Dynamics of impurity spin in a dielectric

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The susceptibility of an impurity spin interacting with nuclear spins of a dielectric is determined under the condition that the radius of this interaction is large enough. Expressions are presented for the static adiabatic susceptibility and for the isothermal susceptibility, which are different in this case. The impurity-spin paramagnetic-resonance line shape is investigated. It is shown that the line is neither Gaussian nor Lorentzian, but has an exponential shape. The results have been obtained under temperatures much higher than the interaction of the impurity spin with the nuclear spins.

1. INTRODUCTION

At the present time there is no consistent microscopic theory of paramagnetic-resonance line shape, and it is customary to use phenomenological or semiphenomenological approximations, as well as a number of other methods, the most frequently used among which, in the high-temperature region, is the method of moments (see, e.g., the book by Abragam[1]). In the method of moments the line shape is estimated, as a rule, from the ratio of the fourth moment to the square of the second moment. Thus, the line is taken to be Gaussian if this ratio is close to three and Lorentzian if much larger than three. This raises the question whether it is possible to calculate the line shape accurately even if the model employed is simplified, and to compare the result with the line shape as stimated by the method of moments. The present paper is devoted to this question.

We consider the following model. An impurity paramagnetic atom is situated in a dielectric lattice, and the spin relaxation of the atom is due to the interaction with the spins of the surrounding nuclei. The radius of this interaction is large, so that the atom interacts with a large number $z \gg 1$ of nuclear spins, which do not interact in turn with one another. Finally, since the magnetic moments μ_N of the nuclei are small, we neglect their interaction with the external magnetic field. This means in fact that $T \gg \mu_N H$. Thus the Hamiltonian of the system takes the form

$$H=-\sum_l V_l \mathbf{I}_l \mathbf{S} - g\mu_0 \mathbf{H}\mathbf{S}, \tag{1}$$

where S is the spin of the atom (with $S = 1/2$), I_l are the spins of the nuclei, V_l is the potential of the interaction with the l-th nucleus, g is the g-factor of the atom, and μ_0 is the Bohr magneton. Within the framework of this model, using the spin-temperature diagram technique (see the papers of Abrikosov[2] and of Vaks, Larkin, and Pikin[3]), we can sum the principal terms of the perturbation-theory series and obtain a formula for the dynamic susceptibility. It turns out here that the line has an exponential shape, whereas an estimate by the method of moments would lead to the conclusion that its shape is Gaussian.

Before we proceed directly to this solution of the problem, let us discuss briefly the question of the possible realization of our model. If we assume that the interaction is due to the magnetic dipole forces, then, as follows from the formula given below, our parameter turns out to be of the order of the number of the nearest neighbors. Thus, this approach makes it possible to estimate the influence exerted on the line shape by the dipole-dipole interaction. Another possible interaction mechanism is exchange of virtual electron-hole pairs (see[4]). In this case, if the forbidden band is narrow enough, the radius of this interaction is large, and if the exchange parameters are of the appropriate value, we obtain a situation corresponding to the considered model. We shall not discuss this question in greater detail, since, in our opinion, this problem still has mainly the character of a theoretical model.

2. IMPURITY-SPIN SUSCEPTIBILITY

To determine the susceptibility we shall use a temperature diagram technique based on representation of the spin operators in the form[2,3]

$$\hat{S}^i = a_\beta^+ S^i_{\beta\beta'} a_{\beta'}, \qquad \hat{I}_l^i = b_{l\alpha}^+ I^i_{l\alpha\alpha'} b_{l\alpha'}, \tag{2}$$

where a_β and $b_{l\alpha}$ are Fermi operators, and $S^i_{\beta\beta'}$ and $I^i_{l\alpha\alpha'}$ are the spin matrices of the impurity and of the nucleus.

We determine the temperature Green's function of the impurity spin

$$G_{ij}(\omega_n) = \int_0^{1/T} e^{i\omega_n\tau} \langle \hat{S}_i(\tau)\hat{S}_j(0)\rangle d\tau, \tag{3}$$

where $\omega_n = 2n\pi T$ and $\langle \ldots \rangle$ is the Gibbs averaging with the Hamiltonian H.

In the limit of low impurity concentration N_0, the magnetic susceptibility tensor χ_{ij} is connected with the retarded Green's function G^R_{ij}, which is an analytic continuation of (3), by the relation

$$\chi_{ij}(\omega) = N_0 (g\mu_0)^2 G_{ij}^R(\omega). \tag{4}$$

The temperature Green's functions of the noninteracting impurity and nucleus, which are needed for the calculations, are given by

$$g^{(0)}_{\beta\beta'}(\omega_n, \mathbf{h}) = \left(\frac{1}{i\omega_n - \mathbf{S}\mathbf{h} - \lambda}\right)_{\beta\beta'}, \tag{5}$$

$$f^{(0)}_{l\alpha\alpha'}(\omega_n) = \delta_{\alpha\alpha'}/(i\omega_n - \Lambda), \tag{6}$$

where $\omega_n = (2n+1)\pi T$ and $\mathbf{h} = -g\mu_0\mathbf{H}$. In the calculation of the contributions of the diagrams it is necessary, after summing over the frequencies, to let λ and Λ go to infinity.

We proceed to analyze the diagrams for G_{ij}. Some of them are shown in Fig. 1, where the solid line corresponds to the Green's function $g^{(0)}$ of the impurity, the dashed line to the Green's function $f_l^{(0)}$ of the nucleus, and the wavy line to the interaction V_l. Since there is only one impurity, each diagram contains only one impurity loop. The vertices of this loop are set in correspondence with the impurity spin operators $\hat{S}^k$, and the vertices of the nuclear loops are set in correspondence with the spin operators $\hat{I}_l^k$ of the nuclei. The Gibbs aver-

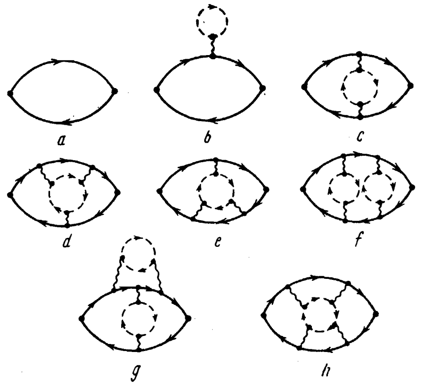

FIG. 1

age in expression (3) should be suitably normalized[2]. Accordingly, for each interacting nuclear spin one introduces in the diagram a normalization factor $e^{\Lambda/T}/(2I + 1)$. An impurity-spin normalization factor pertaining to the entire diagram expansion will be determined in the next section. It is the product of two parts, one of which is independent of the field and of the interaction with the nuclei. This part can be taken into account directly by introducing the factor $e^{\lambda/T}/(2S + 1)$. We note also that diagrams of the type shown in Fig. 1c, containing a nuclear loop with one vertex, are equal to zero.

We shall show now that at $z \gg 1$ in the temperature region $T \gg Vz^{1/3}$, where V is the characteristic energy of the interaction of the impurity spin with the nuclear spin, the principal role is played by diagrams obtained from diagram a of Fig. 1 by all possible insertions of the simplest two-vertex nuclear loops. Indeed, the diagram h of Fig. 1 and the other diagrams with the nuclear four-point diagram are smaller by a factor of z than the diagrams f and g of Fig. 1, which are taken into account in the main sequence, and can therefore be neglected. It is easy to see that this reasoning also holds true for more complicated diagrams. Further, diagrams d and e of Fig. 1 are of the order of zV^3/T^4, and are small in comparison with diagram a of Fig. 1 in the indicated temperature region[1)]. Therefore the series obtained from them by means of all possible insertions of two-vertex nuclear loops should be small in comparison with the series generated by diagram a of Fig. 1. In fact, however the region of applicability of the results obtained in this manner turns out to be narrower. Namely, inasmuch as the diagrams taken into account by us contain in each succeeding order an additional factor zV^2/T^2, it follows that the region in which the summation of these diagrams can be carried out is $T \gtrsim V\sqrt{z}$. Thus, in the indicated region the susceptibility is determined by the series shown in Fig. 2. We note also the following. At finite frequencies, the total aggregate of the diagrams yields for the susceptiblity a series in the moments $\langle\omega^{2n}\rangle/\omega^{2n}$, where the numerator contains a moment of order 2n. Our summation procedure is equivalent to reconstructing the susceptibility from its moments, each of which is calculated approximately accurate to terms of order zV^3/T^3 and $1/z$ relative to the principal terms.

Let us proceed to sum the series shown in Fig. 2. Since the two-vertex nuclear loop is proportional to a δ function in the frequency[3], its insertion in the impurity lines does not change the frequencies that correspond to these lines. It is this circumstance which enables us to sum the series. Indeed, single differentiation

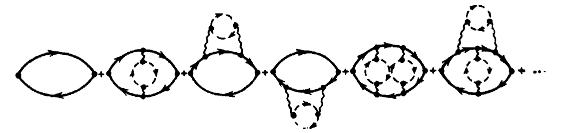

FIG. 2

of the Green's function of the impurity $g^0(\omega_n, \mathbf{h})$ with respect to the field is equivalent to the appearance of a vertex in the corresponding line of the diagram. This leads to the following relation between the terms of the series:

$$\frac{A}{2}\left(\frac{\partial}{\partial \mathbf{h}}\right)^2 \tilde{G}_{ij}^{(p)}(\omega_n, \mathbf{h}) = \tilde{G}_{ij}^{(p+1)}(\omega_n, \mathbf{h}), \tag{7}$$

where $A = I(I+1)\Sigma V_l^2/3$ is a factor from the two-vertex nuclear loop and $\tilde{G}_i^{(p)}$ is a sum of diagrams of order p. The tilde indicates that we are considering diagrams without taking into account the general normalization, which depends on the field and on the interaction with the nuclei. Taking (7) into account, we arrive at the following equation for the entire series shown in Fig. 2:

$$\frac{A}{2}\left(\frac{\partial}{\partial \mathbf{h}}\right)^2 \tilde{G}_{ij}(\omega_n, \mathbf{h}) - \tilde{G}_{ij}(\omega_n, \mathbf{h}) = -\tilde{G}_{ij}^{(0)}(\omega_n, \mathbf{h}). \tag{8}$$

Thus, as a result of summing the series we have obtained a second-order differential equation with a free term $\tilde{G}_{ij}^{(0)}$ determined by the zero-order diagram (a of Fig. 1).

The function $\tilde{G}_{ij}(\omega_n, \mathbf{h})$ is a second-rank tensor, and can therefore be expressed in the form

$$\tilde{G}_{ij}(\omega_n, \mathbf{h}) = K(\omega_n, h^2)\delta_{ij} + L(\omega_n, h^2)h_i h_j + P(\omega_n, h^2) i e_{ijk} h_k, \tag{9}$$

and as a result Eq. (8) reduces to a system of the following three equations:

$$\begin{aligned} &uP''(\omega_n, u) + {}^5/_2 P'(\omega_n, u) - P(\omega_n, u) = -P^{(0)}(\omega_n, u),\\ &uL''(\omega_n, u) + {}^7/_2 L'(\omega_n, u) - L(\omega_n, u) = -L^{(0)}(\omega_n, u),\\ &uK''(\omega_n, u) + {}^3/_2 K'(\omega_n, u) - K(\omega_n, u) = -K^{(0)}(\omega_n, u) - AL(\omega_n, u), \end{aligned} \tag{10}$$

where we have introduced the dimensionless variable $u = h^2/2A$, and the functions $P^{(0)}$, $L^{(0)}$, and $K^{(0)}$ are the coefficients in the expansion (9) for $\tilde{G}_{ij}^{(0)}$:

$$P^{(0)}(\omega_n, u) = -\frac{i\omega_n \operatorname{sh}\sqrt{Au/2T^2}}{\sqrt{2Au}(2Au + \omega_n^2)}, \tag{11}$$

$$L^{(0)}(\omega_n, u) = -\sqrt{\frac{2}{Au}}\frac{\operatorname{sh}\sqrt{Au/2T^2}}{2Au + \omega_n^2}, \tag{12}$$

$$K^{(0)}(\omega_n, u) = \sqrt{\frac{Au}{2}}\frac{\operatorname{sh}\sqrt{Au/2T^2}}{2Au + \omega_n^2}. \tag{13}$$

Let us consider the first equation of (10). We express its solution in the form

$$\begin{aligned} P(\omega_n, u) = {} & (\operatorname{ch} 2\sqrt{u})'' \int_0^u (\operatorname{sh} 2\sqrt{v})'' P^{(0)}(\omega_n, v) v\sqrt{v}\, dv \\ & - (\operatorname{sh} 2\sqrt{u})'' \int_0^u (\operatorname{ch} 2\sqrt{v})'' P^{(0)}(\omega_n, v) v\sqrt{v}\, dv \\ & + c_1(\omega_n)(\operatorname{sh} 2\sqrt{u})'' + c_2(\omega_n)(\operatorname{ch} 2\sqrt{u})''. \end{aligned} \tag{14}$$

Here $c_1(\omega_n)$ and $c_2(\omega_n)$ are functions that do not depend on the magnetic field and are chosen by starting from the following two conditions. The first is that the susceptibility be finite in a zero field. It follows from this condition that $c_1(\omega_n) = 0$. The second condition is that $\tilde{G}_{ij}$ is determined in the absence of interaction by diagram a of Fig. 1. This means that as $A \to 0$ we have

$$\lim_{u\to\infty} P(\omega_n, u) = P^{(0)}(\omega_n, h^2), \tag{15}$$

and leads to the following expression for $c_2(\omega_n)$:

$$c_2(\omega_n) = \int_0^\infty (e^{-2\sqrt{v}})'' P^{(0)}(\omega_n, v)\, v\sqrt{v}\, dv. \tag{16}$$

We note that by virtue of (11) this expression diverges at $T \le (A/8)^{1/2}$.

It will be shown later on that the other functions K and L also contain such a singularity. At the present time, the physical nature of this divergence is not clear. It may be connected with a restructuring of the entire system in such a way that the directions of the nuclear spins are correlated in the interaction region with the direction of the impurity spin. We shall assume here that the divergence indicates at least the region of applicability of the obtained representation for the susceptibility, which is determined by the condition $T \gg \sqrt{A/8}$. Finally, since $\sqrt{A} \approx \sqrt{z}V$, the region of applicability of our approximation is $T \gg \sqrt{z/8}V$ and $z \gg 1$. The analytic continuation of the expression for $P(\omega_n, u)$ from discrete frequencies to the real axis is in this case trivial and reduces to the substitution $i\omega_n \to \omega + i\delta$. Thus, we have for $P(\omega, u)$

$$\begin{aligned} P(\omega, u) = &-\frac{\omega}{\sqrt{2A}}(\mathrm{ch}\, 2\sqrt{u})'' \int_0^u \frac{v(\mathrm{sh}\, 2\sqrt{v})''\, \mathrm{sh}\sqrt{Av/2T^2}\, dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} \\ &+\frac{\omega}{\sqrt{2A}}(\mathrm{sh}\, 2\sqrt{u})'' \int_0^u \frac{v(\mathrm{ch}\, 2\sqrt{v})''\, \mathrm{sh}\sqrt{Av/2T^2}\, dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} \\ &-\frac{\omega}{\sqrt{2A}}(\mathrm{ch}\, 2\sqrt{u})'' \int_0^\infty \frac{v(e^{-2\sqrt{v}})''\, \mathrm{sh}\sqrt{Av/2T^2}\, dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)}. \end{aligned} \tag{17}$$

The remaining two equations of the system (10) are solved analogously and the solutions take the form

$$\begin{aligned} L(\omega, u) = &\sqrt{\frac{2}{A}}(\mathrm{ch}\, 2\sqrt{u})''' \int_0^u \frac{v^2(\mathrm{sh}\, 2\sqrt{v})'''\, \mathrm{sh}\sqrt{Av/2T^2}\, dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} \\ &-\sqrt{\frac{2}{A}}(\mathrm{sh}\, 2\sqrt{u})''' \int_0^u \frac{v^2(\mathrm{ch}\, 2\sqrt{v})'''\, \mathrm{sh}\sqrt{Av/2T^2}\, dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} \\ &+\sqrt{\frac{2}{A}}(\mathrm{ch}\, 2\sqrt{u})''' \int_0^\infty \frac{v^2(e^{-2\sqrt{v}})'''\, \mathrm{sh}\sqrt{Av/2T^2}\, dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)}, \end{aligned} \tag{18}$$

$$\begin{aligned} K(\omega, u) = &\sqrt{\frac{A}{2}}(\mathrm{sh}\, 2\sqrt{u})' \int_0^u \sqrt{v}(\mathrm{ch}\, 2\sqrt{v})' \\ &\times\left(\frac{\sqrt{v}\,\mathrm{sh}\sqrt{Av/2T^2}}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} + \sqrt{2A}\, L(\omega, v)\right) dv - \sqrt{\frac{A}{2}}(\mathrm{ch}\, 2\sqrt{u})' \\ &\times\int_0^u \sqrt{v}(\mathrm{sh}\, 2\sqrt{v})' \left(\frac{\sqrt{v}\,\mathrm{sh}\sqrt{Av/2T^2}}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} + \sqrt{2A}\, L(\omega, v)\right) dv \\ &+\sqrt{\frac{A}{2}}(\mathrm{ch}\, 2\sqrt{u})' \int_0^\infty e^{-2\sqrt{v}} \left(\frac{\sqrt{v}\,\mathrm{sh}\sqrt{Av/2T^2}}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)} + \sqrt{2A}\, L(\omega, v)\right) dv. \end{aligned} \tag{19}$$

Expression (9), together with (17)–(19), determines the susceptibility tensor, apart from a normalization that depends on the field and on the interaction with the nuclei. This normalization factor is determined in the next section, and is equal to

$$\begin{aligned} N(u) &= e^{A/8T^2}\left(\mathrm{ch}\sqrt{\frac{Au}{2T^2}} + \sqrt{\frac{A}{8T^2u}}\,\mathrm{sh}\sqrt{\frac{Au}{2T^2}}\right) \\ &= e^{A/8T^2}\left(\mathrm{ch}\frac{h}{T} + \frac{A}{2hT}\,\mathrm{sh}\frac{h}{T}\right). \end{aligned} \tag{20}$$

We thus have ultimately

$$\chi_{ij}(\omega, \mathbf{h}) = \frac{N_0(g\mu_0)^2}{N(h^2)}(K(\omega, h^2)\delta_{ij} + L(\omega, h^2)h_ih_j + P(\omega, h^2)i\varepsilon_{ijk}h_k), \tag{21}$$

where $K(\omega, h^2)$, $L(\omega, h^2)$, $P(\omega, h^2)$ are given by expressions (17)–(19), in which it is necessary to put $u = h^2/2A$.

The line shape is determined by the imaginary part of the susceptibility tensor. We usually consider the response of the system to the transverse magnetic field. If the z axis is taken to be the direction of the constant magnetic field, and the x axis is taken to be the direction of the alternating magnetic field, then the line shape is determined by the imaginary part $\chi''_{xx}(\omega, u)$. Since χ''_{xx} is an odd function of the frequency, it suffices to consider the region $\omega \gg 0$, where

$$\begin{aligned} \chi_{xx}''(\omega, u) = &\frac{\pi N_0(g\mu_0)^2 e^{-2\sqrt{u}}}{N(u)\sqrt{u}}\left(\frac{\omega}{8A}\right)\mathrm{sh}\frac{\omega}{2T}\cdot \\ &\times\left[\left(\frac{1}{\sqrt{u}}+\frac{1}{u}\right)(\mathrm{ch}\, 2\sqrt{v})'''v\sqrt{v} + 2\,\mathrm{sh}\, 2\sqrt{v} - (\mathrm{ch}\, 2\sqrt{v})'''\sqrt{v}\right], \\ &0 \le \omega \le \sqrt{2Au} = \mu_0 gH, \end{aligned} \tag{22}$$

$$\begin{aligned} \chi_{xx}''(\omega, u) = &\frac{\pi N_0(g\mu_0)^2}{N(u)}\frac{\omega}{8A}\mathrm{sh}\frac{\omega}{2T}\cdot \\ &\times e^{-\omega\sqrt{2/A}}\left[\frac{1}{u}\left(\mathrm{ch}\, 2\sqrt{u} - \frac{\mathrm{sh}\, 2\sqrt{u}}{2\sqrt{u}}\right)\cdot\right. \\ &\left.\times e^{2\sqrt{v}}(e^{-2\sqrt{v}})'''v\sqrt{v} + \frac{\mathrm{sh}\, 2\sqrt{u}}{\sqrt{u}}\left(2+\frac{1}{\sqrt{v}}+\frac{1}{2v}\right)\right], \quad \omega \ge \sqrt{2Au} = \mu_0 gH, \end{aligned} \tag{23}$$

where it is necessary to put $v = \omega^2/2A$ after the differentiation. We note that the $\chi''_{xx}(\omega, u)$ are continuous at $\omega = \mu_0 gH$, but $d\chi''_{xx}(\omega, u)/d\omega$ has a discontinuity.

We consider the expression for the line shape in certain limiting cases. Let $h = \mu_0 gH \gg \sqrt{2A}$, i.e., the interaction of the impurity spin with the field is much stronger than the interaction with the nuclei. In the region $\omega \gg \sqrt{2A}$ we have

$$\begin{aligned} f(\omega) &= \frac{\chi_{xx}''(\omega)}{\chi_{xx}''(\omega_0)} \\ &= \frac{\omega}{\omega_0}\frac{\mathrm{sh}(\omega/2T)}{\mathrm{sh}(\omega_0/2T)}\exp\left\{-\sqrt{\frac{2}{A}}|\omega-\omega_0|\right\}, \end{aligned} \tag{24}$$

where $\omega_0 = \mu_0 gH$. It is seen from (24) that the line has a resonant character, with an absorption maximum at the same frequency as in the absence of the interaction. The line is symmetrical near the maximum ω_0. The influence of the interaction has led to the appearance of an absorption band of width $\Delta\omega \approx \sqrt{2A}$. A characteristic feature of the line is the exponential decrease of $f(\omega)$ with frequency. This circumstance takes place not only in the considered limiting case, inasmuch as the arguments of the exponentials in the general expressions (22) and (23) for $\chi''_{xx}(\omega, u)$ contain ω in the first degree. Thus, the line is neither Gaussian nor Lorentzian. The reason is that in our case $S = 1/2$ and in the magnetic field there are only two spin states between which transitions with change of spin projection not exceeding unity can take place as a result of the interaction with the nuclear spins. On the other hand, if $S \gg 1/2$, we shall actually have a classical spin that precesses in a magnetic field acted upon by the random field of the nuclear spins, and the line should then be Gaussian.

The method of moments yields in our case for the ratio of the fourth moment M_4 to the square of the second moment M_2

$$\frac{M_4}{M_2^2}=\frac{\mathrm{Sp}([H'_{int},[H'_{int},\hat{S}_x]]^2)}{\{\mathrm{Sp}([H'_{int},\hat{S}_x]^2)\}^2}=3+\frac{\sum V_l^4}{\left(\sum V_l^2\right)^2}\left(\frac{9\,\mathrm{Sp}\,I_z^4}{2I+1}-1\right)$$

$$\approx 3+\frac{1}{z}\left(\frac{9\mathrm{Sp}\,I_z^4}{2I+1}-1\right). \qquad (25)$$

Here $H'_{int}=\hat{S}_z\sum V_l\hat{I}_{lz}$ is the so-called secular part of the interaction Hamiltonian relative to $H_0=-g\mu_0\hat{S}_zH_z$. In the derivation of (25) we have put $V_l\approx V$. In our approximation ($z\gg 1$) this ratio is equal to 3, which coincides with the result for the Gaussian line.

Assume now that the interaction of the impurity spin with the nuclei is of the same order as the interaction with the field. In this case the general character of the line is preserved. The maximum, with the exception of a very weak temperature shift, will occur at the frequency ω_0. The shape of the right-hand line wing at $\omega\gg\sqrt{2A}$, apart from a numerical coefficient on the order of unity, will be determined by (24). The line shape will change somewhat only near the maximum ω_0.

We now proceed to the susceptibility in a zero field. Substituting $L(\omega, u)$ from (18) in (19) and taking the normalization (20) into account we obtain at $u = 0$

$$\chi_{ij}(\omega)=\frac{2\sqrt{2}}{3}\frac{N_0(g\mu_0)^2e^{-A/8T^2}\sqrt{A}}{1+A/2T^2}$$

$$\times\int_0^\infty\frac{\sqrt{v}\,e^{-2\sqrt{v}}\,\mathrm{sh}\sqrt{Av/2T^2}\,dv}{(\sqrt{2Av}-\omega-i\delta)(\sqrt{2Av}+\omega+i\delta)}\delta_{ij}. \qquad (26)$$

The maximum value of $\chi''_{ii}(\omega)$ is reached at the frequency

$$\omega_0=\sqrt{2A}\,(1+A/12T^2). \qquad (27)$$

For the line shape we obtain

$$f(\omega)=\frac{\omega}{\omega_0}\frac{\mathrm{sh}(\omega/2T)}{\mathrm{sh}(\omega_0/2T)}\exp\left\{-\sqrt{\frac{2}{A}}(\omega-\omega_0)\right\}. \qquad (28)$$

Let us discuss the static limit of the susceptibility. As is well known (see the paper by Kubo[5]), either isothermal or adiabatic static susceptibility can be considered. The isothermal susceptibility describes the reaction of a system in thermal contact with a thermostat to the external field, while the adiabatic susceptibility describes the reaction of an isolated system to which an external field is applied adiabatically. Generally speaking, the two susceptibilities are not equal. In our case the expression (21) for $\chi_{ij}(\omega, h)$ at $\omega = 0$ yields the adiabatic susceptibility. Indeed, in the determination of χ_{ij} we summed diagrams at nonzero external discrete frequencies and continued the resultant expression analytically to the real axis. We have thus obtained a susceptibility that yields at $\omega\neq 0$ the reaction of the system to an adiabatically turned-on alternating field, so that the continuation of the solution to $\omega = 0$ yields the adiabatic susceptibility. It follows therefore that when the interaction with the nuclear spins is turned off we should obtain a zero value for the parallel part of the static susceptibility, since an infinitesimally slow turning on of the stationary field cannot lead to a transition between states of the system. It is easy to show that this does indeed take place in our case. To this end it suffices to substitute the functions (11), (12) and (13) with $\omega_n = 0$ into the expression obtained from (21) for the parallel part of the susceptibility.

On the other hand, to find the isothermal susceptibility it is necessary, in view of its definition, to set the external frequency equal to zero from the very outset, and then to sum over the internal frequencies. In the expression for the zeroth-order diagram this changes only the function $L^{(0)}$:

$$L^{(0)}(0,u)\to L^{(0)T}(0,u)=L^{(0)}(0,u)+\frac{\mathrm{ch}\sqrt{Au/2T^2}}{ATu} \qquad (29)$$

(the superscript T labels isothermal quantities from now on). Consequently, the function $P^T(0, u)$ is given by expression (17), the function $L^T(0, u)$ by expression (18) with $L^{(0)}$ replaced by $L^{(0)T}$, and the function $K^T(0,u)$ is determined by (19) with L replaced by L^T. Thus, the isothermal susceptibility χ_{ij}^T is determined by expression (21), which contains in the right-hand side the isothermal functions P^T, L^T, and K^T, which have been defined above.

We present, in particular, expressions for the static susceptibilities in a zero field.

$$\chi_{ii}^T(0)=\chi_{ii}(0)+\frac{N_0(g\mu_0)^2T\exp\{-(T_c/T)^2\}}{12(T^2-T_c^2)}$$

$$=\frac{N_0(g\mu_0)^2T\exp\{-(T_c/T)^2\}}{12(T^2-T_c^2)}\left\{1+\frac{2T^2}{T^2+T_c^2}\right\}. \qquad (30)$$

Here $T_c=\sqrt{A/8}$ and $\chi_{ii}(0)$ is the adiabatic susceptibility determined from (26) at $\omega = 0$:

$$\chi_{ii}(0)=\frac{N_0(g\mu_0)^2T^3\exp\{-(T_c/T)^2\}}{6(T^2-T_c^2)(T^2+T_c^2)}. \qquad (31)$$

After turning off the interaction with the nuclear spins, we should obtain the susceptibility of an isolated spin. It is easy to see that expression (30) at $T_c = 0$ does indeed yield this value for the isothermal susceptibility. We note also that the divergence of the susceptibility as $T\to T_c$ is the same as in the Curie-Weiss law. The physical cause of this divergence was already discussed earlier.

3. NORMALIZATION AND GREEN'S FUNCTION OF IMPURITY

A general expression for the normalization of the mean values in spin diagram technique in the presence of a magnetic field is given in[2]. In our case, we have for the part of the normalization not taken into account in the calculation of the diagram contribution

$$N(u)=\frac{e^{\lambda/T}}{2S+1}\langle a_\beta^+a_\beta\rangle=\frac{e^{\lambda/T}}{2S+1}T\sum_{\omega_n}g_{\alpha\alpha}(\omega_n,u)e^{i\omega_n\tau},\qquad \tau\to+0. \qquad (32)$$

We have used here the well-known expression $\langle a_\beta^+a_\beta\rangle$ in terms of the Green's function (see[6]). As usual, on going from summation to integration along the corresponding contour, we obtain

$$N(u)=-\frac{2}{\pi}\int_0^\infty \mathrm{Im}\,g_{\alpha\alpha}^R(t,u)\,\mathrm{ch}\frac{t}{T}\,dt, \qquad (33)$$

where g^R is the retarded Green's function.

To determine the Green's function of the impurity, let us consider its self-energy part Σ. A diagram analysis similar to that in the preceding section shows that in the main the diagrams for Σ at $z\gg 1$ in the region $T\gg z^{1/3}V$ will be the diagrams shown in Fig. 3. The heavy lines correspond here to the impurity Green's function g. Single differentiation of the Green's function leads to the appearance of a vertex in the corresponding line. We therefore arrive at the following differential equation for the self-energy part:

$$\Sigma_{\alpha\beta}(t,\mathbf{h})=A\mathbf{S}_{\alpha\gamma}g_{\gamma\delta}(t,\mathbf{h})(\mathbf{S}_{\delta\beta}+\partial\Sigma_{\delta\beta}(t,\mathbf{h})/\partial\mathbf{h}), \qquad (34)$$

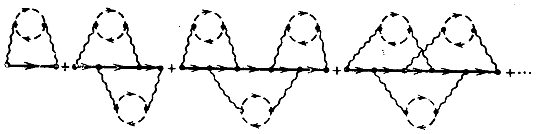

FIG. 3

where we have introduced a new variable $t = i\omega_n - \lambda$. Therefore, using Dyson's equation, we obtain an equation for the Green's function of the impurity

$$AS_{\alpha\delta}\frac{\partial g_{\delta\beta}(t,\mathbf{h})}{\partial \mathbf{h}} - g_{\alpha\delta}^{(0)-}(t,\mathbf{h})\, g_{\delta\beta}(t,\mathbf{h}) + \delta_{\alpha\beta} = 0. \qquad (35)$$

Since the tensor $g_{\alpha\beta}$ can be represented in the form

$$g_{\alpha\beta}(t,\mathbf{h}) = g_1(t,h^2)\,\delta_{\alpha\beta} + (\mathbf{Sh})_{\alpha\beta}\, g_2(t,h^2), \qquad (36)$$

we arrive at a system of equations for g_1 and g_2:

$$g_1'(t,u) + g_1(t,u) - t g_2(t,u) = 0, \quad {}^1/_2 A g_2'(t,u) + {}^1/_2 A (u + {}^3/_2)\, g_2(t,u) - t g_1(t,u) - 1 = 0. \qquad (37)$$

Substituting g_2 from the first equation of the system in the second, we obtain

$$u(e^u g_1(t,u))'' + \frac{3}{2}(e^u g_1(t,u))' - \frac{2t^2}{A} e^u g_1(t,u) = -\frac{2t}{A} e^u. \qquad (38)$$

The requirement that $g_1(t, u)$ be finite at $u = 0$ enables us to write the solution in the form

$$g_1(t,u) = -e^{-u}\sqrt{\frac{2}{Au}}\,\mathrm{sh}\sqrt{\frac{8t^2u}{A}}\int_0^u dv\, e^v\,\mathrm{ch}\sqrt{\frac{8t^2v}{A}} + e^{-u}\sqrt{\frac{2}{Au}}\,\mathrm{ch}\sqrt{\frac{8t^2u}{A}}\int_0^u dv\, e^v\,\mathrm{sh}\sqrt{\frac{8t^2v}{A}} - c(t)\, e^{-u}\sqrt{\frac{2}{Au}}\,\mathrm{sh}\sqrt{\frac{8t^2u}{A}}. \qquad (39)$$

The function $c(t)$ is chosen here to satisfy the condition that the Green's function go over in the absence of interaction into the free function $g^{(0)}$. This leads to the following expression for $c(t)$:

$$c(t) = \int_{-\infty}^{0}\exp\left[v + \sqrt{\frac{8t^2v}{A}}\right] dv = e^{-t^2/A} D_{-2}\left(-2i\sqrt{\frac{t^2}{A}}\right), \qquad (40)$$

where D_{-2} is a parabolic-cylinder function.

We need the retarded Green's function. Choosing in (40) that branch of the root which ensures analyticity of $c(t)$ in the upper half-plane, we get

$$g_1^R(t,u) = -e^{-u} c^R(t)\sqrt{\frac{2}{Au}}\,\mathrm{sh}\sqrt{\frac{8t^2u}{A}} - e^{-u}\sqrt{\frac{2}{Au}}\left(\mathrm{sh}\sqrt{\frac{8t^2u}{A}}\int_0^u dv\, e^v\,\mathrm{ch}\sqrt{\frac{8t^2v}{A}} - \mathrm{ch}\sqrt{\frac{8t^2u}{A}}\int_0^u dv\, e^v\,\mathrm{sh}\sqrt{\frac{8t^2v}{A}}\right), \qquad (41)$$

$$c^R(t) = e^{-t^2/A} D_{-2}(2it/\sqrt{A}). \qquad (42)$$

For the function $g_2^R(t, u)$ we obtain

$$g_2^R(t,u) = \frac{1}{t}\left(g_1^R(t,u) + g_2^{R'}(t,u)\right). \qquad (43)$$

Expressions (36) and (41)–(43) determine the retarded Green's function of the impurity. The quantity $\mathrm{Im}\, g_1^R(t,u)$ needed for the normalization and determined from (41) is given at real t by

$$\mathrm{Im}\, g_1^R(t,u) = -2\sqrt{\frac{\pi}{A^2u}}\, e^{-u-2t^2/A}\,\mathrm{sh}\sqrt{\frac{8t^2u}{A}}. \qquad (44)$$

Substituting this expression in (33), we obtain on normalization

$$N(u) = e^{A/8T^2}\left(\mathrm{ch}\sqrt{\frac{Au}{2T^2}} + \sqrt{\frac{A}{8T^2u}}\,\mathrm{sh}\sqrt{\frac{Au}{2T^2}}\right) = e^{A/8T^2}\left(\mathrm{ch}\frac{h}{T} + \frac{A}{2hT}\,\mathrm{sh}\frac{h}{T}\right). \qquad (45)$$

We note in conclusion that at $A = 0$ we obtain from (45) the usual normalization factor for the mean values in a magnetic field.

[1)] All other third-order diagrams vanish.

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