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IMPURITY ATOM IN A FERROMAGNETIC CRYSTAL WITH NEGATIVE EXCHANGE INTERACTION

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A spin-wave theory is developed for a ferromagnetic cubic crystal containing an impurity atom with negative exchange interaction with the matrix. It is shown that owing to zero-point oscillations of the spin system the ground state of such a crystal is magnetically inhomogeneous. This signifies that the impurity spin projection on the direction of the spontaneous moment is smaller than its maximum value and the decrease is compensated by nonuniform contraction of the matrix atom spins. It is shown that the effect is related to s-type spin system oscillations (in the state terminology used for describing a crystal containing an impurity with ferromagnetic interaction). The temperature dependence of spontaneous crystal magnetization in the range of validity of the Bloch law is calculated and the corresponding results are compared with the case of a ferromagnetic impurity. From measurements of the saturation magnetization at zero temperature and of the temperature dependence of magnetization of a crystal containing a low impurity concentration, one can determine the impurity atom spin and its exchange integral with the matrix.

1. INTRODUCTION

In a series of papers[1-4] the authors have developed a spin-wave theory of a ferromagnetic crystal containing an impurity atom with a distinct value of spin S′ and an exchange integral I′ with its nearest neighbors in the matrix. In this theory it was assumed that I′ > 0, so that in the ground state the spin of the impurity atom is directed along the total spontaneous moment of the crystal. In the case of a negative exchange interaction of the impurity atom with the matrix the impurity spin should have in the ground state, obviously, a negative projection on the direction of the spontaneous moment; however, there is no reason to believe that the absolute magnitude of this projection is the same as S′, as it is in the case of ferromagnetic coupling of the impurity with the matrix.

In order to find the ground state of the system it is necessary to solve for the eigenvalues and eigenfunctions of the exchange Hamiltonian

$$\mathscr{H} = -I \sum_{n,\Delta}{}' (\mathbf{S}_n \mathbf{S}_{n+\Delta}) + 2|I'| \sum_{\Delta} (\mathbf{S}_0 \mathbf{S}_{0+\Delta}), \qquad (1.1)$$

in which $\mathbf{S}_n$ is the spin operator of an atom on site n (the impurity atom is situated at site n = 0): Δ signifies summation over the nearest neighbors; the prime on the first summation means that $n \neq 0$ and $n + \Delta \neq 0$.

From the classical point of view, when the spin vectors $\mathbf{S}_n$ are treated as c-numbers, the minimum of the energy (1.1) corresponds to the state in which the spin of the impurity atom is completely inverted with respect to the totality of the ordered spins of the matrix. This state corresponds to a total spin of the system

$$S^0 = (N-1)S - S', \qquad (1.2)$$

where N is the number of magnetic atoms in the crystal. We shall assume that the true ground state corresponding to the quantum Hamiltonian (1.1) is characterized by this same magnitude of total spin (1.2). However, since in such a system of oppositely oriented spins it is inevitable that zero-point vibrations arise (as in the case of ferri- and antiferromagnets), causing a reduction of the spins (more accurately, of their projections on the direction of the spontaneous moment), the ground state should be inhomogeneous. This is because the total spin of the system is a conserved quantity, since its operator commutes with the exchange Hamiltonian. Hence, if exchange interaction causes contraction of the spin projection of the impurity, this contraction must be compensated by contractions of the spins in the matrix; it follows, keeping in mind the limiting transition $N \to \infty$, that the ground state of the system must be inhomogeneous.

In this paper we calculate with the aid of the retarded Green functions the projection of the spin of the impurity site and the matrix atoms surrounding it as a function of distance from the impurity, and investigate the temperature dependence of the spontaneous magnetization of a crystal containing a low concentration of impurity atoms. We have traced through in detail the distinguishing characteristics of the Green functions for a crystal with an "antiferromagnetic" impurity, which lead to a contraction of the spins. The values of the contractions found by this theory agree with those obtained by Ishii et al.[5] by means of a completely different approach.

2. THE GREEN FUNCTION FOR SINGLE-PARTICLE SPIN EXCITATIONS

Following the assumption about the character of the ground state of the system, we transform from the spin operators S_n and S_0 to a single system of Bose spin deviation operators a_n and a_n^+, introducing them through the Holstein-Primakoff relations[6] 1)

$$S_n^z = S - a_n^+a_n, \quad S_n^+ = (2S)^{1/2}a_n, \quad S_n^- = (2S)^{1/2}a_n^+,$$

$$n \neq 0; \quad S_0^z = -S' + a_0^+a_0, \quad S_0^+ = (2S')^{1/2}a_0^+,$$

$$S_0^- = (2S')^{1/2}a_0, \tag{2.1}$$

where the chosen form of the transformation of the spin of the impurity atom takes into account that the deviation arising at the impurity site increases the z projection of the total spin of the crystal. This leads to the appearance in the Hamiltonian (1.1), expressed in the spin-wave approximation through the operators a_n and a_n^+, of terms of the type $a_0a_{0+\Delta}$(and their Hermitian conjugates), which are characteristic of the Hamiltonian of an antiferromagnetic crystal, along with the usual terms of the type $a_n^+a_m$.

Let us consider the single-particle advanced Green function $G_{nm}(t-t')$, defining it in the following form:[7]

$$G_{nm}(t-t') = i\theta(t'-t)\langle[a_n(t), a_m^+(t')]\rangle, \tag{2.2}$$

where all symbols have their customary significance. It is easy to see that the equation of motion for $G_{nm}(t-t')$ leads to another Green function

$$K_{nm}(t-t') = i\theta(t'-t)\langle[a_n^+(t), a_m^+(t')]\rangle. \tag{2.3}$$

By setting up the equation of motion for it, we obtain a pair of closed equations for G_{nm} and K_{nm}. We expand the function $G_{nm}(t-t')$ in a Fourier integral:

$$G_{nm}(t-t') = \frac{1}{2\pi}\int_{-\infty}^{+\infty} G_{nm}(E)e^{-iE(t-t')}dE \tag{2.4}$$

(we expand $K_{nm}(t-t')$ in analogous fashion). Performing all the standard operations for setting up equations for Green functions, we write them down in symbolic form:

$$\begin{gathered}[G^0(E)]^{-1}G(E) = 1 + V'G(E) + V''K(E),\\ [G^0(-E)]^{-1}K(E) = V'K(E) + V''G(E).\end{gathered} \tag{2.5}$$

All the quantities here are N × N matrices. The matrix elements of G(E) and K(E) are given by the relations (2.2), (2.3), and (2.4); $G^0(E)$ is the Green function G(E) for an ideal crystal:

$$G^0_{nm} = \frac{1}{N}\sum_{\mathbf{k}}\frac{e^{i\mathbf{k}(\mathbf{n}-\mathbf{m})}}{E-\varepsilon_{\mathbf{k}}}, \tag{2.6}$$

where

$$\varepsilon_{\mathbf{k}} = 2SI\sum_{\Delta}(1-e^{i\mathbf{k}\Delta}) \tag{2.7}$$

is the energy of the spin wave in it (we shall consider the case of a simple cubic lattice later).

The quantities V′ and V″ describe the perturbation due to the impurity, and in the nearest-neighbor approximation they are represented by matrices of dimension (z + 1) × (z + 1). In the case of a cubic lattice V′ and V″ have the form

$$V' = 2SI\begin{vmatrix} \lambda z & 1 & 1\ldots 1\\ 1 & \rho & 0\ldots 0\\ 1 & 0 & \rho\ldots 0\\ \cdot & \cdot & \cdot\;\cdot\;\cdot\;\cdot\\ 1 & 0 & 0\quad\;\rho \end{vmatrix}, \tag{2.8}$$

$$V'' = 2SI\begin{vmatrix} 0 & 1+\gamma & 1+\gamma\ldots 1+\gamma\\ 1+\gamma & 0 & 0\;\;\ldots\;\;0\\ 1+\gamma & 0 & 0\;\;\ldots\;\;0\\ \cdot\;\cdot\;\cdot & \cdot\;\cdot\;\cdot & \cdot\;\cdot\;\cdot\;\cdot\;\cdot\;\cdot\\ 1+\gamma & 0 & 0\;\;\ldots\;\;0 \end{vmatrix}, \tag{2.9}$$

where for brevity we use the symbols

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[1] At first sight it may appear that the use of the Holstein-Primakoff relations presupposes that in the ground state the matrix has total spin (N − 1)S, and the z component of the impurity spin equals −S′, i.e., the ground state is characterized by the spins of the matrix and impurity separately. We shall show below, however, that the quantum-mechanical averages of the operators $a_n^+a_n$ and $a_0^+a_0$ in the ground state of the crystal are not equal to zero. Together with this there occurs a compensation of the spin contractions in the matrix by a spin contraction of the impurity, so that the ground state is actually characterized by the value of the total spin (1.2). The only limitation on the ground state associated with the application of Eq. (2.1) is the requirement that the values of $a_n^+a_n$ and $a_0^+a_0$ in the ground state of the crystal be small.

$$\lambda = \frac{|I'|}{I} - 1, \quad \rho = \frac{|I'|S'}{IS} - 1,$$

$$\gamma = \frac{|I'|}{I}\left(\frac{S'}{S}\right)^{1/2} - 1. \qquad (2.10)$$

It should be noticed that formally these parameters λ, ρ, and γ coincide with those introduced by Wolfram and Callaway[8] for a "ferromagnetic impurity." However, in the case of an "antiferromagnetic" impurity the perturbation they determine is completely different, and only in the limiting case of a nonmagnetic impurity, when $S' = 0$ and $I' = 0$, do we have agreement with the result which one can also obtain by starting with a "ferromagnetic" impurity.

From Eq. (2.5), it is easy to obtain

$$G(E) = \frac{1}{1 - G^0(E)V(E)} G^0(E). \qquad (2.11)$$

Here V(E) is the true perturbation operator:

$$V(E) = V' + V'' \frac{1}{1 - G^0(-E)V'} G^0(-E)V''. \qquad (2.12)$$

Thus, for an "antiferromagnetic" impurity the symbolic solution for the Green function G(E) has the same form (2.11) as in the case of a "ferromagnetic" impurity, corresponding to the Dyson equation; however, the expression for the perturbation operator is described by the more complicated relation (2.12), which depends on E.

Direct calculation of the perturbation matrix (2.12) and then of the matrix elements of the Green function G(E) according to Eq. (2.11) is extremely complicated. However, use of a unitary matrix satisfying the point symmetry of the cubic group greatly simplifies the task of finding G(E), although even in this case long calculations are still required. For a simple cubic lattice one can, after all these calculations, represent G_{nm} in the form of several contributions:

$$G_{nm} = G_{nm}^0 + \Delta_{nm}(s) + \Delta_{nm}(p) + \Delta_{nm}(d), \qquad (2.13)$$

which belong to the irreducible representation of the cubic point group Γ_1, Γ_{15}, and Γ_{12}, or in the nomenclature used by Wolfram and Callaway and earlier by us, contributions of the s, p, and d type. Here

$$\Delta_{mn}(s) = \frac{1}{B(E)B(-E)}\left\{\left[(2+\lambda+\rho)E - \rho\frac{E^2}{2ISz}\right]\right.$$

$$\times B(-E)G_{n0}^0 G_{m0}^0 - \left(1+\lambda-\rho\frac{E}{2ISz}\right)$$

$$\times B(-E)\left[G_{n0}^0(E)\delta_{0m} + \delta_{n0}G_{m0}^0(E)\right]$$

$$+\left(1+\lambda-\rho\frac{E}{2ISz}\right)B(-E)G_{00}^0(E)\delta_{n0}\delta_{m0}$$

$$\left.+\left[\frac{\rho}{2ISz}+\left(1+\rho+\rho\frac{E}{2ISz}\right)G_{00}^0(-E)\right]B(E)\delta_{n0}\delta_{m0}\right\}, \qquad (2.14)$$

$$\Delta_{nm}(p) = 2IS\frac{\rho}{2}\frac{1}{D_p}\left\{(G_{n1}^0 - G_{n2}^0)(G_{m1}^0 - G_{m2}^0)\right.$$

$$+ (G_{n3}^0 - G_{n4}^0)(G_{m3}^0 - G_{m4}^0) + (G_{n5}^0 - G_{n6}^0)$$

$$\left.\times (G_{m5}^0 - G_{m6}^0)\right\}, \qquad (2.15)$$

$$\Delta_{nm}(d) = 2IS\frac{\rho}{4}\frac{1}{D_d}\{(G_{n3}^0 + G_{n4}^0 - G_{n5}^0 - G_{n6}^0)$$

$$\times (G_{m3}^0 + G_{m4}^0 - G_{m5}^0 - G_{m6}^0)$$

$$+ {}^1/_3(2G_{n1}^0 + 2G_{n2}^0 - G_{n3}^0 - G_{n4}^0 - G_{n5}^0 - G_{n6}^0)$$

$$\times (2G_{m1}^0 + 2G_{m2}^0 - G_{m3}^0 - G_{m4}^0 - G_{m5}^0 - G_{m6}^0)\}. \qquad (2.16)$$

In the denominators of these expressions are found the quantities:

$$B(E) = 1 + \lambda - \rho\frac{E}{2ISz} - \left[(2+\lambda+\rho)E - \rho\frac{E^2}{2ISz}\right]$$

$$\times G_{00}^0(E), \qquad (2.17)$$

$$D_p(E) = 1 - 2IS\rho\,[G_{00}^0(E) - G_{12}^0(E)], \qquad (2.18)$$

$$D_d(E) = 1 - 2IS\rho\,[G_{00}^0(E) + G_{12}^0(E) - 2G_{13}^0(E)], \qquad (2.19)$$

the zeroes of which are determined by the poles of the Green functions corresponding to states of the s, p, and d type. The expressions (2.15), (2.16), as well as (2.18) and (2.19), which are associated with p and d states, coincide exactly with the corresponding expressions for a "ferromagnetic" impurity[3] and will therefore not be examined here. As was shown earlier,[3] resonant levels of the p and d type do not exist near the bottom of the band; hence, by restricting ourselves to weakly excited states of the system, we shall keep in mind that the quantities $\Delta_{nm}(p)$ and $\Delta_{nm}(d)$ do not have singularities near the bottom of the band of the quasi-continuous excitation spectrum.

As regards expressions (2.14) and (2.17), which characterize the s state, they are essentially different from those for the ferromagnetic impurity. It can be shown that the equation

$$\operatorname{Re} B(E) = 0 \qquad (2.20)$$

(analogous to the equation $\mathrm{Re}D_s(E) = 0$ for a ferromagnetic impurity) has, in addition to possible solutions in the positive region corresponding to resonant or local s type states, also always has a negative solution $E_s < 0$. These solutions can be obtained in the general case only numerically; however, for a weak impurity

$$|I'|S'/IS \ll 1 \tag{2.21}$$

there exists a negative solution of (2.17) near the bottom of the spin-wave band approximately equal to

$$E_s \approx -2|I'|Sz. \tag{2.22}$$

(We recall that for a ferromagnetic impurity in the analogous case the energy of the resonant s level near the bottom of the band was found to be $E_s \approx 2I'Sz$.) At the same time it is easy to see that even for a weak impurity there is no positive solution of Eq. (2.20) near the bottom of the band.

Further, as is seen from (2.14), the poles of the Green function G_{nm} associated with s states are determined not only from Eq. (2.20), but also from the equation

$$\mathrm{Re}\,B(-E) = 0. \tag{2.23}$$

The situation becomes much clearer if in accordance with (2.13) and (2.14) we write an expression for the diagonal elements of G_{nm} corresponding to the impurity ($n = 0$) and the atoms of the matrix ($n \neq 0$):

$$G_{nn}(E) = G_{00}{}^0(E) + \frac{[(2+\lambda+\rho)E - \rho E^2/2ISz][G_{n0}{}^0(E)]^2}{B(E)} + \Delta_{nm}(p) + \Delta_{nn}(d), \tag{2.24}$$

$$G_{00}(E) = \frac{(1+\rho+\rho E/2ISz)\,G_{00}{}^0(-E) + \rho/2ISz}{B(-E)}. \tag{2.25}$$

It is seen from this that the Green function for the impurity atom and the matrix atoms has singularities determined respectively by Eqs. (2.23) and (2.20), and to the solutions of one equation there correspond solutions of the other which are equal in modulus but opposite in sign. This is like a ferrite with two antiparallel sublattices, where the poles of the Green function set up for one of the sublattices differ from the poles of the Green function of the other sublattice only in sign (see, for example, [9]).

3. NEGATIVE POLES AND CONTRACTION OF SPINS

Here we shall show that the negative poles of the the Green functions G_{00} and G_{nm} are associated with the contractions of the spin projections of the atoms of the impurity and the matrix from their maximum values S' and S already at zero temperature, i.e., they are associated with a change of the ground state of the system. To this end, we shall calculate first the temperature correlations $\langle a_0^+ a_0\rangle$ and $\langle a_n^+ a_n\rangle$, using the known spectral relations.[7] We have, in particular, an expression

$$\langle a_0^+ a_0\rangle = \frac{1}{\pi}\int_{-\infty}^{+\infty} \frac{\mathrm{Im}G_{00}(E-i\varepsilon)}{e^{E/kT}-1}\,dE \tag{3.1}$$

an expression which is conveniently represented in the following form:

$$\langle a_0^+ a_0\rangle = \frac{1}{\pi}\int_0^{\infty} [\mathrm{Im}\,G_{00}(E-i\varepsilon) - \mathrm{Im}\,G_{00}(-E-i\varepsilon)]\frac{dE}{e^{E/kT}-1} + \langle a_0^+ a_0\rangle_{T=0}, \tag{3.2}$$

where the term

$$\langle a_0^+ a_0\rangle_{T=0} = -\frac{1}{\pi}\int_0 \mathrm{Im}\,G_{00}(-E-i\varepsilon)\,dE \tag{3.3}$$

describes precisely the change in the spin projection of the impurity site at $T = 0$.

In calculating $\mathrm{Im}\ G_{00}(-E - i\epsilon)$ according to Eq. (2.25) it should be noted that its singularities are associated with the roots of the equation $\mathrm{Re}\ B(E) = 0$. Since the integration in (3.3) is carried out over the positive values of E, the negative root of this equation does not yield a contribution to (3.3), and we have

$$\langle a_0^+ a_0\rangle_{T=0} = \int_0^{\infty} \frac{(1+\rho)(1+\lambda)}{[\mathrm{Re}\,B(E)]^2 + [\mathrm{Im}\,B(E)]^2}\,g_0(E)\,dE. \tag{3.4}$$

The direct calculation of this integral is difficult, since it requires integration in the whole spin-wave band of the quasi-continuous spectrum and inclusion of the contribution of local s type modes if such exists above the band for the given perturbation parameters. This difficulty, however, can be avoided by calculating the expression $\sum_{n=1}^{N-1}\langle a_n^+ a_n\rangle_{T=0}$, which represents the total change of the spin projections of the matrix atoms at zero temperature. As in the case of (3.2), application of the spectral theorem gives

$$\sum_{n=1}^{N-1}\langle a_n^+ a_n\rangle = \frac{1}{\pi}\int_0^{\infty}\Big[\operatorname{Im}\sum_{n=1}^{N-1} G_{nn}(E-i\varepsilon) - \operatorname{Im}\sum_{n=1}^{N-1} G_{nn}(-E-i\varepsilon)\Big]\frac{dE}{e^{E/kT}-1} + \sum_{n=1}^{N-1}\langle a_n^+ a_n\rangle_{T=0}, \tag{3.5}$$

where

$$\sum_{n=1}^{N-1}\langle a_n^+ a_n\rangle_{T=0} = -\frac{1}{\pi}\int_0^{\infty}\operatorname{Im}\sum_{n=1}^{N-1} G_{nn}(-E-i\varepsilon)\,dE. \tag{3.6}$$

We write now expressions for the imaginary parts that appear here in the integrals, which can be calculated most simply by using Eq. (2.24) for G_{nn}. For $E > 0$ we have

$$\frac{1}{\pi}\operatorname{Im}\sum_{n=1}^{N-1} G_{nn}(E-i\varepsilon) = Ng_0(E) - \frac{1}{\pi}\operatorname{Im} G_{00}(-E-i\varepsilon) + \frac{1}{\pi}\operatorname{Im}\frac{B'(E-i\varepsilon)}{B(E-i\varepsilon)} + \frac{3}{\pi}\operatorname{Im}\frac{D_p'(E-i\varepsilon)}{D_p(E-i\varepsilon)} + \frac{2}{\pi}\operatorname{Im}\frac{D_d'(E-i\varepsilon)}{D_d(E-i\varepsilon)}, \tag{3.7}$$

$$\frac{1}{\pi}\operatorname{Im}\sum_{n=1}^{N-1} G_{nn}(-E-i\varepsilon) = \delta(E-|E_s|) - \frac{1}{\pi}\operatorname{Im} G_{00}(E-i\varepsilon). \tag{3.8}$$

Using (3.6) and (3.8), we find

$$\sum_{n=1}^{N-1}\langle a_n^+ a_n\rangle_{T=0} = \frac{1}{\pi}\int_0\operatorname{Im} G_{00}(E-i\varepsilon)\,dE - 1. \tag{3.9}$$

This formula expresses the change in the sum of the spin projections of the matrix atoms through the values of $\operatorname{Im} G_{00}(E-i\epsilon)$ in the positive region of E, whereas formula (3.3) expressed $\langle a_0^+ a_0\rangle$ via $\operatorname{Im} G_{00}(E-i\epsilon)$ in the negative region of E. However, this integral connection exists between them:

$$\frac{1}{\pi}\int_{-\infty}^{+\infty}\operatorname{Im} G_{00}(E-i\varepsilon)\,dE = 1, \tag{3.10}$$

which comes from the definition of the function G_{00} and the spectral theorem. Combining (3.9), (3.10), and (3.3), we obtain the following sum rule:

$$\sum_{n=1}^{N-1}\langle a_n^+ a_n\rangle_{T=0} = \langle a_0^+ a_0\rangle_{T=0}, \tag{3.11}$$

which means that the contraction of the spin projection of the impurity atom should be compensated by the sum of the spin contractions in the matrix, so that the total spin moment of the crystal in the ground state remains equal to $(N-1)S - S'$, as we assumed at the beginning.

The sum rule permits calculation of the contraction of the spin at the impurity site without recourse to Eq. (3.4) but by using the relation (3.9). In fact, it is easy to see from (2.25) that $\operatorname{Im} G_{00} \times (E-i\epsilon) \sim \delta(E-|E_s|)$, where E_s is the negative root of Eq. (2.20) (or a positive root of Eq. (2.23)). As a consequence of this the integration over the energies in (3.9) is removed, and we obtain

$$\langle a_0^+ a_0\rangle_{T=0} = \frac{(1+\rho)(1+\lambda)}{[|E_s|(2+\lambda+\rho)+\rho|E_s|^2/2ISz]\,|\operatorname{Re}B'(-|E_s|)|} - 1 \tag{3.12}$$

It is easy to see that this expression is always positive.

Of course, this result is valid only for such values of the parameters λ and ρ for which $\langle a_0^+ a_0\rangle_{T=0} \ll 1$, as required by the spin wave approximation. Otherwise it is impossible to calculate the spin contraction using the spectrum of single-particle excitations.

We now calculate the magnitude of the spin contractions as a function of distance from the impurity. As in (3.3) and (3.6) we have

$$\langle a_n^+ a_n\rangle_{T=0} = -\frac{1}{\pi}\int_0^{\infty}\operatorname{Im} G_{nn}(-E-i\varepsilon)\,dE. \tag{3.13}$$

Substituting (2.24) herein, the imaginary part of which contains only the second term, which in turn has the factor $1/B(-E)$ and is therefore proportional to $\delta(E-|E_s|)$ when $E > 0$, we immediately obtain after integration over E

$$\langle a_n^+ a_n\rangle_{T=0} = \frac{(2+\lambda+\rho)|E_s| + \rho|E_s|^2/2ISz}{|\operatorname{Re}B'(-|E_s|)|} \times [\operatorname{Re} G_{n0}^0(-|E_s|)]^2. \tag{3.14}$$

In general, the dependence on n for fixed values of the parameters λ and ρ can be obtained only from numerical calculations of $\operatorname{Re} G_{n0}^0(E)$. In the limit of large distances $R \equiv R_{n0}$ it is possible to make use of the asymptotic Green function G_{n0}^0 for $E < 0$:

$$\operatorname{Re} G_{n0}^{0}(-E) = -\frac{1}{8\pi IS}\frac{a}{R}e^{-\varkappa R}, \qquad \varkappa = \frac{1}{a}\sqrt{\frac{|E_s|}{2ISz}}, \tag{3.15}$$

from which we obtain the asymptote for (3.14):

$$\langle a_n^+ a_n \rangle_{T=0} \sim e^{-2\varkappa R} / R^2. \tag{3.16}$$

Thus, the closer the level E_s is to the bottom of the band, the greater is the distance in the matrix over which the spin contractions are propagated.

The distribution of the spin contractions in the matrix may be characterized by a form factor

$$F(\mathbf{q}) = \sum_{n=1}^{N-1} \langle a_n^+ a_n \rangle_{T=0} e^{-i\mathbf{q}\mathbf{n}}, \tag{3.17}$$

which can be measured experimentally from the incoherent elastic scattering of neutrons from crystals containing a low concentration of impurity. Substituting herein the expression (3.14) and summing over n, it is easy to obtain that

$$F(\mathbf{q}) \sim \frac{1}{N}\sum_{\mathbf{k}} \frac{1}{(|E_s| + \varepsilon_{\mathbf{k}})(|E_s| + \varepsilon_{\mathbf{k}-\mathbf{q}})}. \tag{3.18}$$

This expression can be calculated only numerically.

Note that because of the sum rule we have

$$F(0) = \langle a_0^+ a_0 \rangle_{T=0}. \tag{3.19}$$

4. TEMPERATURE DEPENDENCE OF THE SPONTANEOUS MAGNETIZATION

We now calculate the average spontaneous moment of the crystal; according to (2.1), this is written in the form

$$M = g\mu_0 \left[(N-1)S - S' - \sum_{n=1}^{N-1} \langle a_n^+ a_n \rangle + \langle a_0^+ a_0 \rangle \right] \tag{4.1}$$

(it is assumed that the g factors of the impurity and matrix atoms are the same). The correlations appearing here are given by the relations (3.2) and (3.5). If we substitute (3.7) and (3.8) in them, we may write the temperature dependent part in (4.1) in the form

$$\sum_{n=1}^{N-1} \langle a_n^+ a_n \rangle - \langle a_0^+ a_0 \rangle = \int_0^\infty \left[N g_0(E) + \frac{1}{\pi}\operatorname{Im}\frac{B'(E-i\varepsilon)}{B(E-i\varepsilon)} + \frac{3}{\pi}\operatorname{Im}\frac{D_p'(E-i\varepsilon)}{D_p(E-i\varepsilon)} + \frac{2}{\pi}\operatorname{Im}\frac{D_d'(E-i\varepsilon)}{D_d(E-i\varepsilon)} - \delta(E - |E_s|) \right] \frac{dE}{e^{E/kT} - 1}. \tag{4.2}$$

The contribution of the zero-point vibrations fall out on account of the sum rule (3.11). Here the second, third, and fourth terms respectively describe the contribution of the s, p, and d states to the magnetization; the last term also derives from s states and has the same nature as the spin contraction of the impurity site. It is associated with the negative poles of the Green functions.

The contribution in (4.2) due to p and d states is the same as in a ferromagnet. At low temperatures, when the principal contribution to the integral in (4.2) comes from small E, it is possible to obtain approximately

$$\frac{1}{\pi}\operatorname{Im}\frac{B'(E-i\varepsilon)}{B(E-i\varepsilon)} \approx -\frac{3}{2}\left(1 + \frac{S'}{S}\right) g_0(E),$$

$$\frac{3}{\pi}\operatorname{Im}\frac{D_p'(E-i\varepsilon)}{D_p(E-i\varepsilon)} \approx -\frac{3\rho}{1+\Lambda\rho} g_0(E),$$

$$\frac{2}{\pi}\operatorname{Im}\frac{D_d'(E-i\varepsilon)}{D_d(E-i\varepsilon)} \sim E g_0(E). \tag{4.3}$$

The above equations (4.1) to (4.3) now permit the spontaneous moment of the crystal to be written easily in the form

$$\frac{M}{Ng\mu_0} = (1-c)S - cS' - \left[1 + c\frac{3}{2}\left(-1 - \frac{S'}{S} - \frac{2\rho}{1+\Lambda\rho}\right)\right] \eta\left(\frac{T}{T_C}\right)^{3/2} + c\frac{1}{e^{|E_s|/kT} - 1} \tag{4.4}$$

where we have written c for the quantity 1/N, since it plays the role of the concentration. Now, in Eq. (4.4) c may be understood as a finite, but small impurity concentration for which the interaction between individual impurity atoms may be neglected. The quantity η is the coefficient in the Bloch $T^{3/2}$ law, which determines the temperature dependence of an ideal ferromagnetic crystal.

For comparison, we give the analogous result that we obtained earlier[3] for the case of a ferromagnetic impurity:

$$\frac{M}{Ng\mu_0} = (1-c)S + cS' + \left[1 + c\frac{3}{2}\left(-1 + \frac{S'}{S} - \frac{2\rho}{1+\Lambda\rho}\right)\right] \eta\left(\frac{T}{T_C}\right)^{3/2} \tag{4.5}$$

The essential difference between these results consists in the appearance of the last term in (4.4), which increases the magnetization as the temperature is lowered. This is due to local "demagnetization" of the impurity with temperature, which also leads to a positive contribution to the temperature dependence of the total spontaneous moment of the crystal. The spin-wave approximation requires that the average spin deviation at an arbitrary site, including that of the impurity, be less than one. This immediately leads to the condition

$$(e^{|E_s|/kT}-1)^{-1} \ll 1, \tag{4.6}$$

whence follows the equivalent requirement:

$$kT \lesssim |E_s| \tag{4.7}$$

along with the usual requirement $T \ll T_C$, the Curie point of an ideal crystal. The condition (4.7) imposes certain limitations on the parameters λ and ρ, namely, the exchange interaction of the impurity with the matrix must not be much weaker than that between the matrix atoms. Otherwise the condition of the spin-wave approximation for the impurity atom is destroyed at temperatures when it is still valid for the matrix atoms, and it is then impossible to restrict attention to single-particle excitations of the crystal for the description of its temperature behavior. To describe the temperature behavior of the impurity spin in this situation it would be possible to apply the same method that we used recently for a ferromagnetic impurity.[4]

Equations (4.4) and (4.5) allow the experimental determination of S' from the saturation magnetization at $T = 0$, as well as I' from the temperature measurements of the magnetization in the $T^{3/2}$ law region.

It is not difficult to generalize (4.4) to the case when the g factors of the impurity g' and of the matrix g are not the same. In particular, in this case the contribution of the zero-point vibrations to the spontaneous magnetic moment at zero temperature would not be reduced and would equal

$$\frac{M}{N\mu_0} = (1-c)gS - cg'S' + c(g'-g)\langle a_0^+a_0\rangle_{T=0}. \tag{4.8}$$

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