where $\gamma = zSJ' + S'\alpha$ and the quantity $\alpha = \alpha(J')$, which determines the extent to which the levels are not equidistant, is given by the following relations:

$$\text{For } J' \ll J, \quad \alpha = z(J')^2(I_w - 1)/J, \quad (13a)$$

$$\text{For } J' \gg J, \quad \alpha = J'. \quad (13b)$$

In the case of $g' = g$ under consideration, we could write gH in place of $g'H$ in (12), but the form (12) is more convenient for the latter generalizations.

It can be seen from (12) that, for $H = 0$, the spacing between the levels increases with increasing excitation: the difference between the ground ($m = 2S'$) and first levels is smaller than that between the first and second levels, and so on. Therefore, as the field increases from $H = 0$ to $gH \sim \gamma$, the ground level will first be the level with $m = 2S'$, then that with $m = 2S' - 1$, and so on.

Comparing the results of the first and zeroth approximations, we find the conditions for the applicability of perturbation theory:

$$\text{For } J' \ll J, \quad J'S'/JS \ll 1, \quad (14a)$$

$$\text{For } J' \gg J, \quad S'/zS \ll 1. \quad (14b)$$

The magnetic-field term does not appear in the estimates (14), since this term appears in the energy as an additive constant—it is the quantity $\epsilon_m - mgH$ that is expanded in the interaction. Since z is usually sufficiently large in three-dimensional lattices, it can be seen from (14) that, for small S'/S , the perturbation theory used can be applied for practically all J'/J .

The results in the expressions (12) and (13) can also be obtained in another, more standard, way. For this we note that the perturbation theory used in obtaining (13) corresponds, according to (7) and (14), to small amplitudes $|\varphi_{\mathbf{r} \neq 0}| \ll \varphi_0$, i.e., to small deviations of the spins of the matrix from saturation. This is natural, since the interaction energy of the magnons is proportional to their density (cf., e.g., [9-11]). But then we can use the spin-wave approximation [1] to describe the spins of the matrix, while the impurity-spin operator \hat{S}_0 must be described exactly. Changing from the spin operators of the matrix to Bose operators by means of the Maleev-Dyson transformation [8] and discarding the terms with four-boson interaction (which, as can be verified, give corrections of higher orders in the parameters (14)), we obtain for the Hamiltonian

$$\mathcal{H} = E_{00} + \mathcal{H}_1, \quad E_{00} = -1/2 zJS^2(N-2) - gHS(N-1); \quad (15)$$

$$\mathcal{H}_1 = S \sum_{r,r' \neq 0} J_{rr'} (a_r^+ a_{r'} - a_r^- a_{r'}^-) - gH \left(\hat{S}_0^z - \sum_{r \neq 0} a_r^+ a_r \right)$$

$$+ \frac{1}{2} \sum_{r'} J_{r0} [\sqrt{2S} (\hat{S}_0^+ a_r^+ + \hat{S}_0^- a_r^-) + 2\hat{S}_0^z (S - a_r^+ a_r^-)].$$

Here, in accordance with (1), $J_{\mathbf{r}0} = J' \sum \delta_{\mathbf{r}\Delta}$, where Δ , as above, are the coordinates of the nearest neighbors of the impurity.

We introduce the eigenfunctions χ_M of the operator S_0^z : $S_0^z \chi_M = M \chi_M$ and denote the "vacuum" state for the spins of the matrix by $|0\rangle$: $a_{\mathbf{r}} |0\rangle = 0$. The wavefunction Ψ_M corresponding to the total spin $(N-1)S + M$ (M is connected with m from (8) by the relation $M = S' - m$) can be written in the form of a series in $a_{\mathbf{r}}^{\pm}$:

$$\Psi_M = A \chi_M |0\rangle + \chi_{M+1} \sum_{r \neq 0} \psi_r a_r^+ |0\rangle + \dots \quad (16)$$

Here, the terms not written out give corrections to the energy in higher approximations and, to the order in which we are interested, can be omitted. Substituting (16) into the Schrödinger equation with the Hamiltonian (15), we obtain for A and $\psi_{\mathbf{r}}$ the system of equations

$$M(zSJ' - gH)A + J'\sqrt{S/2} S_{M,M+1}^- \sum_{\Delta} \psi_{\Delta} = \epsilon_M A; \quad (17a)$$

$$J_{r0} \sqrt{S/2} S_{M+1,M}^+ A + S \sum_{r' \neq 0} J_{rr'} (\psi_{r'} - \psi_r) + [(M+1)(zSJ' - J_{r0}) - (M+2)gH] \psi_r = \epsilon_M \psi_r. \quad (17b)$$

Here,

$$\epsilon_M = E_M - E_{00}, \quad S_{M,M'}^{\pm} = \langle \chi_M, \hat{S}_0^{\pm} \chi_{M'} \rangle.$$

In the lowest approximation, $\psi_{\mathbf{r}} = 0$ and $\epsilon_M^{(0)} = M(zSJ' - gH)$. In the next approximation we determine the quantity ψ_{Δ} in the s -state [1] from (17b) and substitute it into (17a). Using the explicit form of the matrix elements $S_{MM'}^{\pm}$ [12] and the above-mentioned relation $M = S' - m$, we see that in the limiting cases considered above we again obtain the expressions (12), (13) for the energy.

The case of unequal gyromagnetic ratios $g' \neq g$ is treated completely analogously. The expression for the energy has the form (12), as before. Also, for not too large fields $g'H \lesssim zSJ'$, the constants γ and α in the case of weak coupling $J' \ll J$ remain the same as in (13a), and in the case of large $J' \gg J$ corrections containing $g' - g$ appear in them:

$$\gamma = zSJ' + S'\alpha, \quad \alpha = \frac{J'}{1 - (g' - g)H/zSJ'} \quad (13c)$$

(here $J' \gg J$, $S'/zS \ll 1$, and $g'H \lesssim zSJ'$).

3. THERMODYNAMICS OF A FERROMAGNET WITH ANTIFERROMAGNETIC IMPURITIES

We shall consider temperatures that are low compared with the Curie point T_C of the pure magnet, so that the density of spin waves is small and the spin-wave approximation (15) can be used, as before, to describe them.

The impurity concentration c will be assumed small, so that we can regard the impurities as independent and neglect the concentration broadening of the local levels. Then the impurities can be regarded as an ideal gas, and the free energy F per unit cell can be written in the form

$$F = F_{00} + F_{sw}(T) + F_i(T). \quad (18)$$

Here,

$$F_{00} = -1/2 (1-2c) zJS^2 + czJ'SS' - gHS(1-c) - g'HS'c, \quad (19)$$

F_{sw} gives the thermodynamic contribution of the spin wave (the magnons of the continuous spectrum), and F_i is the contribution from the impurity local levels:

$$F_{sw}(T) = T \int_{\epsilon_{\min}}^{\epsilon_{\max}} \ln(1 - e^{-\beta\epsilon}) \rho(\epsilon) d\epsilon, \quad (20)$$

$$F_i(T) = -cT \ln Z_i = -cT \ln \sum_{m=0}^{2S'} e^{-\beta\epsilon_m}; \quad \beta = \frac{1}{T}. \quad (21)$$

Here $\rho(\epsilon)$ in (20) is the energy density of spin wave states in the presence of the impurities [11]. In (18) we have omitted the thermodynamic contribution from scattering of spin waves by each other and by impurities, which gives higher powers of T/T_C in F (cf., e.g., [9-11, 13]). For the small T considered, small $\epsilon \ll T_C$ are important in the integral (20), so that $\rho(\epsilon)$ can be written in the form $\rho = A(\epsilon - gH)^{1/2}$ [10, 11]. For the small c considered, the constant A contains a contribution linear in the concentration but independent of H and T : $A = A_1 + cA_2$; the explicit forms of A_1 and A_2 are given in, e.g., [11]. Thus, in the term F_{sw} the presence of impurities leads only to an unimportant renormalization factor, and this term is not considered below.

To discuss the contribution F_i of interest to us, we remark that the spacing between the impurity levels is, according to (12), a quantity of the order of $zSJ' - g'H$. Therefore, if the temperatures and fields are not great ($T, g'H \ll T_C$), the term F_i can vary appreciably only in the case of weakly coupled impurities, $J' \ll J$. In connection with this, below we shall mainly have in mind precisely this case, although formally only the general expression (12) will be used. For the same reason, we do not consider the possible contribution of the higher, so-called p and d local levels [1-3], which are absent for small J' (and for $J' \gtrsim J$ have energies $\epsilon_{p,d} \gtrsim T_C \gg T$).

For simplicity, we shall consider first the "two-level" case $S' = 1/2$. For the total magnetic moment, we have from (18)

$$M_z = gS(1-c) + M_{sw}(T) + g'c[1/2 - (e^{\beta\epsilon} + 1)^{-1}]. \quad (22)$$

Suppose $H = 0$. Then according to (12) $\epsilon_1 < 0$ and for $T \rightarrow 0$ we have $M_z = gS(1-c) - g'c/2$, which corresponds to the "Néel" ground state—the impurity spins are oriented "downward." As the temperature is raised, "mixing" over both states of the impurity spin occurs, and for $T \gg zSJ'$ the last term in (22) tends to zero, i.e., M_z is increased by $g'c/2$. At the same time, the term M_{sw} gives the well-known Bloch correction, of the form $\text{const}(T/T_C)^{3/2}$, to the moment [10, 11]. Therefore, if

$$c \gtrsim (zSJ'/T_C)^{3/2}, \quad (23)$$

the combination of these two factors leads to the appearance of a maximum in the curve $M(T)$ (see Fig. 2). This maximum, which was discussed qualitatively in [11], is completely analogous to the similar maximum for ferrites with a weakly coupled magnetic sublattice, which has been observed in a number of iron-garnets [14]. In the present case, a rigorous quantitative description of this temperature dependence is possible.

For a fixed temperature T , the moment M_i of the impurities increases with increasing H , reaching saturation when $g'H \gg \max\{T, \gamma\}$. If $T = 0$, at the point $g'H = \gamma$ the moment increases discontinuously by an amount $\sim c$, i.e., there occurs a "metamagnetic" phase transition with respect to the field [3], associated with a re-orientation of the quantized moment of each of the impurities. At a finite temperature, we obtain for the

impurity contribution χ_i^{ZZ} to the longitudinal susceptibility*

$$\chi_i^{ZZ} = \chi_0 c h^{-2} (e_i/T), \quad \chi_0 = c(g')^2/4v_c T. \quad (24)$$

Here v_c is the volume of the unit cell and χ_0 is the susceptibility of the system of free spins for $H = 0$. It can be seen from (24) that for $\epsilon_1 = 0$, i.e., $g'H = \gamma$, there is a maximum in the dependence of χ on H (Fig. 3a), corresponding to temperature broadening of the above-mentioned transition with respect to the field. At this point, $\chi = \chi_0$ (just as for $T \gg |\epsilon_1|$), since the impurity spins become "free." In addition, for any fixed H , there is also a maximum, at $T \sim |\epsilon_1|$, in the dependence of χ_i on T .

Finally, from (21) we can find the impurity specific heat at constant H :

$$C_i = c \frac{\epsilon_1^2}{4T^2} c h^{-2} \frac{\epsilon_1}{2T}. \quad (25)$$

It can be seen that the presence of impurities leads to a maximum, at $T \sim |\epsilon_1|$, in the dependence of the specific heat C_i on T , corresponding to "unfreezing" of the spins in this region. In the dependence of C_i on the field H at fixed T there will be two maxima, at $g'H - \gamma \sim \pm T$; at the above-mentioned point of "neutral equilibrium" of the spins ($\epsilon_1 = 0, g'H = \gamma$) the specific heat C_i vanishes (Fig. 4a). At this point, the entropy per impurity spin has the constant value $\ln 2$ for all T , so that $C_i = T \partial S_i / \partial T = 0$.

We now discuss the general case $S' > 1/2$. Substituting (12) into (21) and differentiating F with respect to H , we find that the dependence of M_z on T in the absence of the field will remain qualitatively the same as for $S' = 1/2$, and under the condition (23) will have a maximum,

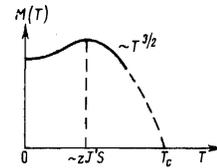


FIG. 2

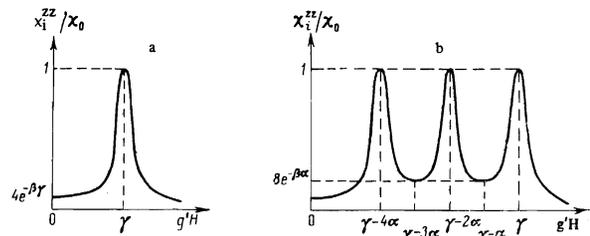


FIG. 3. Dependence of χ on H : a) $S' = 1/2, T \ll \gamma$; b) $S' = 3/2, T \ll \alpha$.

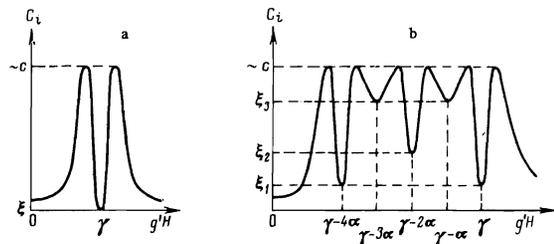


FIG. 4. Dependence of C_i on H : a) $S' = 1/2, T \ll \gamma$; b) $S' = 3/2, T \ll \alpha$. Notation: $\xi = c(\beta\gamma)^2 e^{-\beta\gamma}$, $\xi_1 = 2c(\beta\gamma)^2 e^{-2\beta\alpha}$, $\xi_2 = 4c(\beta\alpha)^2 e^{-2\beta\alpha}$, $\xi_3 = 2c(\beta\alpha)^2 e^{-\beta\alpha}$.

at $T \sim zJ'SS'$, of the type drawn in Fig. 2. But the dependence of M on H for $T \ll \alpha$ has a steplike character with jumps at the points $H_n = \gamma - 2n\alpha$, $n = 0, 1, 2, \dots$, $2S' - 1$, in accordance with the above-mentioned sequential occupation of the different states of the $(2S' + 1)$ -multiplet. Correspondingly, χ_1^{ZZ} has maxima at these points, and, near the n -th maximum for $T \ll \alpha$, is given by the formula (24) with ϵ_1 replaced by $g'(H - H_n)$. Thus, e.g., for $S' = 3/2$ the dependence of χ_1^{ZZ} on H for $T \ll \alpha$ is depicted schematically in Fig. 3b. On increase of T the maxima become less sharp, and merge into one maximum for $T \gtrsim \alpha$. But if $T \gg \alpha$, we can omit the term with α in the spectrum (12) when calculating the free energy (21). Then the magnetic moment is expressed in terms of the Brillouin function, as in the molecular-field approximation for the impurities [11].

The specific heat can be treated analogously. For $H = 0$ there will be a maximum in the dependence of C_i on T at $T \sim \gamma \gg \alpha$; as we have noted, to describe this maximum we can now use the molecular-field approximation. But if $T \ll \alpha$, the specific heat C_i as a function of H has $2S'$ double maxima at the points $g'(H - H_n) \sim \pm T$. In this case, in the regions $\alpha e^{-\beta\alpha} \ll g'|H - H_n| \ll \alpha$, the specific heat has the form (25) with ϵ_1 replaced by $g'(H - H_n)$. For spin $S' = 3/2$ this dependence for $T \ll \alpha$ is depicted in Fig. 4b.

The spin-wave contributions (which we have not considered) from F_{sw} to χ^{ZZ} and C (like the neglected phonon contributions to the specific heat, etc.) have a known smooth dependence on H and T in the region considered [10,11]. Therefore, the sharply nonmonotonic anomalies described can be observed even for small c .

4. CONTRIBUTION OF IMPURITIES TO THE TRANSVERSE SUSCEPTIBILITY AND FERROMAGNETIC RESONANCE

The susceptibility of a system in a uniform oscillating field can be expressed in the usual way in terms of the retarded Green function; cf., e.g., [11]. After standard computations, for the contribution of the impurities to the transverse susceptibility in the "gas" approximation considered we can obtain the expression

$$\chi^{+-}(\omega) = \frac{1}{Z_i} \frac{1}{N\nu_c} \sum_{m=1}^{2S'} \left| \left(\sum_r g_r S_r^+ \right)_{m-1,m} \right|^2 \frac{e^{-\beta\epsilon_m} - e^{-\beta\epsilon_{m-1}}}{\omega - (\epsilon_m - \epsilon_{m-1}) + i\delta}. \quad (26)$$

Here Z_i and ϵ_m are the same as in (21) and (12); $\delta \rightarrow +0$.

To calculate the matrix element appearing in (26) we shall make use of the following device. We write the operator equation of motion for \hat{S}_r^+ , sum both parts of the equality over r and take the matrix element of it. As a result, we obtain

$$(\epsilon_m - \epsilon_{m-1}) \left(\sum_r S_r^+ \right)_{m-1,m} = H \left(\sum_r g_r S_r^+ \right)_{m-1,m}. \quad (27)$$

If there is one impurity at the site $r = 0$, we have from (27)

$$\left(\sum_r g_r S_r^+ \right)_{m-1,m} = (g' - g) \frac{\epsilon_m - \epsilon_{m-1}}{\epsilon_m - \epsilon_{m-1} - gH} (S_0^+)_{m-1,m}. \quad (28)$$

In the approximation under consideration ("weak coupling" of the impurity spin with the matrix, when the second term in (16) is much smaller than the first), we can take for the matrix element of S_0^+ its "free" expression (cf. [11]):

$$(S_0^+)_{m-1,m} = \sqrt{(S' - M)(S' + M + 1)}, \quad M = S' - m. \quad (29)$$

The final expression for χ^{+-} is obtained by substituting (28), (29) into (26) and multiplying by the number of impurities. It is easily verified that, for $T = 0$ and sufficiently small (large) H , when the ground level is the level with $m = 2S'$ ($m = 0$), χ^{+-} goes over into the expressions (20.14) and (20.8) of the book by Izumov and Medvedev [11], near the corresponding resonances.

We shall write down the final expression for the imaginary part of $\chi^{+-}(\omega)$, which determines the absorption of energy by the local levels:

$$\text{Im} \chi^{+-}(\omega) = c \frac{2\pi S'}{Z_i \nu_c} (g' - g)^2 \left(\frac{\omega}{\omega - gH} \right)^2 \sum_{m=1}^{2S'} m \left(1 - \frac{m-1}{2S'} \right) [\exp(-\beta\epsilon_{m-1}) - \exp(-\beta\epsilon_m)] \delta(\omega - \epsilon_m + \epsilon_{m-1}). \quad (30)$$

In agreement with a remark in [11], absorption from a uniform field with $\omega \neq gH$ is possible only for $g' \neq g$ and is proportional to the square of the difference $g' - g$. It can be seen that there are δ -function peaks in $\text{Im} \chi^{+-}$ at the transition frequencies $\omega_m = \epsilon_m - \epsilon_{m-1}$ between neighboring levels. Allowance for the relativistic interactions, concentration broadening, and temperature broadening as a result of absorption of spin-waves (see below) leads to a finite width of these peaks. The continuous spectrum gives a contribution to $\text{Im} \chi^{+-}$ only for frequencies $\omega \geq gH$.

We shall study how the dependence of $\text{Im} \chi^{+-}$ on ω changes with increase of the magnetic field H for $T < \alpha$. For definiteness, we put $S' = 3/2$. For $H \rightarrow 0$ the ground level is the level ϵ_3 , which is the most highly populated. Therefore, the line $\omega = \omega_3 = -(\gamma - 4\alpha)$ has the greatest intensity. The next lines $\omega = \omega_2 = -(\gamma - 2\alpha)$ and $\omega = \omega_1 = -\gamma$ have intensities that decrease with increasing $|\omega|$, as depicted schematically in Fig. 5a. We recall that the sign of ω in the formalism used simply denotes the sign of the circular polarization of the absorbed radiation [11]. Therefore, the fact that all the ω_m are negative for small H means that only a wave with a definite polarization ($h_x + ih_y = h e^{i\omega t}$) is absorbed at these frequencies. But absorption at frequencies $\omega \geq gH$ of the continuous spectrum will occur only for the opposite sign of the circular polarization, in accordance with the momentum and energy conservation laws. With increasing H , all the frequencies ω_m will be shifted to the right; the intensities of the lines at ω_1 and ω_2 will increase, and that at ω_3 will decrease. Fig. 5b corresponds to a field $g'H \gtrsim \gamma - 4\alpha$, when $\omega_3 \gtrsim 0$. On further increase of $g'H$

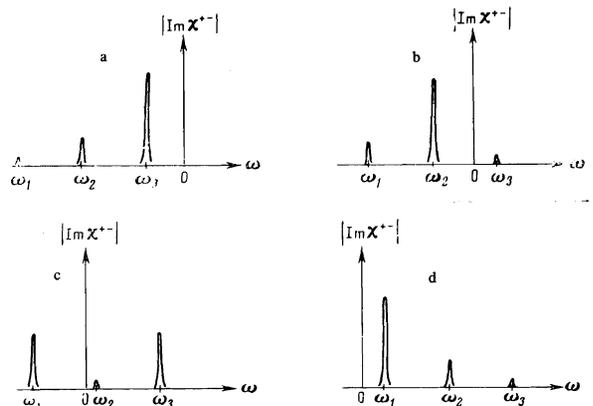


FIG. 5. Change of the ferromagnetic resonance lineshape with variation of the magnetic field H : a) $g'H = 0$, $S' = 3/2$; b) $g'H \gtrsim \gamma - 4\alpha$, $S' = 3/2$; c) $g'H \gtrsim \gamma - 2\alpha$, $S' = 3/2$; d) $g'H \gtrsim \gamma + \alpha$, $S' = 3/2$.

from $\gamma - 4\alpha$, to $\gamma - 2\alpha$, the intensities at ω_1 and ω_3 increase, and that at ω_2 decreases (see Fig. 5c). On increase of $g'H$ from $\gamma - 2\alpha$ to γ , the intensities of the lines at ω_1 and ω_3 decrease, and that at ω_2 increases. Finally, for $g'H > \gamma$, the level $\epsilon_0 = 0$ becomes the ground (most populated) level, and for $g'H - \gamma \gtrsim \alpha$ the intensity distribution is shown in Fig. 5d.

5. IMPURITY LEVELS IN A FERRITE

We shall consider a two-sublattice model of a ferrite. Again, we shall use the Heisenberg model (1) with nearest-neighbor interaction in a cubic lattice, and shall assume that the sublattices alternate so that the nearest neighbors of each spin are always spins of the other sublattice. Let the spin of the first sublattice be greater than that of the second ($S_1 > S_2$), and let the impurity spin S' be situated in the first sublattice at the site $\mathbf{r} = 0$.

Exact expressions for the energy and wave function of the ferrite are not known, and a consistent theory exists only for large spins $S_1, S_2 \gg 1$. This will be assumed to be the case below, although it may be thought that, as in the pure case (cf. [10]), the results will still be sufficiently accurate for $S_1, S_2 \geq 1$. We shall also assume that the impurity spin is weakly coupled with the matrix ($|J'| \ll J$), so that, as above, for the spins of the matrix we can go over from spin operators to Bose operators (a, a^+ for the first sublattice and b, b^+ for the second [11]). As a result, the Hamiltonian of the system takes the form, in place of (15),

$$\begin{aligned} \mathcal{H} &= E_{00} + \mathcal{H}_{0m} + \mathcal{H}_{01} + V, \quad E_{00} \\ &= -\frac{1}{2} zJS_1S_2(N-2) - g_1HS_1\left(\frac{N}{2} - 1\right) + g_2HS_2\frac{N}{2}; \\ \mathcal{H}_{0m} &= \frac{1}{2} J \sum_{\mathbf{r} \neq 0} \left[\sqrt{S_1S_2} (a_{\mathbf{r}} b_{\mathbf{r}+\Delta} + a_{\mathbf{r}}^+ b_{\mathbf{r}+\Delta}^+) + S_2 a_{\mathbf{r}}^+ a_{\mathbf{r}} + S_1 b_{\mathbf{r}+\Delta}^+ b_{\mathbf{r}+\Delta} \right] \\ &\quad + g_1 H \sum_{\mathbf{r} \neq 0} a_{\mathbf{r}}^+ a_{\mathbf{r}} - g_2 H \sum_{\mathbf{r}+\Delta} b_{\mathbf{r}+\Delta}^+ b_{\mathbf{r}+\Delta}; \\ \mathcal{H}_{01} &= -(zS_2J' + g'H) S_0^z, \\ V &= J' \sum_{\Delta} \left[\sqrt{\frac{S_2}{2}} (S_0^+ b_{\Delta} + S_0^- b_{\Delta}^+) + S_0^z b_{\Delta}^+ b_{\Delta} \right]. \end{aligned} \quad (31)$$

Here \mathbf{r} and $\mathbf{r} + \Delta$ label the sites of the first and second sublattices respectively, and it is taken into account that the exchange integral $J_{\mathbf{r}, \mathbf{r}+\Delta} = J > 0$ for the spins of the matrix, in accordance with the antiferromagnetic character of the interaction. As in (15), terms with four-boson interaction, which give higher powers of J' , $1/S_1$ and $1/S_2$, have been omitted in (31).

The Hamiltonian \mathcal{H}_{0m} , which describes free magnons in a lattice with a "hole" at the site $\mathbf{r} = 0$, can be diagonalized by means of an ordinary canonical transformation (cf., e.g., [11]):

$$\begin{aligned} a_{\mathbf{r}} &= \sum_{\nu} (\alpha_{\nu} \varphi_{\mathbf{r}\nu} + \beta_{\nu} \chi_{\mathbf{r}\nu}^*), \quad b_{\mathbf{r}} = \sum_{\nu} (\alpha_{\nu}^+ \psi_{\mathbf{r}\nu} + \beta_{\nu} \xi_{\mathbf{r}\nu}^*); \\ \mathcal{H}_{0m} &= E_{vac} + \sum_{\nu} (\epsilon_{1\nu} \alpha_{\nu}^+ \alpha_{\nu} + \epsilon_{2\nu} \beta_{\nu}^+ \beta_{\nu}). \end{aligned} \quad (32)$$

Here α_{ν} and β_{ν} are Bose operators, and $\rho = \mathbf{r} + \Delta$. The explicit form of the functions $\varphi_{\mathbf{r}\nu}^{\nu}$, $\chi_{\mathbf{r}\nu}^{\nu}$, $\psi_{\mathbf{r}\nu}^{\nu}$ and $\xi_{\mathbf{r}\nu}^{\nu}$ and of the corresponding energies $\epsilon_{1\nu}$ and $\epsilon_{2\nu}$ of the "acoustic" and "optical" magnon branches respectively can be found as in [11]. But the appropriate expressions are cumbersome, and their concrete form is unimportant for the following discussion.

The wave function of the system, corresponding to the

total spin $M^Z = S_1(N/2 - 1) - S_2N/2 + M$, can be written, analogously to (16), in the form of an expansion:

$$\Psi_M = A \chi_M |0\rangle + \chi_{M+1} \sum_{\nu} c_{\nu} \alpha_{\nu}^+ |0\rangle + \chi_{M-1} \sum_{\nu} d_{\nu} \beta_{\nu}^+ |0\rangle + \dots \quad (33)$$

Here $|0\rangle$ is the vacuum state for the operators α_{ν} and β_{ν} ; χ_M is the same as in (16), and the terms not written out give corrections of higher orders in J'/J to the energy.

Substituting (33) into the Schrödinger equation, we obtain a system of three equations for A , c_{ν} and d_{ν} . Solving it by expanding in J' up to and including terms $\sim J'^2$, we find

$$\begin{aligned} \epsilon_M &= -M \left[g'H + zS_2J' \left(1 - \frac{1}{zS_2} \sum_{\Delta} |\psi_{\Delta}^{\nu}|^2 \right) \right] \\ &- (J')^2 S_0^z \left\{ \frac{(S'-M)(S'+M-1)}{2} \sum_{\nu} \frac{|\psi_{\nu}^{\nu}|^2}{\epsilon_{1\nu} - zS_2J' - \sigma'H - i\delta} \right. \\ &\left. + \frac{(S'+M)(S'-M+1)}{2} \sum_{\nu} \frac{|\xi_{\nu}^{\nu}|^2}{\epsilon_{2\nu} + zS_2J' - \sigma'H - i\delta} \right\}. \end{aligned} \quad (34)$$

Here,

$$\epsilon_M = E_M - E_{00} - E_{vac}, \quad \psi_{\nu} = \sum_{\Delta} \psi_{\Delta}^{\nu}, \quad \xi_{\nu} = \sum_{\Delta} \xi_{\Delta}^{\nu},$$

and the auxiliary quantity $\delta \rightarrow +0$ enables us, as usual, to find the imaginary part of ϵ_M (the damping of the level) if the corresponding denominator can vanish.

If we introduce, as above, the quantity $m = S' - M$, the expression (34) again takes the form (12). In the approximation linear in J' , the energy has the form (10), and, accurate to the term $\sim 1/S_2 \ll 1$, we have $\epsilon_M^{(0)} \approx m(g'H + zS_2J')$.

It can be seen that if $J' > 0$, i.e., J' and J have the same sign, then $\epsilon_M > 0$ and in the ground state the impurity spin is oriented parallel to the magnetization of the crystal. In this case, the denominator of the first term in the curly brackets in (34) can vanish (for the small fields $g'H \lesssim zS_2J'$ of interest to us), and this corresponds to the possibility of emission of an acoustic magnon with a transition to a level with smaller m (larger M). The corresponding imaginary part and the damping of the level are proportional to $(J')^{5/2}$. Thus, the levels are quasi-local, and all their properties, including their dependence on the field H , are analogous to the properties of the levels of a ferromagnetic impurity in a ferromagnet [11].

On the other hand, if $J' < 0$, in weak fields $g'H \lesssim zS_2J'$ the levels ϵ_m are negative, so that the impurity spin is oriented against the magnetization. Moreover, if the magnitude of zS_2J' is less than the gap, of order $zJ(S_1 - S_2)$ [11, 10], in the spectrum of the optical magnons, then neither denominator in (34) vanishes, i.e., emission of magnons is forbidden by the energy and momentum conservation laws. Thus, there is no damping of the impurity states, and the structure of the levels and their dependence on H are found to be the same as in the case (12).

The case when the impurity spin is in the second sublattice can be treated analogously. In this case, the energy again has the form (12), and the levels ϵ_m are negative and local for $J' > 0$ and positive and quasi-local for $J' < 0$.

All these results are a manifestation of the general analogy, noted in [11], between impurity levels in a fer-

rite and in a ferromagnet. It can be seen that, in the case under study of a weakly coupled impurity, the properties of the levels in the ferrite and in the ferromagnet are the same when the directions of the impurity spin (in the ground state) with respect to the total magnetization are the same. This analogy is connected with the fact that, in processes associated with transitions between levels of a weakly coupled impurity, only long-wavelength magnons with $\epsilon_\nu \lesssim zS_1 J'$ play an important role. But for such magnons, as is well known, the short-wave, "optical" magnetic structure becomes unimportant, all phenomena are determined by the averaged characteristics of the unit cell, and the ferrite becomes equivalent to a ferromagnet with total spin $S = S_1 - S_2$ [9,10].

Thus, the anomalies treated in Secs. 3 and 4 can also be observed in ferrites, if the impurity spin in the ground state is antiparallel to the magnetization (an impurity in the first sublattice with ferromagnetic interaction with its nearest neighbors, or one in the second with antiferromagnetic interaction). The opposite case is analogous to the case of a ferromagnetic impurity in a ferromagnet. At the same time, from the point of view of the phenomena treated, an impurity antiferromagnet differs sharply from both the ferromagnet and the ferrite. In this case, there is no gap in the spectrum for magnons of either polarization, so that for small J' all impurity states are quasi-local and the behavior in a magnetic field can depend substantially on the lattice structure, the type of impurity, and so on (cf., e.g., [1,15]).

6. CONCLUSION

A highly idealized Heisenberg model of the magnet was used above. However, the anomalies in the dependences on H and T described in Secs. 3 and 4 have a very simple physical nature: the reversal of the spin of the antiferromagnetic impurity under the influence of the field or the temperature in the presence of the unequal spacing of the levels that arises from the magnon-magnon interaction. It may be thought, therefore, that these phenomena will also remain in a more complete and realistic description.

For example, the presence of a crystal field usually splits the spin multiplets of the individual ions into a series of sublevels. But in lattices of sufficiently high symmetry these sublevels usually still contain a sufficiently large number of degenerate levels, for the description of which it is often sufficient to use a Heisenberg model with low effective spin, with the possible addition of an anisotropy field (cf., e.g., [4]). It is evident that the effect of the anisotropy field in the region $T \ll T_C$ under consideration basically reduces, as usual [11], to a redefinition of the effective field H . Allowance for interactions between non-nearest neighbors and for the real structure of the lattice usually also gives no qualitative changes, especially for the lower maximum-symmetry levels under consideration [1,4].

Less clear is the generalization of the results to the case of ferromagnetic metals (if it will be possible to find such systems with antiferromagnetic impurities). The direct interaction of the impurity with the conduction electrons can, generally speaking, give certain additional effects. It may be thought, however, that for the low $T \ll T_C$ under consideration the described proper-

ties of the impurity levels are not radically changed; cf., e.g., [16].

Only the region of low temperatures and concentrations has been considered above; we shall discuss the effect of increase of T or c . Quantitatively, as is well known, the study of the region of higher $T \sim T_C$ is difficult, but to understand the qualitative results we can usually use the self-consistent field approximations [9,4,1]. Then, with increasing T , the average spin of the matrix, $\langle S_z(T) \rangle$, appears in place of the spin S in the formulas (12) and (13), i.e., the splitting of the levels (in the absence of the field H) will decrease. In addition, the presence of a finite density of spin waves, and of their interaction with each other, leads to the appearance of damping of the impurity levels. Thus, e.g., processes of absorption of a spin wave by a level with component M with a transition to the lower level $(M-1)$ and emission of two spin waves in accordance with the scheme $M + \epsilon \rightarrow (M-1) + \epsilon' + \epsilon''$ are possible. It is easy to convince oneself that the momentum and energy conservation laws will be fulfilled, although for small T/T_C and J'/J the corresponding width will be extremely small. However, for observation of the above-described oscillations in a magnetic field, the total width Γ of the levels should also be small compared with the "degree of nonequidistance": $\Gamma < \alpha \sim (J')^2/J$.

An increase in the concentration c also leads to broadening of the levels, so that the concentration broadening (the width of the impurity band near the m -th local level) should also be less than α . The oscillations described are a quantum effect, associated with the quantization and splitting of the levels of an individual impurity in the combined field of the medium and H . With increasing concentration, inter-impurity interaction appears, so that the impurity system is characterized by a resultant macroscopic moment, for which the quantum nature of the variation in the magnetic field is unimportant. Therefore, for higher c , a classical monotonic increase in magnetization with increasing H (cf., e.g., [17]) will be observed in place of the steplike increase corresponding to Figs. 3-5.

In conclusion, we give some numerical estimates. If for the estimates we use parameters characteristic for iron garnets with rare-earth impurities [17] ($T_C \sim 500^\circ$, $J'/J \sim 0.1$), then fairly restrictive conditions are necessary for the experimental observation of the described oscillations in a magnetic field. "Flipping" of the impurity spins occurs in fields of the order of several hundred kOe (e.g., for a Gd impurity in $Y_3Fe_5O_{12}$, $H_{eff} \sim 300$ kOe [17]), and the spacing $\Delta H \sim 20$ kOe between the maxima should be greater than the temperature: $T \lesssim \alpha \sim 3^\circ$. Decrease of J' lowers the magnitude of the field H , but requires a further lowering of T . To make these estimates more precise, the calculations described above must be performed for specific lattices, as in the analogous treatment in [4].

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