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PECULIARITIES OF THE SPIN WAVE SPECTRUM OF A FERROMAGNET CONTAINING IMPURITIES AND THE TEMPERATURE DEPENDENCE OF SPONTANEOUS MAGNETIZATION

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A numerical calculation is carried out for the conditions of appearance of virtual and local magnetic oscillations in a Heisenberg ferromagnet with a simple cubic lattice containing an impurity magnetic atom. It is shown that only virtual levels of the s type can arise on the bottom of the spin-wave band and only in those cases when the exchange coupling between the impurity and the atoms of the matrix is weaker than the exchange coupling between the atoms of the matrix. Approximate formulas are also obtained for the density of states near the bottom of the band and, respectively, for the spontaneous magnetization at low temperatures for a ferromagnet with an impurity.

1. AT the present time there exists a general spin-wave theory of a ferromagnetic crystal containing an impurity magnetic atom having a spin and exchange integrals different from those of the atoms of the matrix.[1, 2] If one is limited to the nearest-neighbor approximation for the exchange interactions of the atoms, the perturbation introduced by the impurity is a local one, and all the general conclusions obtained at one time for systems with a continuous spectrum[3] are valid for the analysis of the energy spectrum. In particular, the changes in the density of states of the energy spectrum are resonant in character. For a monoatomic ferromagnetic crystal with a simple cubic lattice the density of states g(E) in the spin-wave spectrum, calculated for one atom (N is the total number of atoms in the crystal), is in this situation described by the following formula:[2]

$$g(E)=g_0(E)+\frac{1}{N\pi}\operatorname{Im}\frac{D'(s)}{D(s)}+\frac{3}{N\pi}\operatorname{Im}\frac{D'(p)}{D(p)}+\frac{2}{N\pi}\operatorname{Im}\frac{D'(d)}{D(d)},\qquad (1)$$

where $g_0(E)$ is the density of spin-wave states of an ideal crystal, the prime indicates a derivative d/dE with respect to energy E, and the quantities D(s), D(p), and D(d) are functions of E (they are defined for the values $E - i\delta$; $\delta = 0^+$)

$$D(s)=1+\varepsilon+\frac{\rho}{2JSz}E-\left[(\varepsilon-\rho)E+\frac{\rho}{2JSz}E^2\right]G_{11}{}^0(E),\qquad (2)$$

$$D(p)=1-\rho\cdot 2JS(G_{11}{}^0(E)-G_{23}{}^0(E)),\qquad (3)$$

$$D(d)=1-\rho\cdot 2JS(G_{11}{}^0(E)+G_{23}{}^0(E)-2G_{24}{}^0(E)).\qquad (4)$$

We denote the spin of an atom of the ideal crystal and the exchange integral between nearest neighbors by S and J, the spin of the impurity atom by S′, and the exchange integral between the latter and its nearest neighbors in the matrix by J′. The perturbation parameters ε and ρ are

$$\varepsilon=J'/J-1;\quad \rho=J'S'/JS-1.\qquad (5)$$

The quantities $G^0_{nm}(E)$ in Eqs. (2)–(4) are the Green functions of the spin waves of the ideal crystal:

$$G_{nm}{}^0(E)=\frac{1}{N}\sum_{\mathbf{k}}\frac{\exp\{i\mathbf{k}(\mathbf{R}_n-\mathbf{R}_m)\}}{E-\mathscr{E}_{\mathbf{k}}},\qquad (6)$$

where $\mathscr{E}_{\mathbf{k}}$ is the energy of the spin wave with quasimomentum **k**. The impurity atom is at the site n = 1. Besides G^0_{11}, we also have G^0_{24}, which couples two arbitrary neighboring atoms in the first coordination sphere, appearing in Eqs. (2)–(4), and G^0_{23} couples two arbitrary opposed atoms (relative to the impurity atom) in the same sphere.

By separating the real and imaginary parts of the expressions

$$D(\mu)=D(\mu,E-i\delta)=\operatorname{Re}D(\mu)+i\operatorname{Im}D(\mu)$$
$$(\mu=s,p,d),$$

we can now write down in these terms each of the additional terms in the density of states (1):

$$\Delta_\mu g(E) \equiv \frac{1}{\pi}\mathrm{Im}\frac{D'(\mu)}{D(\mu)}$$

$$= \frac{1}{\pi}\frac{\mathrm{Re}\,D(\mu)\,\mathrm{Im}\,D'(\mu) - \mathrm{Re}\,D'(\mu)\,\mathrm{Im}\,D(\mu)}{[\mathrm{Re}\,D(\mu)]^2 + [\mathrm{Im}\,D(\mu)]^2}. \qquad (7)$$

If at some point $E = E_\mu^0$ the quantity

$$\mathrm{Re}\,D(\mu, E_\mu{}^0) = 0 \qquad (8)$$

and Im $D(\mu)$ is small at the same time, Eq. (7) has a sharp maximum near this point. The form of the function (7) near the singular point can be obtained approximately by expanding Re $D(\mu, E)$ in a series:

$$\mathrm{Re}\,D(\mu, E) = \mathrm{Re}\,D'(\mu, E_\mu{}^0)\,(E - E_\mu{}^0) + \ldots$$

In this case we have for $E \sim E_\mu^0$

$$\Delta_\mu g(E) = \frac{1}{\pi}\frac{\Gamma_\mu}{(E - E_\mu{}^0)^2 + \Gamma_\mu{}^2}, \qquad (9)$$

and the quantity

$$\Gamma_\mu = -\mathrm{Im}\,D(\mu, E_\mu{}^0)\,/\,\mathrm{Re}\,D'(\mu, E_\mu{}^0) \qquad (10)$$

characterizes the width of the maximum.

If the solution of Eq. (8) E_μ^0 lies above the top of the spin-wave band of the ideal crystal, it is easy to see from Eqs. (2)–(4) that Im $D(\mu, E_\mu^0) \equiv 0$, and hence Γ_μ as well. In this case Eq. (9) reduces to the following

$$\Delta_\mu g(E) = \delta(E - E_\mu{}^0). \qquad (11)$$

This means that the system has an isolated discrete level, called a local level. In a simple cubic crystal there are three types, corresponding to values of μ equal to s, p, and d, and the p-level is triply degenerate, the d-level doubly degenerate. The coefficients 3 and 2 in Eq. (1) in fact indicate just this degeneracy.

If E_μ^0 falls in the band of the continuous spectrum, then Γ_μ is non-zero and the change in the density of states near E_μ^0 is described approximately by the Lorentz form (9). An important fact is that Γ_μ can be not only positive but also negative. If $\Gamma_\mu > 0$, we have a jump in the density of states, and we shall treat this situation as the appearance of a virtual level E_μ^0 in the system. In the opposite case $\Gamma_\mu < 0$, there is a dip in the density, corresponding to the so-called antiresonance point. As a rule, in fact, Eq. (8) has two solutions for any μ, one of which corresponds to the resonant peak in the density of states and the other to the dip, so that the total change in the density of states is equal to zero, since the total number of states both for the ideal crystal and for the crystal with impurity is N.

2. The determination of the energies of the local or virtual states obtained from setting the determinants of (2), (3), and (4) to zero can only be made numerically. The necessary Green functions G_{11}^0, G_{23}^0, and G_{24}^0 (their real and imaginary parts) for the simple cubic lattice have been calculated by Wolfram and Callaway,[1] whose data we shall use here. We begin with an analysis of p- and d-type oscillations, which is simpler than for the s-type. According to (3) and (4), the equations determining the energy of these oscillations can be written in the following form:

$$2/\rho = 4JS\,\mathrm{Re}\,\{G_{11}{}^0(E) - G_{23}{}^0(E)\}\ \ (p), \qquad (12)$$

$$2/\rho = 4JS\,\mathrm{Re}\,\{G_{11}{}^0(E) + G_{23}{}^0(E) - 2G_{24}{}^0(E)\}\ \ (d). \qquad (13)$$

It is seen that the p- and d-type oscillations are characterized by the single parameter ρ (whereas the s-type depend on the two parameters ρ and ε). The parameter ρ can vary in the interval $-1 < \rho < \infty$, hence the quantity $2/\rho$ takes values $-\infty < 2/\rho < -2$ or $0 < 2/\rho < \infty$; thus the interval $(-2, 0)$ is forbidden for the quantity $2/\rho$.

The right-hand sides of Eqs. (12) and (13) are plotted in Fig. 1. The negative values of both curves lie in the region of forbidden values for the parameters $2/\rho$; hence Eqs. (12) and (13) do not have solutions for E near the bottom of the band (the width of the spin-wave band is 24JS). Therefore, we have to conclude that in a simple cubic lattice there are no p- and d-type levels at the bottom of the band. However, they are possible near its top.

It is easy to determine from Fig. 1 the critical values of the parameter $2/\rho$ (and then also $\rho + 1 = J'S'/JS$) for which a local or virtual level only just appears. We thus arrive at the following results:

a) virtual and local p- and d-type levels are absent when

$$0 < J'S'/JS < 4.09\ \ (p), \qquad 0 < J'S'/JS < 2.83$$

b) virtual levels appear near the top of the band when

$$4.09 < J'S'/JS < 5.76\ \ (p), \qquad 2.83 < J'S'/JS < 6.41$$

c) local levels appear when

$$5.76 < J'S'/JS\ \ (p), \qquad 6.41 < J'S'/JS\ \ (d).$$

From this we see that a sufficiently "strong" impurity (in terms of the magnitude of the exchange interaction) is required for the appearance of virtual or local p- and d-levels. The dependence of

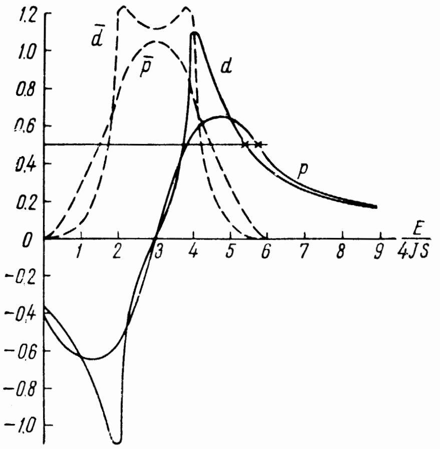

FIG. 1. Graphical solution of Eqs. (12) and (13), which determine the local or virtual p- and d-type levels in the case of the simple cubic lattice. The solid curves describe the righthand sides of Eqs. (12) and (13). The dashed curves give the imaginary parts of the determinants –D(p) and – D(d), viz., curve $\bar{d}$ is

$$4JS\,\mathrm{Im}\{G_{11}^0(E)+G_{23}^0(E)-2G_{24}^0(E)\},$$

and $\bar{p}$ is

$$4JS\,\mathrm{Im}\{G_{11}^0(E)-G_{23}^0(E)\}.$$

From this it is easy to determine the sign of the quenching of the virtual levels.

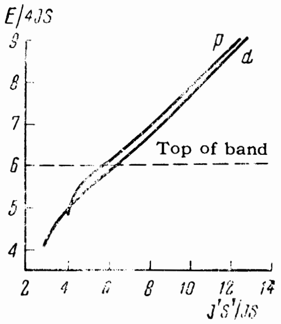

FIG. 2. Energy of the p- and d-levels as a function of the parameter J 'S '/JS.

the energy of p- and d-levels on the parameter J′ S′/JS is shown in Fig. 2.

In the case of the s-oscillations, the situation becomes much more complicated. The position of the local and virtual levels is determined from the equality to zero of the real part of Eq. (2) and therefore depends on two parameters: ε and ρ. Figure 3 shows the results of the solution for several of the most interesting values of the parameters σ = S′/S as functions of J′/J. It is seen from Fig. 3 that the local s-type oscillations exist for a given fixed value of σ, beginning each time from some value of J′/J, which increases with decreasing σ. The larger σ, the higher the energy of the local oscillation (for a fixed J′/J). When $\sigma \geq 1$ the virtual levels near the top of the band do not exist. They appear when $\sigma < 1$ and can go into the band rather deeply.

For the properties of a ferromagnet at low temperatures, the only region where the spin-wave approximation is valid, the virtual levels lying near the bottom of the spin-wave band are of greatest interest. Their energy is shown as a function of J′/J in Fig. 4 for a wide interval of selected values of the parameter σ from 0.1 to 10, and their width Γ_S is shown in Fig. 5. We see that these levels appear only when J′/J < 1 and their energy depends weakly on σ. It can be seen from Fig. 3 that for large values of σ and in a certain region of values of J′/J two s-levels can exist simultaneously: one of them is virtual, the other local.

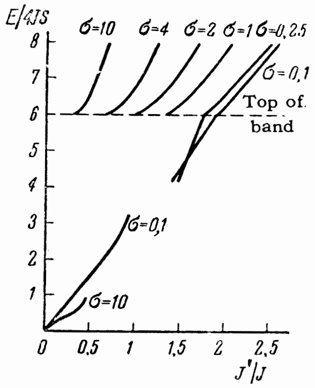

FIG. 3. Dependence of the energy of the s-oscillation on the parameter J ′/J for different values of σ = S ′/S.

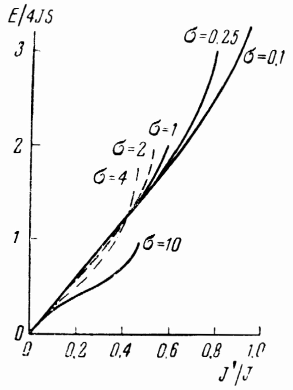

FIG. 4. Dependence of the energy of the virtual s-oscillation on the parameter J ′/J.

A practical conclusion of these numerical calculations is the following: in a cubic crystal there are low-lying virtual s-levels for ratios of the exchange integrals J′/J < 1. Their energy does not depend very strongly on the parameter σ in all cases of practical interest.

Figure 6 shows the results of a numerical calculation of the density of states of a ferromagnetic crystal with impurity concentration c = 0.01, for

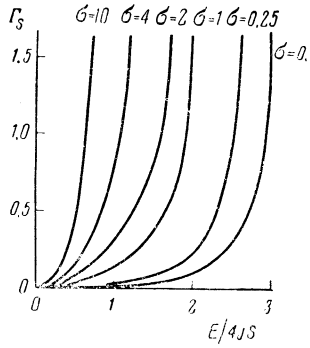

FIG. 5. Dependence of the width $\Gamma_s = -\operatorname{Im} D(s, E^0/4JS)/\operatorname{Re} D'(s, E^0/4JS)$ of the virtual s-oscillation on its energy, determined by the parameter J'/J for different values of σ.

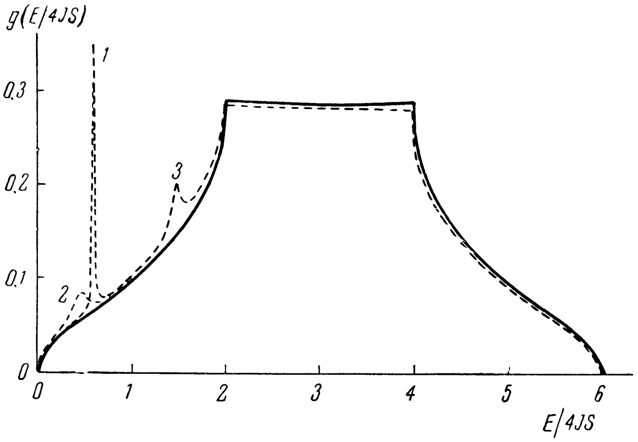

FIG. 6. Density of states $g(E/4JS)$ of a ferromagnetic crystal with impurity concentration $c = 0.01$ for the cases: $1 - J'/J = 0.2$; $\sigma = 0.25$; $2 - J'/J = 0.2$; $\sigma = 4$; $3 - J'/J = 0.5$; $\sigma = 0.25$. The solid curve gives the density of states of the ideal ferromagnetic crystal $g_0(E/4JS)$.

three cases in which virtual levels of the s-type exist.

3. We have investigated numerically the position of the local and virtual levels as a function of the parameters ε and ρ. We now attempt to calculate the change in the density of states near the bottom of the band. It turns out that this can be done analytically if the virtual levels are not located close to the very bottom. In every case we shall consider a region of E such that the lowest virtual level is above it. A still more favorable situation for this approach comes about when, for given perturbation parameters, the resonant states can arise only at the top of the spin-wave band or above it, or, for example, in those cases when the perturbation is insufficiently strong (the parameters ε and ρ small) to cause the formation of resonant states (Eq. (8) cannot be satisfied for all μ at any E), and only leads to a smooth redistribution of the density of states.

The density of states of the ideal crystal can be calculated from the following general formula:

$$g_0(E) = \frac{1}{\pi} \operatorname{Im} G_{11}{}^0(E) = \frac{1}{N} \sum_{\mathbf{k}} \delta(E - \mathscr{E}_{\mathbf{k}}). \tag{14}$$

For small E we can limit ourselves to small values of the quasimomentum, taking for the simple cubic lattice $\mathscr{E}_{\mathbf{k}} = 2JSa^2k^2$ (where a is the lattice parameter). Calculation of the integral over $\mathbf{k}$ in (14) leads to the well-known formula

$$g_0(E) = \frac{1}{4\pi^2 (2JS)^{3/2}} E^{1/2} + \ldots \tag{15}$$

Substituting expression (2) into formula (7) for $\mu = s$ and taking into account the last two relations, we find in the lowest order in $E/2JSz$ the following relation for the change in density of states due to oscillations of the s-type:

$$\Delta_s g(E) \approx 3(\sigma - 1) g_0(E)/2. \tag{16}$$

To calculate the contributions from the p- and d-oscillations it is necessary to find $\operatorname{Im} G^0_{nm}$. It is easy to show that for small E

$$\frac{1}{\pi} \operatorname{Im} G_{nm}{}^0(E) = \frac{1}{N} \sum_{\mathbf{k}} \cos \mathbf{k}(\mathbf{R}_n - \mathbf{R}_m)\, \delta(E - \mathscr{E}_{\mathbf{k}})$$
$$= \left\{1 - \frac{E}{2JS} \frac{(\mathbf{R}_n - \mathbf{R}_m)^2}{6a^2}\right\} g_0(E) + \ldots \tag{17}$$

Substituting expressions (3) and (4) into formula (7) and taking the last relation into account, we find the contribution from one p- and d-state:

$$\Delta_p g(E) \approx \frac{1}{\pi} \frac{\operatorname{Im} D_E'(p)}{\operatorname{Re} D(p, E=0)} \approx -\frac{\rho}{1 + 0.21\rho} g_0(E),$$
$$\Delta_d g(E) \approx 0, \tag{18}$$

or more exactly, $\Delta_d g(E) \sim E g_0(E)/2JS$, and therefore it can be neglected for small E.

Summing the results (16) and (18) and using (1) we express the density of states for small E as

$$g(E) = \left\{1 + \frac{1}{N} \frac{3}{2}\left(\sigma - 1 - \frac{2\rho}{1 + 0.21\rho}\right)\right\} g_0(E). \tag{19}$$

In the presence of many impurities in the crystal, $1/N$ in (19) must be replaced by the concentration of impurities c.

Depending on the parameters σ and ρ there can be different signs in the additional term in (19), i.e., in the presence of impurities the density of states may either increase or decrease. If the impurity atoms are non-magnetic ($\sigma = 0$, $\rho = -1$), the additional term becomes positive.

4. We now calculate the spontaneous magnetiza-

tion M of the crystal for the low-temperature region ($T \ll T_C$, the Curie temperature), where the spin-wave approximation is valid. We use the general relation

$$\frac{M}{Ng\mu_0} = (1-c)S + cS' - \int\limits_0^\infty \frac{g(E)\,dE}{e^{E/kT}-1}. \qquad (20)$$

In the integral the values $E \lesssim kT$ are important; therefore, if in this energy interval there is no virtual state, we can substitute into (20) the relation for the density of states (19) (in which 1/N is replaced by c). As a result we have the usual Bloch law for the temperature dependence of M:

$$\frac{M}{Ng\mu_0} = (1-c)S + cS' - \left\{1 + c\cdot\frac{3}{2}\left(\sigma - 1 - \frac{2\rho}{1+0.21\rho}\right)\right\} \times \gamma\left(\frac{T}{T_c}\right)^{3/2}, \qquad (21)$$

where γ is a numerical coefficient for the ideal crystal. A comparison of the temperature dependences of M of the ideal crystal and the crystal with impurity for different concentrations enables one to determine the parameter ρ from Eq. (21), i.e., in essence the ratio J′/J.

If, however, there are virtual levels at the bottom of the band in the impurity ferromagnet,[1)] then as soon as the thermal energy kT becomes with increasing temperature comparable to the energy of the virtual level E_S^0, formula (19) for g(E) already becomes inapplicable, and the principal change in the density of states is found to have a maximum in the vicinity of E_S^0 of the type (9) (see [4]). If, neglecting the width of the maximum Γ_S, (9) is approximated by the functions $\delta(E - E_S^0)$, an additional term $-c\{\exp(E_S^0/kT) - 1\}^{-1}$ results from the integral in (20), which for certain cases can be comparable with the Bloch term $-\gamma(T/T_C)^{3/2}$ (this is determined by the concentration c and how close the level E_S^0 is to the bottom of the spin-wave band).

Thus we have the following result: at low temperatures the magnetization decreases according to the $(T/T_C)^{3/2}$ law, as in the ideal crystal, but as soon as the average thermal energy kT becomes close to E_S^0, the magnetization begins to fall sharply by virtue of the filling of the virtual levels, and the filling of all single-particle virtual levels of the s-type leads to a decrease in $M/Ng\mu_0$ by the magnitude of the concentration c. However, it is physically obvious that in the presence of very low-lying virtual levels, when kT exceeds E_S^0, a situation comes about whereby the spins of the ferromagnetic matrix atoms are in great bulk still weakly excited, and the spins on the impurity atoms and close to them are already deflected.[2)] Hence to describe the behavior of the spins on the impurity atoms and their neighbors by means of the approximate transition from spin operators to Bose operators according to the method of Holstein and Primakoff is already impossible. This phenomenon can be observed even at temperatures at which we still would not have gotten out of the framework of the spin-wave description for the ideal ferromagnet. Strong excitation of the spins on the impurities produces a further decrease in the magnetization of the crystal, but it is not possible to take this into account correctly by means of the density of single-particle states; what is required is a self-consistent solution of the system of equations for the magnetization of individual sites, which can be obtained with the aid of the Green temperature functions made up of spin operators.

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[1)] A similar effect takes place for the heat capacity of a crystal containing a heavy impurity atom (see[5]).

[2)] We recall (Fig. 4) that the virtual levels of the s-type appear at the bottom of the band when J′/J < 1; the smaller this ratio, the closer they are to the bottom. This means that the effect considered appears when the exchange coupling of the impurity with the matrix is markedly weaker than between the atoms of the matrix.

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