CONTRIBUTION TO THE THEORY OF THE TRANSVERSE COMPLEX PARAMAGNETIC SUSCEPTIBILITY

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The perturbation theory[5] developed for two-time temperature Green's functions is used to calculate the paramagnetic relaxation function and complex transverse magnetic susceptibility. Use of the Dyson-Dembinski[6,7] representation for spin operators is found to be essential in this case. Near resonance, the expression obtained for the susceptibility in not very small stationary fields is the same as given by the phenomenological theory.[9]

1. WE consider a system described by a Hamiltonian $\mathscr{H}$ if isolated or under a time-independent action. Application of an alternating external action F(t), represented in the Hamiltonian by a term $\hat{\mathscr{H}}'(t) = -\hat{A}F(t)$, causes the mean value B(t) of a quantity B pertaining to the system to deviate from its equilibrium value B_0. In an approximation linear in F(t), this deviation is given by the formula[1]

$$\Delta B(t) \equiv B(t) - B_0 = \int_{-\infty}^{+\infty} dt'\, G_{BA}(t-t')F(t'), \quad (1)$$

where $G_{BA}(t-t')$ is a retarded two-time Green's function defined in the following manner:

$$G_{BA}(t-t') = i\theta(t-t')\langle[\hat{B}(t), \hat{A}(t)]\rangle, \quad (2)$$

$$\hat{B}(t) = \exp\{i\hat{\mathscr{H}}t\}B\exp\{-i\hat{\mathscr{H}}t\}, \quad (3)$$

$$\theta(t) = \begin{cases} 1 & t > 0 \\ 0 & t < 0 \end{cases}; \quad (4)$$

$\langle\ \rangle$ denotes averaging over the Gibbs statistical operator with Hamiltonian $\hat{\mathscr{H}}$.

It follows from (1) that the susceptibility $\chi_{BA}(\omega)$ defined by the relation

$$\Delta B(\omega) = \chi_{BA}(\omega)F(\omega), \quad (5)$$

where $\Delta B(\omega)$ and $F(\omega)$ are the Fourier components of the quantities $\Delta B(t)$ and F(t), coincides with the Fourier component of the Green's function:

$$\chi_{BA}(\omega) = G_{BA}(\omega) = \int_{-\infty}^{+\infty} dt e^{i\omega t} G_{BA}(t). \quad (6)$$

At high temperatures it is sufficient to retain only the linear term in the expansion of $G_{BA}(t-t')$ in powers $\beta\hat{\mathscr{H}}$, where $\beta = 1/kT$. In this approximation we have

$$G_{BA}(t-t') = i\beta\frac{d}{dt}g_{BA}(t-t') + \delta(t-t')\chi_{BA}(0); \quad (7)$$

we use here the notation

$$g_{BA}(t-t') \equiv i\theta(t-t')\langle\hat{B}(t)\hat{A}(t')\rangle_0,$$
$$\chi_{BA}(0) \equiv \beta\langle\hat{B}\hat{A}\rangle_0, \quad \langle\ldots\rangle_0 \equiv \mathrm{Sp}(\ldots)/\mathrm{Sp}(1). \quad (8)$$

The susceptibility $\chi_{BA}(\omega)$ is connected with the Fourier component $g_{BA}(\omega)$ of the function $g_{BA}(t-t')$ by the relation

$$\chi_{BA}(\omega) = \beta\omega g_{BA}(\omega) + \chi_{BA}(0). \quad (9)$$

It can be shown[2] that the relaxation of the quantity $\Delta B(t)$, that is, its change in the case when

$$F(t) = \theta(-t)e^{\varepsilon t}F, \quad \varepsilon \to +0$$

can be expressed in terms of the susceptibility in the following fashion:

$$\Delta B(t) = -\frac{i}{2\pi}F\int_{-\infty}^{+\infty} d\omega\frac{e^{-i\omega t}}{\omega}[\chi_{BA}(\omega) - \chi^*_{AB}(\omega)]. \quad (10)$$

2. We now consider a solid dielectric paramagnet with pure spin magnetism under conditions when the spin system can be regarded as isolated from the lattice (see, for example, [3]). The system referred to in Sec. 1 is an aggregate of N spins localized at the crystal lattice points and coupled with one another by dipole-dipole interaction. The system is in a magnetic field consisting of a constant field H parallel to the z axis, and an alternating field h(t) parallel to the x axis. The Hamiltonian describing the "natural" motion of the system is written in the form

$$\hat{\mathscr{H}} = -\omega_L\sum_j \hat{S}_j^0 + \frac{1}{2}\sum_{j,l} A_{jl}(\hat{S}_j^0\hat{S}_l^0 - \hat{S}_j^+\hat{S}_l^-). \quad (11)$$

Here

$$\hat{S}_j^{\pm} = 2^{-1/2}(\hat{S}_j^x \pm i\hat{S}_j^y), \qquad \hat{S}_j^0 = \hat{S}_j^z,$$
$$A_{jj} = 0, \qquad A_{jl} \equiv \gamma^2 r_{jl}^{-3}(1 - 3\cos^2\theta_{jl}),$$

$\hat{S}_j$ are the spin operators, r_{jl} is the distance between the j-th and l-th spins, θ_{jl} is the angle between the z axis and a line joining the j-th and l-th spins, $\omega_L \equiv \gamma H$, and γ is the gyromagnetic ratio. As usual,[4] we take into account only the secular part of the dipole-dipole interaction. The alternating external action is written in the form:

$$\mathcal{H}'(t) = -\gamma \sum_j \hat{S}_j^x h(t). \tag{12}$$

Our problem is to find the component $\chi_{xx}(\omega)$ of the magnetic susceptibility tensor in the case of high temperatures. Putting in (9)

$$\hat{B} = \hat{A} = \gamma \sum_j \hat{S}_j^x = \frac{\gamma}{\sqrt{2}} \sum_j (\hat{S}_j^- + \hat{S}_j^+),$$

we find that to solve our problem we must calculate the Fourier component of the function

$$g_{xx}(t) = \frac{\gamma^2}{2} \sum_{j,l} \{g_{-+}(\mathbf{r}_j - \mathbf{r}_l;\, t) - \text{c.c.}\},$$
$$g_{-+}(\mathbf{r}_j - \mathbf{r}_l;\, t) = i\theta(t)\langle \hat{S}_j^-(t)\hat{S}_l^+\rangle_0. \tag{13}$$

3. We go over to the Dyson-Dembinski representation[5, 6] for the spin operators, that is, we make everywhere the substitution

$$\hat{S}_j^+ \to \sqrt{s}\, a_j^+(1 - a_j^+ a_j/2s), \qquad \hat{S}_j^- \to \sqrt{s}\, a_j, \tag{14}$$
$$\hat{S}_j^0 \to -s + a_j^+ a_j;$$

here the operators a_j and a_j^+ obey the commutation relations

$$[a_j, a_l^+] = \delta_{jl}, \qquad [a_j, a_l] = [a_j^+, a_l^+] = 0. \tag{15}$$

The operation of taking the trace in $g_+(\mathbf{r}_j - \mathbf{r}_l;\, t)$ must in this case be modified as follows:

$$\mathrm{Sp}\ldots \to \sum_u E_u(u|\ldots|u),\; |u) = \prod_j |u_j),\; E_u = \prod_j E_{u_j}, \tag{16}$$

where $|u_j)$ is a complete orthonormal system of eigenstates of the operator $a_j^+ a_j$ and

$$E_{u_j} = \begin{cases} 1 & 0 \leqslant u_j \leqslant 2s \\ 0 & u_j > 2s \end{cases}, \tag{17}$$

where s is the spin. It is convenient, further, to expand the operators $a_\mathbf{l}$ (here $\mathbf{l}$ is used for brevity in lieu of $\mathbf{r}_l$) in a Fourier series

$$a_\mathbf{l} = \frac{1}{\sqrt{N}} \sum_\mathbf{q} e^{i\mathbf{lq}} a_\mathbf{q}, \qquad a_\mathbf{q} = \frac{1}{\sqrt{N}} \sum_\mathbf{l} e^{-i\mathbf{ql}} a_\mathbf{l}. \tag{18}$$

In the terms of the operator $a_\mathbf{q}$, the Hamiltonian $\mathcal{H}$ and the function $g_+(\mathbf{r}_j - \mathbf{r}_l;\, t)$ are represented in the form

$$\mathcal{H} = -\sum_\mathbf{q} \omega_\mathbf{q} a_\mathbf{q}^+ a_\mathbf{q} + \frac{1}{4N} \sum_{\mathbf{q}_1\mathbf{q}_2\mathbf{q}_3\mathbf{q}_4} \Gamma(\mathbf{q}_1\mathbf{q}_2|\mathbf{q}_3\mathbf{q}_4)$$
$$\times \Delta(\mathbf{q}_1 + \mathbf{q}_2 - \mathbf{q}_3 - \mathbf{q}_4) a_{\mathbf{q}_1}^+ a_{\mathbf{q}_2}^+ a_{\mathbf{q}_3} a_{\mathbf{q}_4}, \tag{19}$$

where

$$\omega_\mathbf{q} \equiv \omega_L + s(A_0 + {}^1/_2 A_\mathbf{q}), \tag{20}$$
$$\Gamma(\mathbf{q}_1\mathbf{q}_2|\mathbf{q}_3\mathbf{q}_4)$$
$$= {}^1/_2[A_{\mathbf{q}_1-\mathbf{q}_3} + A_{\mathbf{q}_1-\mathbf{q}_4} + A_{\mathbf{q}_2-\mathbf{q}_3} + A_{\mathbf{q}_2-\mathbf{q}_4} + A_{\mathbf{q}_3} + A_{\mathbf{q}_4}), \tag{21}$$
$$A_\mathbf{q} = \sum_\mathbf{f} e^{-i\mathbf{qf}} A_\mathbf{f}, \qquad A_\mathbf{f} = \frac{1}{N} \sum_\mathbf{q} e^{i\mathbf{fq}} A_\mathbf{q}, \tag{22}$$

f stands for $\mathbf{r}_j - \mathbf{r}_l$ (see (11))

$$\Delta(\mathbf{q}) = \begin{cases} 1 & \mathbf{q} = 0 \\ 0 & \mathbf{q} \neq 0 \end{cases}; \tag{23}$$

$$g_{-+}(\mathbf{r}_j - \mathbf{r}_l;\, t) = \frac{s}{N} \sum_\mathbf{q} e^{i(\mathbf{j}-\mathbf{l})\mathbf{q}} \langle a_\mathbf{q}|_t, \tag{24}$$

with

$$\langle a_\mathbf{q}|_t = i\theta(t)\langle a_\mathbf{q}(t)\alpha_\mathbf{q}^+\rangle_b, \quad \alpha_\mathbf{q}^+ = \frac{1}{\sqrt{N}} \sum_\mathbf{l} e^{i\mathbf{ql}} a_\mathbf{l}^+ \left(1 - \frac{a_\mathbf{l}^+ a_\mathbf{l}}{2s}\right), \tag{25}$$
$$\langle\ldots\rangle_b = \sum_u E_u(u|\ldots|u) \Big/ \sum_u E_u(u|u). \tag{26}$$

4. The calculation of the Fourier component $\langle a_\mathbf{q}|_\omega$ of the function $\langle a_\mathbf{q}|_t$ will be carried out in accordance with the well known scheme.[7] The system of coupled equations for $\langle a_\mathbf{q}|_\omega$ has in our case the form

$$(\omega + \omega_\mathbf{q})\langle a_\mathbf{q}|_\omega = -\langle a_\mathbf{q}\alpha_\mathbf{q}^+\rangle_b$$
$$+ \frac{1}{2N} \sum_{\mathbf{q}_1\mathbf{q}_2} \Gamma(\mathbf{q}\mathbf{q}_1|\mathbf{q}_2\mathbf{q}_1 - \mathbf{q}_2 + \mathbf{q})\langle a_{\mathbf{q}_1}^+ a_{\mathbf{q}_2} a_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}|_\omega, \tag{27}$$

$$(\omega - \omega_{\mathbf{q}_1} + \omega_{\mathbf{q}_2} + \omega_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2})\langle a_{\mathbf{q}_1}^+ a_{\mathbf{q}_2} a_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}|_\omega$$
$$= -\langle a_{\mathbf{q}_1}^+ a_{\mathbf{q}_2} a_{\mathbf{q}_1+\mathbf{q}-\mathbf{q}_2}\alpha_\mathbf{q}^+\rangle_b - \frac{1}{2N} \sum_{\mathbf{q}_3\mathbf{q}_4} \Gamma(\mathbf{q}_1 + \mathbf{q}_3 - \mathbf{q}_4\mathbf{q}_4|\mathbf{q}_3\mathbf{q}_1)$$
$$\times \langle a_{\mathbf{q}_1+\mathbf{q}_3-\mathbf{q}_4}^+ a_{\mathbf{q}_4}^+ a_{\mathbf{q}_3} a_{\mathbf{q}_2} a_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}|_\omega$$
$$+ \frac{1}{2N} \sum_{\mathbf{q}_3\mathbf{q}_4} \Gamma(\mathbf{q}_2\mathbf{q}_3|\mathbf{q}_4\mathbf{q}_2 + \mathbf{q}_3 - \mathbf{q}_4)$$
$$\times \langle a_{\mathbf{q}_1}^+ a_{\mathbf{q}_3}^+ a_{\mathbf{q}_4}^+ a_{\mathbf{q}_2+\mathbf{q}_3-\mathbf{q}_4} a_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}|_\omega$$
$$+ \frac{1}{2N} \sum_{\mathbf{q}_3\mathbf{q}_4} \Gamma(\mathbf{q} + \mathbf{q}_1 - \mathbf{q}_2\mathbf{q}_3|\mathbf{q}_4\mathbf{q} + \mathbf{q}_1 - \mathbf{q}_2 + \mathbf{q}_3 - \mathbf{q}_4)$$
$$\times \langle a_{\mathbf{q}_1}^+ a_{\mathbf{q}_2} a_{\mathbf{q}_3}^+ a_{\mathbf{q}_4} a_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2+\mathbf{q}_3-\mathbf{q}_4}|_\omega, \tag{28}$$

. . . .

Defining in accordance with [7] the mass operator $M(\mathbf{q};\omega)$ by means of the relation

$$M(\mathbf{q};\omega)\langle a_{\mathbf{q}}|_{\omega}=\frac{1}{2N}\sum_{\mathbf{q}_1\mathbf{q}_2}\Gamma(\mathbf{q}\mathbf{q}_1|\mathbf{q}_2\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2)$$

$$\times\langle a_{\mathbf{q}_1}^{+}a_{\mathbf{q}_2}a_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}|_{\omega}, \tag{29}$$

we represent the function $\langle a_{\mathbf{q}}|_{\omega}$ in the form

$$\langle a_{\mathbf{q}}|_{\omega}=-\frac{\langle a_{\mathbf{q}}a_{\mathbf{q}}^{+}\rangle_b}{\omega+\omega_{\mathbf{q}}-M(\mathbf{q};\omega)}. \tag{30}$$

Near resonance $\omega=-\omega_{\mathbf{q}}$ we can put $M(\mathbf{q};\omega)\cong M(\mathbf{q};-\omega)\equiv M_{\mathbf{q}}$. Calculating $M_{\mathbf{q}}$ by perturbation theory[7] accurate to the second approximation inclusive, we obtain

$$M_{\mathbf{q}}=s\left(A_0+\frac{1}{2}A_{\mathbf{q}}\right)$$

$$+\frac{s(s+1)}{2N^2}\sum_{\mathbf{q}_1\mathbf{q}_2}\frac{\Gamma(\mathbf{q}\mathbf{q}_1|\mathbf{q}_2\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2)\Gamma(\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2\mathbf{q}_2|\mathbf{q}_1\mathbf{q})}{\omega_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}-\omega_{\mathbf{q}}-\omega_{\mathbf{q}_1}+\omega_{\mathbf{q}_2}+i\varepsilon} \tag{31}$$

or, separating the real and imaginary parts

$$M_{\mathbf{q}}=s(A_0+{}^1/_2A_{\mathbf{q}})+d_{\mathbf{q}}-i\tau_{\mathbf{q}}^{-1}, \tag{32}$$

where

$$d_{\mathbf{q}}=\frac{s(s+1)}{2N^2}\,\mathrm{P}$$

$$\times\sum_{\mathbf{q}_1\mathbf{q}_2}\frac{\Gamma(\mathbf{q}\mathbf{q}_1|\mathbf{q}_2\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2)\Gamma(\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2\mathbf{q}_2|\mathbf{q}_1\mathbf{q})}{\omega_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}-\omega_{\mathbf{q}}-\omega_{\mathbf{q}_1}+\omega_{\mathbf{q}_2}}, \tag{33}$$

$$\tau_{\mathbf{q}}^{-1}=\pi\frac{s(s+1)}{2N^2}\sum_{\mathbf{q}_1\mathbf{q}_2}\Gamma(\mathbf{q}\mathbf{q}_1|\mathbf{q}_2\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2)\Gamma(\mathbf{q}+\mathbf{q}_1$$

$$-\mathbf{q}_2\mathbf{q}_2|\mathbf{q}_1\mathbf{q})\,\delta(\omega_{\mathbf{q}+\mathbf{q}_1-\mathbf{q}_2}-\omega_{\mathbf{q}}-\omega_{\mathbf{q}_1}+\omega_{\mathbf{q}_2}). \tag{34}$$

In our case (unlike in [7]) the correlation functions can be calculated exactly. However, since we have in mind a large system, which corresponds to the transition to the limit as $N\to\infty$ when $N/V=\text{const}$ (V is the volume of the system), with the usual transition from summation to integration

$$\frac{1}{V}\sum_{\mathbf{q}}\ldots\to\frac{1}{(2\pi)^3}\int d\mathbf{q}\ldots,$$

the terms which vanish in this transition have not been written out in $M_{\mathbf{q}}$. In other words, the result has the same form as if we were to use Wick's theorem for the mean values.[8]

5. Thus we obtain for the component $\chi_{xx}(\omega)$ of the magnetic susceptibility tensor near resonance (disregarding the resonance shift characterized by the quantity d_0)

$$\chi_{xx}(\omega)=\chi_0-\chi_0\frac{\omega}{2}\left\{\frac{1}{\omega+\omega_L+i\tau^{-1}}+\frac{1}{\omega-\omega_L+i\tau^{-1}}\right\}, \tag{35}$$

where

$$\tau\equiv\tau_0,\quad \chi_0=N\beta\gamma^2 s(s+1)/3, \tag{36}$$

or, separating the real and imaginary parts,

$$\chi_{xx}(\omega)=\chi_{xx}'(\omega)+i\chi_{xx}''(\omega), \tag{37}$$

$$\chi_{xx}'(\omega)=\chi_0\frac{[1+(\omega_L^2-\omega^2)\tau^2](1+\omega_L^2\tau^2)+2\omega^2\tau^2}{[1+(\omega_L^2-\omega^2)\tau^2]^2+4\omega^2\tau^2}, \tag{38}$$

$$\chi_{xx}''(\omega)=\chi_0\frac{[1+(\omega_L^2+\omega^2)\tau^2]\,\omega\tau}{[1+(\omega_L^2-\omega^2)\tau^2]^2+4\omega^2\tau^2}. \tag{39}$$

Formula (10) is an expression for the relaxation of the magnetic moment:

$$\Delta M_x(t)=\chi_0 h\cos\omega_L t e^{-t/\tau}, \tag{40}$$

from which we see that τ is the relaxation time.

Comparison of formulas (38) and (39) with the results of the phenomenological theory[9] shows that for not too small constant fields the corresponding results coincide, if the relaxation time τ_S of the phenomenological theory is identified with a quantity τ which does not depend on the constant field.

6. It is obviously of interest to calculate the relaxation time τ for concrete crystals. To carry out such a calculation it is necessary to put $q=0$ in (34), take the limiting transition from summation to integration, substitute in it the values of Γ and ω from (21) and (20), write out the values for the given crystal, and carry out the integration. We present the result for the case of a primitive cubic lattice in the nearest-neighbor approximation. By choosing as the coordinate axis the symmetry axis of the crystal we obtain for τ

$$\tau=[{}^9/_2J(s+1)\gamma^2/a^3]^{-1}, \tag{41}$$

where a is the lattice constant and J is a number of the order of unity.

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