Concentration broadening of local levels in a ferromagnetic substance with antiferromagnetic impurities

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Local energy levels of a ferromagnet with two antiferromagnetic impurities are considered and classified. The concentration broadening of the local impurity levels is obtained. The concentration range in which magnetic-susceptibility oscillations can be observed is determined.

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1. INTRODUCTION

The singularities of the magnon spectrum in a ferromagnet with magnetic impurities were investigated in a number of studies.[1–3] It was shown that local levels, namely bound states of magnons on the impurity, are produced in the case of antiferromagnetic interaction between the impurity spin $\mathbf{S}'$ and the matrix spin $\mathbf{S}$. Physically this means that an antiparallel "Néel" alignment of $\mathbf{S}'$ relative to the direction of the total magnetization M_z is energywise favored.

In a paper by Vaks and one of us,[4] henceforth cited as I, it was shown that allowance for the magnon-magnon interaction makes the $2S'+1$ levels of the impurity non-equidistant at $S'>\frac{1}{2}$. This circumstance in turn causes distinctive oscillations of the magnetic susceptibility χ and of the heat capacity C with changing external magnetic field. Neglecting the relativistic interactions that do not conserve the total moment M_z, all the $2S'+1$ levels of the impurity spin are strictly local at a temperature $T=0$, i.e., they have no width. Therefore in the approximation linear in the concentration c, when no account is taken of the interaction between the impurities, these oscillations have a δ-function character at $T=0$ (see I). With increasing c, however, and also with increasing T, the singularities should become smeared out, and this raises the question of the possibility of experimentally observing these oscillatory phenomena.

In this paper we determine the concentration broadening of local impurity levels and the range of concentration values in which susceptibility oscillations can be observed. The analysis is carried out within the framework of the Heisenberg model with nearest-neighbor interaction, and principal attention is paid to the case of weakly-coupled impurities.

Our problem is close to that of the density of states of a particle in a random impurity field.[5] Thus, if the spin-wave approximation[1] is valid for the impurity spins, the magnon-magnon interaction can be neglected and the problem is fully equivalent to the usual one.[5] However, if all the $2S'+1$ local levels are considered ($S'>\frac{1}{2}$), it is precisely the magnon-magnon interaction which leads to their non-equivalence (see I). Therefore in this case, in contrast to the case considered by Lifshitz,[5] we are dealing with a many-particle problem.

The concentration broadening is calculated in the pair approximation, i.e., the contribution made to the magnon spectrum by the interaction of not more than two impurities is taken into account. The region of applicability of this approximation is well known from the theory of concentration broadening of local levels of phonons and the electrons in crystals with impurities,[5] and its application to our problem will be discussed below.

2. SINGLE-MAGNON LOCAL LEVELS AT TWO IMPURITIES

Just as in I (see also [1]), we consider a Heisenberg ferromagnet with cubic lattice with spin $\mathbf{S}$ and nearest-neighbor interaction J. Let two substitution impurities with spin $\mathbf{S}'$ be located at the points $\mathbf{r}=\mathbf{R}_1$ and $\mathbf{r}=\mathbf{R}_2$, and let their interaction with the matrix spin be equal to J'. The gyromagnetic ratios for the spins of the matrix and the impurity will initially be assumed to be equal, $g=g'$.

For the wave function $\varphi_\mathbf{r}$ of the single-magnon state we have the equation (I.5) (the notation is the same as in I):

$$E\varphi_\mathbf{r}+\sum_{\mathbf{r}'}S_{\mathbf{r}'}J_{\mathbf{r}\mathbf{r}'}(\varphi_\mathbf{r}-\varphi_{\mathbf{r}'})=0, \quad E=E_1-gH \tag{1}$$

with the normalization condition (I.4). Here E_1 is the energy of the single-magnon state reckoned from the energy of the fully aligned state. From (1) we obtain for the wave function $\varphi_\mathbf{r}$ in the case when the impurities are not nearest neighbors ($\mathbf{R}=\mathbf{R}_1-\mathbf{R}_2\neq\boldsymbol{\Delta}$),

$$\varphi_\mathbf{r}=SJ\sum_{\boldsymbol{\Delta}}\{[\rho G(\mathbf{r}-\mathbf{R}_1-\boldsymbol{\Delta})-\delta G(\mathbf{r}-\mathbf{R}_1)](\varphi_{\mathbf{R}_1}-\varphi_{\mathbf{R}_1+\boldsymbol{\Delta}})+\ldots\}. \tag{2}$$

The second term of (2), which has not been written out, is obtained from the first by making the substitution $\mathbf{R}_1\to\mathbf{R}_2$. In formula (2), $\boldsymbol{\Delta}$ stands for the distance between the nearest neighbors and z (the lattice constant is assumed equal to unity), and

$$\rho=1+\frac{S'J'}{SJ}, \quad \delta=1+\frac{J'}{J}, \quad G(\mathbf{r})=\sum_\mathbf{k}\frac{e^{i\mathbf{k}\mathbf{r}}}{E-\omega_\mathbf{k}}, \tag{3}$$

where $\omega_\mathbf{k}$ is the same as in (I.6).

Putting $\mathbf{r}=\mathbf{R}_1$, $\mathbf{R}_1+\boldsymbol{\Delta}$, $\mathbf{R}_2$, $\mathbf{R}_2+\boldsymbol{\Delta}$, in (2), we can obtain a closed system of 14 equations for the energy levels E_1. In the general case, however, an investigation of such a system is difficult. In this paper we are interested only in the low-lying most symmetrical so-called s states,[1] and mainly in the case $J' \ll J$. If we confine ourselves at the same time to large distances between impurities (these are precisely the values of R we need) the determination of the energy levels becomes much easier.

We first derive a formula useful for the determination of these energy levels. To this end we put $\mathbf{r} = \mathbf{R}_1$ in (2). We then write an equation for $\varphi_{\mathbf{R}_1+\boldsymbol{\Delta}}$ and sum it over $\boldsymbol{\Delta}$. Using the second equation to eliminate the term with $\varphi_{\mathbf{R}_2+\boldsymbol{\Delta}}$ from the first, we obtain ultimately

$$\frac{E}{zSJ} = (1-\delta)(1-\psi_1), \quad \psi_1 = \frac{1}{z}\sum_{\Delta}\frac{\varphi_{\mathbf{R}_1+\Delta}}{\varphi_{\mathbf{R}_1}}. \tag{4}$$

The term with $\varphi_{\mathbf{R}_1+\boldsymbol{\Delta}}$ can be eliminated in similar fashion. Here again we obtain formula (4) with the substitution $\mathbf{R}_1 \to \mathbf{R}_2$. Since the energy E should not depend on the choice of ψ_i, the equality $\psi_1 = \psi_2$ should be satisfied.

Let us find the energy levels at large distances between impurities, $R \gg 1$, in the form of an expansion in powers of $1/R$:

$$E = E^{(0)} + E^{(1)} + \ldots, \quad \psi_1 = \psi_1^{(0)} + \psi_1^{(1)} + \ldots$$

Determining $\psi_1^{(0)}$ from the equation for $\varphi_{\mathbf{R}_1}$ and substituting it in (4), we obtain the usual equation for the s-state energy with one impurity (I.6). In this, zeroth approximation $\psi_1^{(0)} = \psi_2^{(0)}$ is an identity. In the next approximation in $1/R$ we must put in the equation for $\varphi_{\mathbf{R}_1}$

$$\varphi_{\mathbf{R}_2+\Delta} \approx \varphi^{(0)}_{\mathbf{R}_2+\Delta} = \psi_2^{(0)}\varphi_{\mathbf{R}_2} = \psi_1^{(0)}\varphi_{\mathbf{R}_2},$$

inasmuch as in the s state the wave function of the magnon at an isolated impurity is symmetrical (see I, and also [1]). As a result we obtain an equation for $\psi_1^{(1)}$ in terms of the unknown ratio $\varphi_{\mathbf{R}_1}/\varphi_{\mathbf{R}_2}$.

A similar procedure is used to determine $\psi_2^{(1)}$. From the equality $\psi_1^{(1)} = \psi_2^{(1)}$ we obtain $\varphi_{\mathbf{R}_1}/\varphi_{\mathbf{R}_2} = \pm 1$, so that in this approximation the energy levels are split and take the form

$$E^{(1)} = -\sigma(1-\delta)\left(\rho - \delta - \rho\frac{E^{(0)}}{zSJ}\right) G^0(\mathbf{R}) \Big/ \left\{(\rho G_1^0 - \delta G_0^0)^2 + (1-\delta)\left[\frac{\partial}{\partial E^{(0)}}(\rho G_1^0 - \delta G_0^0)\right]\right\}, \tag{5}$$

$\sigma = \pm 1$. Here $G_0 = G(0)$, $G_1 = G(\boldsymbol{\Delta})$, $\mathbf{R} = \mathbf{R}_1 - \mathbf{R}_2$ and $G^0(\mathbf{r})$ is given by expression (3) with $E = E^{(0)}$.

In the case of weakly-coupled impurities $J' \ll J$, which is of greatest interest to us, using the expression for $E^{(0)}$ from I and the asymptotic form of the function $G(R)$, we obtain ultimately

$$E_1^\sigma = \varepsilon_1 - \sigma\varepsilon_R, \quad \varepsilon_R = zS'\frac{(J')^2}{J}\frac{n}{4\pi}\frac{e^{-\varkappa R}}{R}, \quad \varkappa^2 = n\frac{J'}{J}. \tag{6}$$

Here ε_1 is given by (I.7a), and n is equal to 6, 2, and 3, respectively for the primitive cubic, BCC, and FCC lattices. We note that the relation between R and $1/\varkappa$ in (6) is arbitrary. For (6) to be valid it suffices to satisfy the condition $R \gg 1$.

From (2) with allowance for the normalization condition (I.4) it is easy to obtain the wave function ($J'' \ll J$)

$$\varphi_{\mathbf{r}}^\sigma = \frac{1}{\sqrt{2}}(\varphi_{\mathbf{r}}^{(1)} + \sigma\varphi_{\mathbf{r}}^{(2)}), \quad \sigma = \pm 1, \tag{7a}$$

$$\varphi_{\mathbf{r}}^{(i)} \approx (2S')^{-1/2}\left[\frac{S'J'}{SJ}G(\mathbf{r}-\mathbf{R}_i) + \frac{1}{zSJ}\delta_{\mathbf{r},\mathbf{R}_i}\right]. \tag{7b}$$

It is seen from (7) and (6) that, as expected, the symmetrical state $\varphi_{\mathbf{r}}^+$ corresponds to a lower energy $E_1^+ = \varepsilon_1 - \varepsilon_R$.

The character of the energy correction of next order in $1/R$ is of interest. To calculate this correction it is necessary to know the function $\varphi(\mathbf{R}_i + \boldsymbol{\Delta})$ with higher accuracy. As a result, we arrive at expression (6) with ε_R replaced by $(1 + J'/J)\varepsilon_R$, i.e., the next-order correction is small in terms of the parameter $J'/J \ll 1$.

We note expression (6) can be obtained also by another more standard method.[6] To this end it is necessary to express the magnon wave function in the form (7), i.e., as a symmetrical and antisymmetrical combination of the wave functions for the isolated impurities, and substitute in (1). We then arrive at the result (6). The next-order corrections, however, are incorrectly described in this approximation and must be discarded.

3. MANY-MAGNON LOCAL LEVELS AT TWO IMPURITIES

We consider now many-particle states with m magnons, with $m \leqslant 4S'$. At $m = 4S'$ the spins of both impurities are antiparallel to the magnetic moment of the system, so that this is the ground state.

The equation for the m-magnon wave function $\varphi_{\mathbf{r}_1\ldots\mathbf{r}_m} \equiv \varphi_{1\ldots m}$ (symmetrical in all the coordinates) was obtained in I, see formula (I.9). The term in the curly bracket of (I.9) describes the magnon-magnon interaction. If this term is disregarded, Eq. (I.9) has as its solution

$$\varphi^{\sigma_1\ldots\sigma_m}_{\mathbf{r}_1\ldots\mathbf{r}_m} = \varphi^{\sigma_1}_{\mathbf{r}_1}\ldots\varphi^{\sigma_m}_{\mathbf{r}_m}, \quad E^{(0)}_m = E^{\sigma_1}_1 + \ldots + E^{\sigma_m}_m, \quad \sigma_i = \pm 1, \tag{8}$$

where $\varphi_{\mathbf{r}}^\sigma$ and E_1^σ make up the solution of the single-particle problem (1) in the field of two impurities. At $J' \ll J$ and $R \gg 1$ we must use expressions (6) and (7) for E_1^σ and $\varphi_{\mathbf{r}}^\sigma$.

Just as in I, assuming the magnon-magnon interaction to be small, we account for it by perturbation theory. For an isolated impurity with l magnons, the energy correction takes the form

$$E_l^{(1)} = {}^1\!/_2\, l(l-1)\alpha, \tag{9}$$

where α is the same as in (I.13). Since $\varepsilon_R \ll \alpha$, all the states (8) must be regarded as degenerate and the wave function must be sought in the form of a linear combination of expressions of the type (8) with unknown coefficients $A_{\sigma_1\ldots\sigma_m}$. The substitution of such a wave function in (I.9) leads to a system of equations for the quantities $A_{\sigma_1\ldots\sigma_m}$, and the condition that this system have a solution yields the sought energy levels.

The solution of the problem with a wave function in this form is in general difficult. We therefore first classify the energy levels E_m at $R \gg 1$. With m given, E_m breaks up in the zeroth approximation in $1/R$ into a number of sublevels corresponding to different distributions of the magnons over the pair of impurities. If one of the impurities has l magnons ($0 \leqslant l \leqslant 2S'$), then the energy of such a state is equal to

$$E_{ml}=m\varepsilon_1+E_l^{(1)}+E_{m-l}^{(1)}$$

with ε_1 from (I.7) and $E_l^{(1)}$ from (9). Allowance for the fact that the distance between impurities is finite leads to a splitting of these sublevels.

We take into consideration the following two circumstances: First, the splitting is connected with tunneling of the magnons from one impurity to the other, so that we can confine ourselves to a one-magnon transition, i.e., the transition between the states $(l,\ m-l)$ and $(l\pm 1,\ m-l\mp 1)$. The probability of many-magnon transitions is small in the parameter $1/R$. Second, the considered sublevels must be degenerate, for otherwise the splitting will be of next order of smallness. It is easily seen that the states satisfying both requirements are $(l_0,\ l_0+1)$ and $(l_0+1,\ l_0)$, i.e., $l_0=\frac{1}{2}(m-1)$, which is possible only if m is odd. We note that these sublevels are lowest in energy at the given m. Thus, in the first-order approximation in $1/R$, the energy sublevels do not split if m is even, and if m is odd only the ground sublevel is split.

To find the splitting of these sublevels we write down the wave function of the system in a form analogous to (7a) $(m=2l+1)$:

$$\varphi_{1\ldots m}=\hat{P}_{1\ldots m}\{\varphi_1^{(1)}\ldots\varphi_{l+1}^{(1)}\varphi_{l+2}^{(2)}\ldots\varphi_m^{(2)}\pm\varphi_1^{(2)}\ldots\varphi_{l+1}^{(2)}\varphi_{l+2}^{(1)}\ldots\varphi_m^{(1)}\}. \quad (10)$$

Here $\varphi_l^{(i)}\equiv\varphi_{\mathbf{r}_l}^{(i)}$ is the same as in (7b), and $\hat{P}_{1\ldots m}$ is the operator of symmetrization with respect to the coordinates $\mathbf{r}_1\ldots\mathbf{r}_m$. We have left out from (10) a normalization factor which is inessential for our purpose. We substitute the wave function (10) in (I.9) and then multiply both sides of the equation by

$$2S_1^{(1)}\varphi_1^{(1)}\ldots 2S_{l+1}^{(1)}\varphi_{l+1}^{(1)}2S_{l+2}^{(2)}\varphi_{l+2}^{(2)}\ldots 2S_m^{(2)}\varphi_m^{(2)}$$

and sum over all the coordinates $\mathbf{r}_1\ldots\mathbf{r}_m$. Here $S_{\mathbf{r}}^{(i)}=S$ at $\mathbf{r}\neq\mathbf{R}_i$ and $S_r^{(i)}=S'$ at $\mathbf{r}=\mathbf{R}_i$, since $\varphi_{\mathbf{r}}^{(i)}$ is the solution of the one-magnon problem for an isolated impurity. Using the normalization condition (I.4) for the functions $\varphi_{\mathbf{r}}^{(i)}$ we obtain as a result $(m=2l+1)$

$$E_m-m\varepsilon_1\pm(l+1)\varepsilon_R=l^2\alpha\pm l(l+1)\varepsilon_R/2S'. \quad (11)$$

Here ε_1, α, and ε_R are given by expressions (I.7a), (I.13), and (6).

The right-hand side of (11) is the result of the term with the magnon-magnon interaction in (I.9). The term $l^2\alpha$ in (11), as can be seen from (9) is equal to $E_l^{(1)}+E_{l+1}^{(1)}$. Therefore the expression for the energy levels at arbitrary m can be written in final form

$$E_{ml}=m\varepsilon_1+E_l^{(1)}+E_{m-l}^{(1)}\mp(l+1)\left(1-\frac{l}{2S'}\right)\varepsilon_R\delta_{m,2l+1}. \quad (12)$$

In (12), $\delta_{m,2l+1}$ is the Kronecker symbol and the energy levels E_m are assigned a second index l in accordance with the classification given above.

The origin of the first three terms in the expression for E_{ml} was explained earlier. It is also clear that the splitting should be proportional to ε_R. It is likewise easy to understand the meaning of ε_R in (12). The splitting is connected with tunneling of one magnon from the impurity with $l+1$ magnons to an impurity with l magnons. Since any of the $l+1$ magnons can take part in the transition, the probability of the transition is proportional to their number multiplied by the probability of finding a free place on another impurity, equal to $(2S'-l)/2S'$, where $2S'$ is the total number of states at one impurity.

Expression (12) goes over into (6) at $l=0$ $(m=1)$. At $l=l_{\max}=2S'-1$, the splitting is the same as in the one-magnon case. Moreover, it is easily seen that the term with the splitting in (12) is invariant to the substitution $l\to l_{\max}-l$. The reason is that when the number of magnons is larger than $2S'$ one can speak of tunneling of "holes" rather than tunneling of magnons. The invariance indicated above is the consequence of the symmetry of the magnons and "holes." The splitting in (12) at half-integer S' is maximal if $l=\frac{1}{2}(2S'-1)$ and is equal to $\mp\varepsilon_R(2S'+1)^2/8S'$. For integer S' the splitting is maximal at $l=S'-1$ and is equal to $\mp\varepsilon_R(S'+1)/2$.

The determination of the energy levels with the aid of a wave function of the type (10) is valid only in the region of a small overlap over the wave functions of the magnons at different impurities. In the general case this calls for an exponentially small overlap, i.e., $\varkappa R\gg 1$. It turns out, however, that the result (12) (at $J'/J\ll 1$) is valid also in the region $1\ll R\ll 1/\varkappa$. For the case $S'=\frac{1}{2}$ this was demonstrated in Sec. 2, where we used only the condition $R\gg 1$ (and $J'/J\gg 1$). At $S'>\frac{1}{2}$ the overlap is apparently small also at $\varkappa R<1$, since the wave function of the magnon is decreased by a factor J'/J already at the site neighboring on the impurity.

To prove this statement, we use another method proposed in I. This method, which is valid also at $R\gtrsim 1$, enables us to obtain a successive expansion in the parameter J'/J. In addition, the results obtained with its aid can be generalized to the case of ferrites. In this method, the spin-wave approximation is used for the matrix spins, while the impurity spin operators are described exactly. The Hamiltonian of the system is written in a form analogous to that given in I. The wave function Ψ_M corresponding to the total spin of the system $(N-2)S+M$ can be represented in a form that coincides formally with (I.16):

$$\Psi_M=\chi_M\Phi_0+\chi_{M+1}\sum_{r\neq\mathbf{R}_1,\mathbf{R}_2}\psi_{\mathbf{r}}\hat{a}_{\mathbf{r}}^{+}\Phi_0+\ldots\ . \quad (13)$$

Here Φ_0 is the wave function of the "vacuum" state for the matrix spins: $\hat{a}_{\mathbf{r}}\Phi_0=0$. The terms not written out in (13) result in higher-approximation corrections to the energy and can be left out at the accuracy of interest to us. Substitution of (13) in the Schrödinger equation yields, just as in I, two equations for the function χ_M and $\psi_{\mathbf{r}}$. Eliminating the function $\psi_{\mathbf{r}}$ we can reduce this system of equations, accurate to terms $\sim(J'/J)^2$, to a single equation

$$\left\{E_M^{(1)}+\frac{\alpha}{2}\sum_{i=1,2}\hat{S}_i^-\hat{S}_i^+-\frac{1}{2}z^2S(J')^2G(\mathbf{R})(\hat{S}_1^-\hat{S}_2^++\hat{S}_1^+\hat{S}_2^-)\right\}\chi_M=0. \quad (14)$$

Here $E_M^{(1)}$ is a correction to the energy and is proportional to $(J')^2$, while $G(\mathbf{R})$ is given by (3) with $E=0$. The "wave function" of the pair of impurities χ_M can be written in the form

$$\chi_M=\sum_\mu A_\mu\chi_\mu^{(1)}\chi_{M-\mu}^{(2)}. \qquad (15)$$

Here A_μ are unknown coefficients, $\chi_\mu^{(i)}$ are the eigenfunctions of the operator $\hat{S}^z_{\mathbf{R}_i}$:

$$\hat{S}^z_{\mathbf{R}_i}\chi_\mu^{(i)}=\mu\chi_\mu^{(i)};$$

the values of μ in (15) are bounded by the conditions $-S'\leqslant\mu\leqslant S'$ and $-S'\leqslant M-\mu\leqslant S'$.

In the case $R\gg 1$ and $l=(m-1)/2$ the equations (14) and (15) for the spectrum are easy to solve, and after adding the correction energy $\propto J'$, which has been left out of (14), we obtain the result (12), the only difference being that the expression for ε_R does not contain the factor $e^{-\varkappa R}$. The reason for its absence is that in the derivation of (14) we have left out terms containing J' raised to a power higher than 2. The expression obtained from (14) for E_{ml} is therefore valid at $\varkappa R\lesssim 1$. In the case of unequal gyromagnetic ratios, $g'\neq g$, for not too strong fields, $g'H\lesssim zSJ'$, expression (12) for the energy levels retains the same form, apart from the substitution $g\to g'$ in ε_1.

4. THERMODYNAMICS OF IMPURITY FERROMAGNET IN THE PAIR APPROXIMATION

We consider temperatures that are low compared with the Curie point $T_c\sim J$ of the pure ferromagnet. The density of the spin waves is then small and they can be described in the spin-wave approximation. The impurity concentration c will be assumed small. Then the free energy, just as in I, can be written in the form

$$F=F_{00}+F_{sw}(T)+F_i(T), \qquad (16)$$

where F_{00} and $F_{sw}(T)$ are given by expressions (I.19) and (I.20), while $F_i(T)$ is the contribution of the local impurity levels. Inasmuch as at the considered small c the concentration broadening of interest to us is connected primarily with pairwise interactions of the impurities, we write down $F_i(T)$ in the form of a sum of free energies of the impurity pairs. The region of applicability of this approximation will be explained below.

Let $F_2(\mathbf{r}_i-\mathbf{r}_j)$ be the free energy of two impurities located at the points $\mathbf{r}_i$ and $\mathbf{r}_j$. Then, in the considered approximation,

$$F_i=\frac{1}{2N}\sum_{ij}F_2(\mathbf{r}_i-\mathbf{r}_j).$$

Changing over in this expression to summation over the relative coordinate $\mathbf{R}_{ij}=\mathbf{r}_i-\mathbf{r}_j$, we write down the expression for F_i in final form

$$F_i=\frac{c}{2}\int F_2(\mathbf{R})W(\mathbf{R})d\mathbf{R}. \qquad (17)$$

Here $W(\mathbf{R})$ is the probability of finding the second impurity at a distance $\mathbf{R}$ from the first. For a Poisson distribution we have

$$W(\mathbf{R})=c\exp(-4\pi cR^3/3). \qquad (18)$$

We note that the transition from summation over the relative coordinate $\mathbf{R}_{ij}$ to integration in (17) and to the continuous distribution (18) is essentially connected with the proposed smallness of the concentration c. It is assumed here that the distances in (17) are large compared with the lattice constant. In particular, the radius of the bound state is $\sim 1/\varkappa\gg 1$ and the average distance between impurities is $\sim c^{-1/3}\gg 1$. In addition, we assume that $c^{1/3}/\varkappa\ll 1$, i.e., the radius of the bound state is small in comparison with the average distance between impurities.

The quantity $F_2(\mathbf{R})$ in (17) is expressed in the usual manner in terms of the partition function of the impurity pair Z_2:

$$F_2(\mathbf{R})=-T\ln Z_2(\mathbf{R})=-T\ln\sum_{m=0}^{4S'}\sum_l\exp(-\beta E_{ml}),\quad \beta=\frac{1}{T}, \qquad (19)$$

where E_{ml} are the energy levels of the bound states at the impurity pair, and are given by formula (12). If we neglect in (12) the term with the splitting, then $Z_2=Z_1^2$, where Z_1 is the partition function of the isolated impurity, and (17) goes over into expression (I.21).

Just as in I, we confine ourselves to the case of weakly-coupled impurities $J'\ll J$, for only in this case are the oscillatory phenomena predicted in I possible at the considered $T\ll T_c$. We confine ourselves below to an investigation of the quantity which seems to be easiest to measure, namely the longitudinal susceptibility χ^{zz}.

We consider first the case $S'=\frac{1}{2}$. From (19) and (12) it is easily seen that Z_2 can be written in the form

$$Z_2=(1+\exp(-\beta E_1^+))(1+\exp(-\beta E_1^-)),\quad E_1^\pm=\varepsilon_1\mp\varepsilon_R. \qquad (20)$$

We then obtain from (17)–(19) for the contribution of the impurities to the longitudinal susceptibility χ_i^{zz}

$$\chi_i^{zz}=\frac{\chi_0}{2}\int\left[\operatorname{ch}^{-2}\left(\frac{\varepsilon_1+\varepsilon_R}{2T}\right)+\operatorname{ch}^{-2}\left(\frac{\varepsilon_1-\varepsilon_R}{2T}\right)\right]W(R)d\mathbf{R},\quad \chi_0=\frac{c(g')^2}{4v_cT}. \qquad (21)$$

Here v_c is the volume of the unit cell, χ_0 is the susceptibility of the system of free spins in a magnetic field $H=0$. Expression (21) does not depend on the sign of $\varepsilon_1=g'H-\gamma$, i.e., the peak in χ_i^{zz} becomes symmetrically smeared out. If we let the temperature T go to zero, then (21) goes over into

$$\chi_i^{zz}=\frac{c(g')^2}{2v_c}\int[\delta(\varepsilon_1+\varepsilon_R)+\delta(\varepsilon_1-\varepsilon_R)]W(R)d\mathbf{R}. \qquad (22)$$

In the approximation linear in the concentration, which was considered in I, the susceptibility χ_i^{zz} has at $T=0$ a δ-function peak, which is obtained from (22) by putting $\varepsilon_R=0$. The presence of this peak is connected with the fact that in the approximation linear in c, as the magnetic field increases, the impurity spins at the point $g'H=\gamma$ flip jumpwise from the "down" position ($g'H<\gamma$) to the "up" position ($g'H>\gamma$). When the finite distance between impurities (finite concentration) is taken into account, the interaction between impurities smears out this so-called metamagnetic phase transition. Expression (22) gives the concentration broadening of the δ-function peak in the pair approximation.

From (22) it is easy to determine the region where the pair approximation is valid. Assume for the sake of argument that $H<H_1=\gamma/g'$. At a fixed distance R, for a selected pair of impurities, the metamagnetic phase transition takes place at $g'H=\gamma-\varepsilon_R=g'H_1-\varepsilon_R$.

If $H_1 - H$ is large enough, so that $g'(H_1 - H) = \varepsilon_R$ corresponds to a distance R between impurities smaller than the average distance $\bar{R} \sim c^{-1/3}$, then it can be assumed that these pairs will make an additive contribution that is independent of the other impurities. In fact, the probability of finding a third impurity located at a distance $R \ll c^{-1/3}$ from the selected pair is small because the concentration c is small. Consequently, the region of applicability of the considered pair approximation is determined by the condition that H be different enough from H_1:

$$g'|H-H_1| \gg \varepsilon(\bar{R}).$$

For the foregoing reasoning to be valid it must be stipulated that the wave functions of the bound states overlap weakly over distances $\bar{R} \sim c^{-1/3}$, and this leads to the condition $c^{1/3}/\varkappa \ll 1$. The inequalities $g'|H - H_1| \gg \varepsilon(\bar{R})$ and $c^{1/3}/\varkappa \ll 1$ give the region of applicability of (22).

We note that formula (22), apart from a coefficient, coincides with the density of states of a particle in a random potential in the pair approximation, if ε_1 is taken to mean the proximity of the particle energy to the energy of the bound state on the attraction impurity. In the region $\varepsilon \gg \varepsilon(\bar{R})$ where it is valid, formula (22) reduces, apart from a coefficient, to the expression obtained by I. Lifshitz[5] for the density of states.

Our main task is to estimate the concentration broadening of the δ-function peak in the susceptibility. Since formula (22) ceases to hold in the region of the maximum χ_i^{zz} as $\Delta H = H - H_1 \to 0$, we use the method used by Maradudin *et al.*[7] to estimate the width of the maximum. We define the half-width of the peak Γ_1 in terms of the first moment of the function χ_i^{zz}:

$$\Gamma_1 = \frac{1}{2}\overline{|g'\Delta H|}$$

$$= \frac{1}{2}\int \chi_i^{zz}(H) g'|\Delta H| dH \Big/ \int \chi_i^{zz}(H)\,dH = \frac{1}{2}\int \varepsilon_R W(R)\,d\mathbf{R}. \qquad (23)$$

An estimate of (23) using (6) shows that at $c^{1/3}/\varkappa \ll 1$ the major role in (23) is played by values $R \lesssim 1/\varkappa \ll c^{-1/3}$, i.e., the conditions for the applicability of (22) are satisfied. As a result we obtain for Γ_1

$$\Gamma_1 = 3cS'J'. \qquad (24)$$

If $T < \Gamma_1$, then the observed broadening of the peak in $\chi_i^{zz}(H)$ will be of concentration origin.

We consider now the general case $S' > \frac{1}{2}$. At low temperatures $T \ll \alpha$, the dependence of χ_i^{zz} on H, just as in I, is oscillatory with maxima at the points[1] $g'H_n = \gamma - n\alpha$, $n = 0, 1, \ldots, 2S' - 1$. The susceptibility χ_i^{zz} near the n-th maximum is given by formulas similar to (21) and (22), except that ε_1 is replaced by $g'(H - H_n)$ and the coefficient ε_R depends on n (in accordance with expression (12)). The condition for the validity of these formulas at $c^{1/3}/\varkappa \ll 1$ is

$$\varepsilon(\bar{R}) \ll g'|H-H_n| \ll \alpha.$$

Thus, for example, at $S' = \frac{3}{2}$ the value of χ_i^{zz} as a function of H will have three maxima. The outer maxima are described by formulas (21) and (22) with the substitutions $\varepsilon_1 \to g'(H - H_1)$ and $\varepsilon_1 \to g'(H - H_3)$, while the half-widths coincide with (24). The expression for χ_i^{zz} near the central maximum is obtained from (21) and (22) by making the replacements $\varepsilon_1 \to g'(H - H_2)$ and $\varepsilon_R \to \frac{4}{3}\varepsilon_R$, and its half-width is equal to $\Gamma_2 = 4\Gamma_1/3$.

The oscillations of the susceptibility (as $T \to 0$) will be noticeable if the sum of the half-widths of the neighboring maxima is smaller than the distance between them: $\Gamma_1 + \Gamma_2 < \alpha$, from which we obtain an approximate estimate for the concentration:

$$c \lesssim c_0 = {}^6/_7 (I_W - 1) J'/S'J. \qquad (25)$$

Here I_W is the Watson integral; its values are given in I. At arbitrary S', the inequality (25) is the condition for the resolution of the extreme maximum and the maximum that follows it. With increasing n, the width of the peaks increases and reaches a maximum at the center of the "comb" when $n \sim S'$. Thus, at $S' \gg 1$ we have $\Gamma_{\max} \approx S'\Gamma_1/2$, and the condition for the resolution of the central maxima is

$$c \lesssim 2(I_W - 1) J'/(S')^2 J. \qquad (26)$$

Since the area of each peak is approximately the same, the height of the central maxima at the considered $T < \Gamma_1$ is apparently smaller than that of the outer maxima. It is natural to expect the results of I to be applicable at higher temperatures $T > \Gamma_{\max}$, when the temperature broadening is larger than the concentration broadening.

Since the characteristic dimension of the bound state of the magnon is $1/\varkappa \gg 1$, it follows that, just as in I, our results are apparently qualitatively valid also for ferrites. Using parameter values typical of iron garnets with raw-earth impurities[8,4] (for example $Y_3Fe_5O_{12}$ with Gd impurities), $J'/J = 0.1$, and putting $S' = \frac{3}{2}$, we obtain from (25) a limit on the concentration: $c \lesssim 0.03$ (primitive cubic lattice) and $c \lesssim 0.02$ (BCC and FCC).

Apropos these estimates we note the following. First, at $J'/J = 0.1$ the condition $\varkappa \ll 1$ is not very well satisfied. Thus, for a primitive cubic lattice we have $\varkappa = (6J'/J)^{1/2} \lesssim 0.8$. Therefore the estimates (25) and (26) are only semi-quantitative. The second and more significant remark is connected with the method of determining the width of the peaks. If the half-width is defined not in terms of the first moment, as above, but for example in terms of the second moment of the function χ_i^{zz}, then the upper bound of the concentration will be different than in (25). However, the numerical estimate for c at the chosen parameters J'/J and S' remains of the same order of magnitude. It is therefore natural to assume that expression (25) gives the correct order-of-magnitude estimate of the impurity-concentration region in which experimental observation of susceptibility oscillations is possible.

In conclusion, we thank V. G. Vaks for valuable advice and interest in the work.

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[1] We take the opportunity to correct errors noted in I. In the last term of expression (I. 7a) for ε_1 it is necessary to replace S with S'. In formula (I. 17b) it is necessary to omit the term in the square bracket, and set the right-hand side equal to zero. On page 1142 [558 of the translation] the seventh line from below [3rd line from top, left column] the

expression for H_n should take the form $g'H_n=\gamma-n\alpha$. Everywhere in Secs. 3 and 4, and particularly in Figs. 3–5, α should be replaced by $\alpha/2$.

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Effect of impurities on the thermoelectric properties of tin

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The thermal conductivity and thermoelectric power of pure tin ($\rho_{273K}/\rho_{4.2K}\sim 6\cdot 10^4$) and of tin containing up to 0.2% of Cd, In, Sb, Te or Pb are measured in the 3.7–7.2°K range. The electronic part of α_e of the thermoelectric power is separated from the component α_{ph} due to electron dragging by the phonons. It is found that α_e, α_{ph} and W_{ph} (the thermal resistance due to electron scattering by phonons) depend on the impurity concentration and increase by several times when ρ_0 is increased up to about 10^{-8} Ω·cm. The changes in α_{ph} and W_{ph} are ascribed to suppression of the anisotropy in the distribution of the nonequilibrium electrons. The variation of α_e is ascribed to the dependence of $\partial \ln \tau_{imp}/\partial \ln E$ on the scattering-center charge (τ_{imp} is the relaxation time for electron scattering by impurities). The values of $\partial \ln \tau/\partial \ln E$ for Cd, In, Sb, Te and Pb impurities in tin and for pure tin are 3.3, 3.1, −1.5, −3.7, 0.3 and −2.4 respectively, on the assumption that $\partial \ln S/\partial \ln E = 1$.

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The thermoelectric power is one of the most sensitive kinetic electronic properties of a metal. Its value depends on the type of impurities introduced into the metal, on the electron structure of the metal in an external magnetic field, etc. All these features of thermoelectric power were observed in various experimental studies devoted to its properties (for a review see[1]), but until recently there were practically no systematic investigations of the thermoelectric power of extremely pure metals or of metals with small amounts of impurities at low temperatures.

The present study was undertaken mainly for the purpose of determining the dependence of the thermoelectric power on a small number of impurities. The investigated object was tin, since we had at our disposal material with an initial impurity concentration less than 10^{-4}% ($\rho_{273\,K}/\rho_{4.2\,K}=6\cdot 10^4$). The impurities employed were In, Sb, Cd, Te and Pb. It is obvious that In, Cd, Sb, and Te differ from tin primarily in charge, whereas lead differs in atomic weight. The known published data on the thermoelectric power of tin at low temperatures[2,3] are insufficient and contradictory.

MEASUREMENT PROCEDURE, SAMPLES

The differential thermoelectric power

$$\alpha=\Delta U/\Delta T\,[\mathrm{V/^\circ K}]. \quad (1)$$

where ΔU is the voltage on the sample along which the temperature difference ΔT is produced, is small and amounts to approximately 10^{-8} V/°K at 4.2 °K. Since in the course of the measurements usually $\Delta T\sim 10^{-2}$ °K, to determine α it becomes necessary to measure voltage differences down to $\sim 10^{-12}$ V.

We determined ΔU by a known method with the SKIMP installation[4] as the null indicator (Fig. 1). The sensitivity of the setup was limited by thermal noise in the resistor R_N and amounted to $\sim 10^{-13}$ V. The low-resistance resistor was a plate 1×1 cm of brass foil 0.25 mm thick. Leads of superconducting wire BT-60 were welded to the plate on both sides, one on top of the other, over the entire length. The resistance between these wires perpendicular to the plane of the plate was 1.28×10^{-5} Ω at 4.2 °K. The resistance was constant during the entire measurement cycle. Random changes of the resistance R_N when installed in the cryostat were possible. These changes of resistance took place in our case during the initial stage of the work. As a result, all the thermoelectric-power values cited by us earlier[5] must be multiplied by 1.177.

The temperature gradient was produced in the apparatus shown in Fig. 1. The sample was thermally insulated from the surrounding helium by means of a dewar turned upside down. The copper rod 4 soldered