MAGNETIC IMPURITY LEVELS IN ANTIFERROMAGNETS

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Submitted to JETP editor February 7, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) 51, 1084-1089 (October, 1966)

The problem considered is that of local magnetic excitations in antiferromagnetic crystals with a single impurity center. A general solution is given for the problem of oscillations of the spin system of an antiferromagnetic crystal containing a magnetic impurity atom. It is shown that the Green function of such a crystal satisfies Dyson's equation and can be expressed in terms of the Green function of an ideal crystal. Expressions that determine the local impurity levels are obtained. It is established that some of these local levels can have a value less than the spin-wave gap in antiferromagnets.

IT was first shown by I. M. Lifshitz[1] that in crystals containing impurity atoms, local levels can appear. These papers gave a method of treating such problems and investigated in detail the local levels that arise on the phonon branch of the spectrum. Recently, Lifshitz's idea has been extended to crystal-excitation branches of other types. For example, a number of papers have considered the problem of oscillations of the spin system of a ferromagnetic crystal that contains a magnetic impurity atom[2-5].

The present paper considers the problem of the occurrence of local magnetic levels in antiferromagnetic crystals containing a magnetic impurity atom.

1. THE GREEN FUNCTION OF AN ANTIFERROMAGNETIC CRYSTAL WITH AN IMPURITY ATOM

We consider an antiferromagnetic crystal described by two magnetic sublattices, with spins $|S_1| = |S_2| = S_0$ and with exchange integrals J between nearest neighbors. We suppose that the magnetic impurity atom, with spin S′ and with exchange coupling J′ to the nearest atoms, replaces one of the sublattices in an elementary cell. The Hamiltonian of such a system can be written in the form

$$\mathcal{H} = J\sum_{n,\Delta}\mathbf{S}_n\mathbf{S}_{n+\Delta} - 2J\sum_{\Delta}\mathbf{S}_1\mathbf{S}_{1+\Delta} + 2J'\sum_{\Delta}\mathbf{S}_1'\mathbf{S}_{1+\Delta}$$
$$+ A\sum_{n,\Delta}S_n^{(z)}S_{n+\Delta}^{(z)} - 2A\sum_{\Delta}S_1^{(z)}S_{1+\Delta}^{(z)} + 2A'\sum_{\Delta}S_1'^{(z)}S_{1+\Delta}^{(z)}$$
$$- \mu g H_z\Big(\sum_n S_n^{(z)} + \sum_{n,\Delta}S_{n+\Delta}^{(z)}\Big) - \mu H_z(gS_1^{(z)} - g_1S_1'^{(z)}), \quad (1)$$

where the n-summation is over all the sites of the lattice, the Δ-summation is over the sites nearest to the given one, the index 1 refers to the impurity, and S_1' is the spin operator of the impurity site. The first term in (1) describes the exchange interaction in an ideal crystal; the first and second terms together give the exchange interaction in a crystal in which there is no magnetic atom at all on site 1; the third term is the exchange energy between the impurity atom and the others; the fourth term describes the magnetocrystalline-anisotropy energy of the ideal crystal; the fourth and fifth together give the anisotropy energy when there is no magnetic atom on site 1; the sixth describes the anisotropy energy of the impurity atom. Finally, the last two terms in (1) are the energy of the ideal crystal and of the impurity atom, respectively, in the external magnetic field.

If we take into account the antiferromagnetic distribution of the spins S_n, S_1, and S_1' in relation to the spins $S_{n+\Delta}$ and $S_{1+\Delta}$, then in the spin-wave approximation the Hamiltonian (1) will have the form

$$\mathcal{H} = p_1\sum_{n,\Delta}a_n^+a_n + p_2\sum_{n,\Delta}b_{n+\Delta}^+b_{n+\Delta}$$
$$+ p_3\sum_{n,\Delta}(a_n^+b_{n+\Delta}^+ + a_nb_{n+\Delta}) + \beta a_1^+a_1$$
$$+ p_5\sum_{\Delta}b_{1+\Delta}^+b_{1+\Delta} + p_6\sum_{\Delta}(a_1^+b_{1+\Delta}^+ + a_1b_{1+\Delta}); \quad (2)$$

$$p_1 = (J+A)S_0 + g\mu H_z, \quad p_2 = (J+A)S_0 - g\mu H_z,$$
$$p_5 = 2(J'+A')S_0' - 2(J+A)S_0, \quad p_6 = 2J'(S_0S_0')^{1/2} - 2JS_0,$$
$$p_3 = JS_0, \quad \beta = 2S_0z(J'-J-A+A') + \mu H_z(g-g_1); \quad (2')$$

here g and g_1 are the spectroscopic splitting factors for the moments of the basic and of the impurity atoms, μ is the Bohr magneton, and a_n and b_n are the Bose operators of the spin deviations for the respective sublattices of the antiferromagnet.

On applying the usual scheme for construction of a Green function[6] in the energy representation, we get

$$\alpha G_1^{lm} + p_3 \sum_{\Delta} G_2^{l+\Delta, m} + \left(\beta_1 G_1^{m} + p_6 \sum_{\Delta} G_2^{l+\Delta, m}\right)\delta_{l,1} = +\delta_{lm},$$

$$\gamma G_2^{lm} - p_3 \sum_{\Delta} G_1^{l-\Delta, m} - \sum_{\Delta} (p_5 G_2^{l+\Delta, m} + p_6 G_1^{lm})\delta_{l, l+\Delta} = 0, \quad (3)$$

where δ_{lm} is the Kronecker symbol and

$$\alpha = E + zp_1, \qquad \gamma = E - zp_2,$$

$$G_1^{lm} = \langle\langle a_l^+; a_m\rangle\rangle, \qquad G_2^{lm} = \langle\langle b_l; a_m\rangle\rangle.$$

When J = J′, A = A′, S = S′, and g = g_1, the system of equations (3) reduces to the equations for the Green function of an ideal antiferromagnetic crystal, that is

$$\alpha G_{01}^{lm} + p_3 \sum_{\Delta} G_{02}^{l+\Delta, m} = -\delta_{lm},$$

$$\gamma G_{02}^{lm} - p_3 \sum_{\Delta} G_{01}^{l-\Delta, m} = 0. \quad (4)$$

The solution of these equations has the form

$$G_{01} = -\frac{\gamma}{\alpha\gamma + p_3^- p_3^+}, \qquad G_{02} = -\frac{p_3^-}{\alpha\gamma + p_3^- p_3^+},$$

$$p_3^{\pm} = \sum_{\Delta} p_3 e^{\pm i\Delta\mathbf{k}} \quad (5)$$

or

$$G_{01,2}^{lm} = \frac{1}{N}\sum_{\mathbf{k}} G_{01,2} e^{i\mathbf{k}(\mathbf{l}-\mathbf{m})}. \quad (5')$$

Equations (3) and (4) can be written formally in matrix form:

$$(V_0 + V)\begin{pmatrix} G_1 \\ G_2 \end{pmatrix} = \begin{pmatrix} -1 \\ 0 \end{pmatrix}, \quad (3')$$

$$V_0 \begin{pmatrix} G_{01} \\ G_{02} \end{pmatrix} = \begin{pmatrix} -1 \\ 0 \end{pmatrix}; \quad (4')$$

here

$$V_0 = \begin{pmatrix} \alpha & p_3^+ \\ p_3^- & -\gamma \end{pmatrix},$$

$$V = \left(\begin{array}{ccc:ccc} \beta & 0 & 0 \ldots 0 & 0 & p_6 & p_6 \ldots p_6 \\ 0 & 0 & 0 \ldots 0 & 0 & 0 & 0 \ldots 0 \\ \multicolumn{3}{c:}{\ldots\ldots 0} & \multicolumn{3}{c}{\ldots\ldots\ldots} \\ 0 & 0 & 0 \ldots 0 & 0 & 0 & 0 \ldots 0 \\ \hdashline 0 & 0 & 0 \ldots 0 & 0 & 0 & 0 \ldots 0 \\ p_6 & 0 & 0 \ldots 0 & 0 & p_5 & 0 \ldots 0 \\ \multicolumn{3}{c:}{\ldots\ldots 0} & \multicolumn{3}{c}{\ldots\ldots\ldots} \\ p_6 & 0 & 0 \ldots 0 & 0 & 0 & 0 \ldots p_5 \end{array}\right).$$

By use of (4′), it is easy to obtain the desired solution of equation (3′):

$$\begin{pmatrix} G_1 \\ G_2 \end{pmatrix} = (1 + V_0^{-1}V)^{-1}\begin{pmatrix} G_{01} \\ G_{02} \end{pmatrix}. \quad (6)$$

When S = S′, J = J′, A = A′, and g = g_1, it is seen from (2) that V = 0, and it follows from (6) that

$$\begin{pmatrix} G_1 \\ G_2 \end{pmatrix} = \begin{pmatrix} G_{01} \\ G_{02} \end{pmatrix},$$

that is, we get the solutions for an ideal antiferromagnetic crystal.

It is easily shown that the operator V_0^{-1} can be expressed in terms of the zero-order Green functions as follows:

$$V_0^{-1} = \begin{pmatrix} G_{01} & G_{02} \\ G_{02} & -\alpha\gamma^{-1}G_{01} \end{pmatrix} = G^0. \quad (7)$$

In the matrix V in (6), the rows and columns enumerate the sites occupied by the impurity and by its z nearest neighbors. Hence the matrix V has dimensions (2z + 2) × (2z + 2). Since the matrix V acts on the matrix G^0, which has dimensions N × N (N = number of magnetic sites), it must be filled up with zeros beyond the edges of the meaningful square of dimensions (2z + 2) × (2z + 2).

To find the spectrum of oscillations of the spin system, we use the Green function, given in matrix form by expression (6):

$$\begin{pmatrix} G_1 \\ G_2 \end{pmatrix} = \sum_p \frac{B_{pm}}{D}\begin{pmatrix} G_{01} \\ G_{02} \end{pmatrix}_{pn};$$

$$B_{pm}/D = [(1 + G^0V)^{-1}]_{mp}, \qquad D = \det|1 + G^0V|. \quad (8)$$

Since the matrix V is abridged to dimensions (2z + 2) × (2z + 2), the determinant D of the matrix $1 + G^0V$ also has dimensions (2z + 2) × (2z + 2).

It follows from (8) that in addition to the poles of the ideal crystal[1)] (that is, the poles of G_{01} and G_{02}), there appear additional poles of the Green function. These poles are determined by the equation D = 0.

We shall now calculate the following Green functions, which are necessary for a complete analysis and for consideration of certain other physical problems in antiferromagnets:

$$G_3 = \langle\langle a_l; a_m^+\rangle\rangle, \quad G_4 = \langle\langle b_l^+; a_m^+\rangle\rangle,$$

$$G_5 = \langle\langle b_l^+; b_m\rangle\rangle, \quad G_6 = \langle\langle a_l; b_m\rangle\rangle,$$

$$G_7 = \langle\langle b_l; b_m^+\rangle\rangle, \quad G_8 = \langle\langle a_l^+; b_m^+\rangle\rangle. \quad (9)$$

[1)] The poles of G_{0_1} and G_{0_2} determine the spectrum of spin waves in the impurity-free antiferromagnet.

It is known that $G_3 = G_1(-E)$ and $G_4 = G_2(-E)$. To calculate the remaining Green functions, we shall start with the equations of motion for G_5 and G_6, analogous to (6):

$$\begin{pmatrix} G_5 \\ G_6 \end{pmatrix} = (1 + GV')^{-1} \begin{pmatrix} G_{05} \\ G_{06} \end{pmatrix}. \tag{10}$$

Here

$$G_{05} = -\frac{c}{cf + p_3^- p_3^+}, \quad G_{06} = -\frac{p_3^-}{cf + p_3^- p_3^+},$$
$$f = E + zp_2, \quad c = E - zp_1,$$

$$G = -\begin{pmatrix} G_{05} & -G_{06} \\ G_{06} & fc^{-1} G_{05} \end{pmatrix},$$

$$V' = \left(\begin{array}{ccccc|ccccc} 0 & 0 & 0 & \dots & 0 & 0 & 0 & 0 & \dots & 0 \\ 0 & p_5 & 0 & \dots & 0 & p_6 & 0 & 0 & \dots & 0 \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ 0 & 0 & 0 & \dots & p_5 & p_6 & 0 & 0 & \dots & 0 \\ \hline 0 & -p_6 & -p_6 & \dots & -p_6 & -\beta & 0 & 0 & \dots & 0 \\ 0 & 0 & 0 & \dots & 0 & 0 & 0 & 0 & \dots & 0 \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ 0 & 0 & 0 & \dots & 0 & 0 & 0 & 0 & \dots & 0 \end{array}\right),$$

or in matrix form

$$\begin{pmatrix} G_5 \\ G_6 \end{pmatrix} = \sum_p \frac{B'_{pm}}{D_t} \begin{pmatrix} G_{05} \\ G_{06} \end{pmatrix}_{pn};$$

$$B_{pm'}/D_t = [(1 + GV')^{-1}]_{mp}, \quad D_t = \det|1 + GV'|;$$
$$G_7 = G_5(-E), \quad G_8 = G_6(-E). \tag{11}$$

We shall now analyze the energy of the impurity levels. For this purpose it is sufficient to know the specific forms of D and of D_t.

2. SPECTRUM OF ADDITIONAL POLES OF THE GREEN FUNCTION

The spectrum of poles of the Green function due to an impurity in antiferromagnets will be determined by the equations $D = 0$ and $D_t = 0$. In the case $z = 2$ (which is the only one we shall consider), these equations have the form

a) $D = 1 + \alpha\gamma^{-1} p_5 [G_{01}(2) - G_{01}(0)] = 0,$

b) $D = 1 + p_5 [G_{05}(2) - G_{05}(0)] = 0,$

c) $D = [1 + \beta G_{01}(0) + 2p_6 G_{02}(1)]$
$\times \{1 + 2p_6 G_{02}(1) - \alpha\gamma^{-1} p_5 [G_{01}(0) + G_{01}(2)]\}$
$- 2[p_6 G_{01}(0) + p_5 G_{02}(1)]\{\beta G_{02}(1) - \alpha\gamma^{-1} p_6 [G_{0,}(0)$
$+ G_{01}(2)]\} = 0,$

d) $D = [1 + fc^{-1}\beta G_{05}(0) - 2p_6 G_{06}(1)]$
$\times \{1 - 2p_6 G_{06}(1) - p_5 [G_{05}(0) + G_{05}(2)]\}$
$+ 2[fc^{-1} p_6 G_{05}(0) - p_5 G_{06}(1)]$
$\times \{\beta G_{06}(1) + p_6 [G_{05}(0) + G_{05}(2)]\} = 0.$ (12)

Here we have used the relations

$$G_0(0) = G_0^{11} = G_0^{22} = G_0^{33},$$
$$G_0(1) = G_0^{12} = G_0^{13} = G_0^{21} = G_0^{31},$$
$$G_0(2) = G_0^{23} = G_0^{32},$$

which follow from the symmetry of the linear chain. To simplify the analysis of the formulas obtained, we set $H = 0$; then

$$\alpha = f = E + 2p, \quad \gamma = C = E - 2p,$$
$$G_{01} = G_{05}, \quad G_{02} = G_{06}.$$

We begin our investigation of the impurity level with an analysis of oscillations of types a) and b); this is simpler than analysis of oscillations of types c) and d). On using (5) and (5′) and on carrying out the summation over nearest neighbors, we can write D of a) and D of b) in the form

$$\text{a)} \quad 1 - 2(E + 2p) p_5 \sum_k \frac{\sin^2 k}{E_0^2 - E^2 + 4p_3^2 \sin^2 k} = 0,$$

$$\text{b)} \quad 1 - 2(E - 2p) p_5 \sum_k \frac{\sin^2 k}{E_0^2 - E^2 + 4p_3^2 \sin^2 k} = 0, \tag{13}$$

where $E_0 = \sqrt{2AJ}$ is the size of the energy gap in the spin-wave spectrum; $(E_0^2 + 4p^2)^{1/2}$ defines the upper limit of the band in the spectrum of the antiferromagnet under consideration. Hereafter we shall be interested in those impurity levels whose energy falls in the gap ($E < E_0$, whence it follows that $E \ll 2p$) in the spin-wave spectrum.

Because $p > 0$ and the sum is positive, it follows from (13) that when $p_5 > 0$, only a level of type a) can be inside the energy gap in the spin-wave spectrum. On carrying out the integration in (13), we get the following equation, which relates the energy of the impurity level a) to the "impurity parameter" σ:

$$\sigma = \frac{2p}{E + 2p}\left\{1 - \frac{(E_0^2 - E^2)^{1/2}}{(E_0^2 + 4p_3^2 - E^2)^{1/2}}\right\}^{-1}, \tag{14}$$

where $\sigma = (J'S' - JS)/JS$. It can be seen from (14) that when

$$(1 + \sqrt{A/2J}) > \sigma > (1 - \sqrt{A/2J}) \tag{14′}$$

the level of type a) falls in the gap.

When $p_5 < 0$, on the other hand, a level corresponding to oscillations of type b) can fall in the gap; then there is no type a) level in the gap. The energy corresponding to oscillations of type b) is determined by the equation

$$\sigma = \frac{2p}{E - 2p}\left\{1 - \frac{(E_0^2 - E^2)^{1/2}}{(E_0^2 + 4p_3^2 - E^2)^{1/2}}\right\}^{-1}. \tag{15}$$

The condition for finding a level b) in the gap has the form

$$-(1+E_0/2p) < \sigma < -(1-E_0/2p). \qquad (15')$$

In the case of oscillations of types c) and d), the situation is much more complicated. The position of each of these levels depends on three parameters: p_5, p_6, and β. We have succeeded in analysing these levels only for the single value $S_0' = S_0$; then $p_5 = p_6 = \frac{1}{2}\beta$, and

$$\text{c)}\quad 1+2p_5(E+2p)\sum_k \frac{\sin^2 k}{E^2-E_0^2-4p_3^2\sin^2 k}=0,$$

$$\text{d)}\quad 1+2p_5(E-2p)\sum_k \frac{\sin^2 k}{E^2-E_0^2-4p_3^2\sin^2 k}=0. \qquad (16)$$

On comparing (16) with (13), we perceive that in the case $S_0' = S_0$, the levels c) and d) coincide, respectively, with the levels a) and b).

In (14′) and (15′), $\sqrt{A/2J} \sim 10^{-1}$ to 10^{-2}. It follows from (14′) and (15′) that the condition for finding an impurity level in the energy gap of an antiferromagnet is $J' \sim J$.

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Translated by W. F. Brown, Jr.
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