Enhanced magnetic susceptibility (Atsarkin effect) at low spin temperatures

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A theoretical study is made of the susceptibility of a spin system to a weak low-frequency field under conditions of dynamic cooling of the reservoir of spin-spin interactions (the "enhanced susceptibility"). An expression is obtained for the enhanced susceptibility under conditions of two-temperature quasi-equilibrium at arbitrary temperatures of the Zeeman subsystem and interaction reservoir. In the nonlinear-spin-temperature-effects approximation, expressions are obtained for the frequency and temperature dependence of the enhanced susceptibility for a regular spin distribution (method of moments) and for a random spin distribution at a low concentration (statistical theory). The expressions obtained are valid for arbitrary temperatures of the Zeeman subsystem and high temperatures of the interaction reservoir.

1. INTRODUCTION

The method of enhanced susceptibility, which was proposed by Atsarkin[1] in 1973, has become very useful for studying relaxation processes and dynamics in paramagnetic spin systems. It has turned out that on dynamic cooling of the reservoir of spin-spin interactions by saturation of the wing of the magnetic resonance line, the susceptibility to a weak low-frequency field increases with respect to the static susceptibility χ_0 by several orders of magnitude, approximately by a factor of ω_0/D, where ω_0 is the center frequency of the magnetic resonance and D is the frequency of the local field created by the spin-spin interactions.

In accordance with the existing experimental situation the theory of the enhanced susceptibility effect (reviewed in Ref. 2) was limited to the high-temperature approximation (HTA):$\omega_0/kT \ll 1$ (we have set $\hbar = 1$). However, interest has recently focused on the study of paramagnetic spin systems in the low-temperature region, down to the ordering temperature.[3] In a recent study of the dynamics of magnetically dilute spins systems by the enhanced-susceptibility method[4] the conditions corresponded to $\omega_0/kT \approx 1$, and so extending the theory of the enhanced-susceptibility effect to the low-temperature case is a timely problem.

The difficulties in constructing a low-temperature theory of the enhanced susceptibility are aggravated by the fact that the Provotorov theory,[5,6] which is used in the theory of the enhanced susceptibility to describe the dynamic cooling of the spin-spin interaction reservoir, is itself a high-temperature theory. The extension of this theory to the low-temperature case[76–9] makes it possible to obtain reliable results only at rather high temperatures of the interaction reservoir, much higher than those which would correspond to the possible spontaneous ordering in a zero effective field. In the present paper we shall also restrict discussion to the case of high temperatures of the interaction reservoir. This case can be realized at fairly high polarizations upon total saturation on the remote wing of the line or upon partial saturation over the entire line. In the latter case the spin system in a rotating coordinate system is found in a state of quasi-equilibrium, characterized by different temperatures of the Zeeman subsystem and the interaction subsystem. In this case the fluctuation dissipation theorem, which has so often been used for evaluating the enhanced susceptibility,[2,10,11] is difficult to use directly, and in this paper the enhanced susceptibility is evaluated using linear response theory. To the best of our knowedge, the enhanced susceptibility under conditions of two-temperature quasi-equilibrium in a rotating coordinate system has never before been studied even in the high-temperature approximation.

We shall consider a system of spins 1/2, coupled by dipole-dipole interactions, in the case of both magnetically regular and magnetically dilute systems.

2. LONGITUDINAL SUSCEPTIBILITY IN A ROTATING COORDINATE SYSTEM

Let us write the Hamiltonian of the spin system in the form

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_d+\mathcal{H}_\perp(t)+\mathcal{H}_{2z}(t), \qquad (1)$$

where $\mathcal{H}_0$ describes the interaction of the spin system with the static magnetic field: $\mathcal{H}_0 = \omega_0 S_x$, $\mathcal{H}_d$ is the Hamiltonian of the dipole-dipole interactions, $\mathcal{H}_\perp(t) = \omega_0 \cos \Omega t S_x$ is the interaction Hamiltonian of the spin system with the saturating transverse magnetic field, and $\mathcal{H}_{2z}(t) = \omega_2 \cos \omega t S_z$ is the interaction Hamiltonian with the weak low-frequency longitudinal field. The z axis is in the direction of the static field. The conditions under which one can neglect the contribution of the lattice to the longitudinal susceptibility are discussed below.

In the linear-response approximation the absorption of energy from the longitudinal field[12] is

$$\chi''(\omega)=\omega\int_0^\infty \Phi_{zz}(\tau)\cos\omega\tau d\tau, \qquad (2)$$

where the zz component of the relaxation tensor $\Phi_{\alpha\beta}$ appears in the relation

$$\langle M_z(\tau)\rangle - M_{0z}=\Phi_{zz}H_2, \qquad (3)$$

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H_2 being a static field turned on at time $\tau = 0$.

Let us pass to a coordinate system rotating at frequency Ω about the z axis. The Hamiltonian in the rotating coordinate system is of the form

$$\mathcal{H}^{\rm RCS} = \tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x} + \mathcal{H}_{2z}(\tau), \quad (4)$$

where $\tilde{\mathcal{H}}_0 = \Delta S_z$, with $\Delta = \omega_0 - \Omega$, $\mathcal{H}'_d$ is the secular part of the dipole-dipole interaction Hamiltonian, and $\tilde{\mathcal{H}}_{1x} = (\omega_1/2)(S_+ + S_1)$. To evaluate the linear response, we replace $\mathcal{H}_{2z}(\tau)$ by a "step":

$$\mathcal{H}^{\rm RCS}(\tau<0) = \tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x} + \mathcal{H}_{2z},$$
$$\mathcal{H}^{\rm RCS}(\tau\geqslant 0) = \tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x}. \quad (5)$$

The evolution of the system for $\tau > 0$ is found from the solution of the Liouville equation

$$\rho(\tau>0) = \exp[-i(\tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x})\tau]\rho(0) \times \exp[i(\tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x})\tau]. \quad (6)$$

We assume that $\rho(0)$ corresponds to quasi-equilibrium in the rotating coordinate system:

$$\rho(0) = \exp[-\alpha(\tilde{\mathcal{H}}_0 + \mathcal{H}_2) - \beta\mathcal{H}_d'] / \mathrm{Sp}\exp[-\alpha(\tilde{\mathcal{H}}_0 + \mathcal{H}_2) - \beta\mathcal{H}_d']. \quad (7)$$

Here α and β are the inverse temperatures of the Zeeman subsystem and dipole-dipole reservoir, respectively. To first order in $\mathcal{H}_2$ we have

$$\rho(0) = \rho_q(1 - \alpha\mathcal{H}_2 + \alpha\langle\mathcal{H}_2\rangle_q), \quad (8)$$

where

$$\rho_q = \exp(-\alpha\tilde{\mathcal{H}}_0 - \beta\mathcal{H}_d') / \mathrm{Sp}\exp(-\alpha\tilde{\mathcal{H}}_0 - \beta\mathcal{H}_d'),$$
$$\langle A\rangle_q = \mathrm{Sp}\, A\rho_q. \quad (9)$$

Substituting (8) into (6), we find the desired component of the relaxation tensor:

$$\Phi_{zz}(\tau) = \alpha\gamma^2[\langle S_z(\tau)S_z\rangle_q - \langle S_z(\tau)\rangle_q\langle S_z\rangle_q], \quad (10)$$

where

$$S_z(\tau) = \exp[i(\tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x})\tau] S_z \exp[-i(\tilde{\mathcal{H}}_0 + \mathcal{H}_d' + \tilde{\mathcal{H}}_{1x})\tau].$$

We expand $S_z(\tau)$ in a series in $\tilde{\mathcal{H}}_{1x}$ to second order, inclusive[10]:

$$S_z(\tau) = S_z - i\int_0^\tau dt_1[\tilde{\mathcal{H}}_{1x}(t_1), S_z] - \int_0^\tau dt_1\int_0^{t_1} dt_2[\tilde{\mathcal{H}}_{1x}(t_1), [\tilde{\mathcal{H}}_{1x}(t_2), S_z]], \quad (11)$$

where

$$\tilde{\mathcal{H}}_{1x}(t) = \exp[i(\tilde{\mathcal{H}}_0 + \mathcal{H}_d')t]\tilde{\mathcal{H}}_{1x}\exp[-i(\tilde{\mathcal{H}}_0 + \mathcal{H}_d')t] = \frac{\omega_1}{2}\exp(i\mathcal{H}_d't)(e^{i\Delta t}S_+ + e^{-i\Delta t}S_-)\exp(-i\mathcal{H}_d't). \quad (12)$$

This approximation is valid for frequencies $\omega \gtrsim W$, where W is the probability of a spin flip caused by the transverse magnetic field. With the aid of (11) we get

$$\Phi_{zz}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\Phi_{zz}(\tau)e^{i\omega\tau}d\tau = \frac{\gamma^2}{8\pi}\alpha\frac{\omega_1^2}{\omega^2}\int_{-\infty}^{\infty}d\tau[e^{i(\Delta+\omega)\tau} + e^{i(\Delta-\omega)\tau}]\{\langle[S_+(\tau), S_-]S_z\rangle_q - \langle[S_+(\tau), S_-]\rangle_q\langle S_z\rangle_q\}, \quad (13)$$

where

$$S_+(\tau) = \exp(i\mathcal{H}_d'\tau)S_+\exp(-i\mathcal{H}_d'\tau).$$

For our further use, and also for comparison with the known results in the HTA, we use the easily verified identity

$$\langle[S_+(\tau), S_-]S_z\rangle_q - \langle[S_+(\tau), S_-]\rangle_q\langle S_z\rangle_q = -\frac{1}{\Delta}\frac{\partial}{\partial\alpha}\langle[S_+(\tau), S_-]\rangle_q. \quad (14)$$

The correlation function $\langle[S_+(\tau), S_-]\rangle_q$, as we know,[3] determines the signal for the absorption of energy from a transverse rf field at arbitrary temperatures of the Zeeman subsystem and dipole-dipole reservoir.

For calculations using the high-temperatrure expansion for the dipole subsystem, it is convenient to transform the correlator obtained above to the form

$$\langle[S_+(\tau), S_-]\rangle_q = (1 - e^{\alpha\Delta})\langle S_+(\tau)S_-\rangle_q + e^{\alpha\Delta}\langle[S_+(\tau), e^{\beta\mathcal{H}_d'}]e^{-\beta\mathcal{H}_d'}S_-\rangle_q. \quad (15)$$

To first order in the indicated expansion

$$\langle[S_+(\tau), S_-]\rangle_q \approx (1 - e^{\alpha\Delta})\langle S_+(\tau)S_-\rangle_q^{(1)} + i\beta e^{\alpha\Delta}\frac{\partial}{\partial\tau}\langle S_+(\tau)S_-\rangle_0, \quad (16)$$

where $\langle A_0\rangle$ is the average over the density matrix

$$\rho_0 = \exp(-\alpha\Delta S_z) / \mathrm{Sp}\exp(-\alpha\Delta S_z),$$

and $\langle S_+(\tau)S_-\rangle_q^{(1)}$ should be evaluated to first order in $\beta\mathcal{H}'_d$. Thus, in this order everything can be expressed in terms of the correlation function $\langle S_+(\tau)S_-\rangle_q$. In particular, the moments M_1–M_4 have been calculated[3] for the function $\langle S_+(\tau)S_-\rangle_0$.

Combining the formulas given above and integrating the corresponding terms by parts, we obtain an expression for the part of the longitudinal susceptibility that is due to the spin-spin interactions:

$$\chi_s''(\omega) = \pi\omega\Phi_{zz}(\omega) = -\frac{\gamma^2}{8}\alpha\frac{\omega_1^2}{\omega}\frac{\partial}{\partial\alpha}\left\{\frac{1 - e^{\alpha\Delta}}{\Delta}\int_{-\infty}^{\infty}d\tau[e^{i(\Delta+\omega)\tau} + e^{i(\Delta-\omega)\tau}]\langle S_+(\tau)S_-\rangle_q^{(1)} + \beta\frac{e^{\alpha\Delta}}{\Delta}\int_{-\infty}^{\infty}[(\Delta+\omega)e^{i(\Delta+\omega)\tau} + (\Delta-\omega)e^{i(\Delta-\omega)\tau}]\langle S_+(\tau)S_-\rangle_0 d\tau\right\}. \quad (17)$$

In the limiting case of the HTA (for both subsystems) Eq. (17) gives

$$\chi_s''(\omega) = \alpha\frac{\gamma^2}{8}\frac{\omega_1^2}{\omega}\int_{-\infty}^{\infty}d\tau[e^{i(\Delta+\omega)\tau} + e^{i(\Delta-\omega)\tau}]\langle S_+(\tau)S_-\rangle, \quad (18)$$

which, in combination with (2), leads to the result obtained in Ref. 10 (for $\alpha = \beta$). In the case when the polarization is rather large and the high-temperature approximation can be used for the dipole subsystem (we shall call this approximation HTA_D), Eq. (17) gives

$$\chi_S''HTA_D(\omega)=\alpha\frac{\gamma^2}{8}\frac{\omega_1^2}{\omega}\int_{-\infty}^{\infty}d\tau\,[e^{i(\Delta+\omega)\tau}+e^{i(\Delta-\omega)\tau}]$$

$$\times\left[e^{\alpha\Delta}\langle S_+(\tau)S_-\rangle_0-\frac{1-\exp(\alpha\Delta)}{\Delta}\frac{\partial}{\partial\alpha}\langle S_+(\tau)S_-\rangle_0\right]. \quad (19)$$

Finally, formula (17) itself can be used to calculate the first (nonlinear-spin-temperature-effect) correction to (19).

Let us first restrict discussion to frequencies ω which are much greater than the spin-lattice relaxation rates τ_{SL}^{-1} and τ_{SSL}^{-1} of the Zeeman subsystem and dipole reservoir. From a theory[1] based on Provotorov's equations under conditions of strong saturation ($W\tau_{SL}\gg1$) we have $|\chi_l''/\chi_S''|\approx\tau_S/\tau_1^*$, where χ_l'' is due to the spin-lattice relaxation, τ_1^* is the spin-lattice relaxation time in the rotating coordinate system, and $\tau\rho_S$ is the time required for the mixing of the subsystems in the rotating coordinate system. For strong saturation we have $\tau_1^*\gg\tau_S$ for any relationship of Δ and D. Under conditions of intermediate saturation ($W\tau_{SL}\sim1$) on the remote wing of the line ($\Delta^2/D^2\gg1$), a similar analysis shows that the quantities τ_S and τ_1^* appearing in the ratio of the susceptibilities are replaced by $D^2/W\Delta^2$ and τ_{SL}, respectively. For a Lorentzian line shape, which is characteristic for low temperatures and/or magnetic dilution, we have $|\chi_l''/\chi_S''|\approx(D/\omega_1)^2(\delta\tau_{SL})^{-1}$, where δ is the half-width of the line. Although we must require $\omega_1/D\ll1$ for the Provotorov theory to apply, the product $\delta\tau_{SL}$ is usually very large, and the condition $|\chi_l''|\ll|\chi_S''|$ can in fact hold. Although the estimates given above correspond to the HTA, it seems that they are not affected very much by the value of the temperature. For this reason we shall confine our analysis in this paper to $\chi_S''|$ the spin-spin part of the longitudinal susceptibility. The question of the spin-lattice relaxation and the related longitudinal susceptibility under conditions of low-temperature quasi-equilibrium is rather complicated and should be the subject of a separate study.

Let us consider the cases of regular and random distributions of spins in the lattice. For regular systems a decrease in the temperature (an increase in the polarization) causes the line shape to change from approximately Gaussian to approximately Lorentzian, to narrow rapidly, to become asymmetric, and, generally speaking, to shift[3]; for $\alpha^{-1}\to0$ (i.e., for complete polarization) the linewidth goes to zero (if the lattice does not contain nonequivalent positions, which would lead to a set of narrow lines). For magnetically dilute systems, on the other hand, the linewidth depends only weakly on the temperature.[13] Qualitatively, this behavior is due to the strong difference between the local dipole fields at different centers, and this scatter in the local fields persists even when the spins are completely polarized.

3. REGULAR SYSTEMS

Let us discuss just the simple case of a spherical sample with a simple cubic lattice of spins. In this case the first moment (the line shift) in HTA_D is equal to zero and, neglecting the small asymmetry,[3] we can approximate the shape function of the line at sufficiently high polarization by a Lorentzian:

$$g(\nu)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\langle S_+(t)S_-\rangle_0e^{i\nu t}dt/\langle S_+S_-\rangle_0\to\frac{\delta}{\pi(\delta^2+\nu^2)}. \quad (20)$$

Cutting off the line at a frequency $\nu_0\gg\delta$, we express δ in terms of the second moment of function (20): $\delta=\pi M_2/2\nu_0$. This moment is known[3] for function (20):

$$M_2=M_2^0(1-p^2),$$

where M_2^0 is the second moment in the HTA, and $p=-\tanh(\alpha\Delta/2)$ is the polarization. We note that the general definition of the polarization is $P=\langle S_z\rangle_q/SN$; in HTA_D one has $P=p=\langle S_z\rangle_0/SN$, and for $S=1/2$

$$\langle S_z\rangle_0=-{}^1/_2N\tanh(\alpha\Delta/2).$$

The frequency ν_0 in this case is of the order of the nearest-neighbor interaction energy $(M_2^0)^{1/2}$; thus

$$\delta\approx\sqrt{M_2^0}(1-p^2),$$

and for $|p|\lesssim1$ we have $\nu_0\gg\delta$. In principle one does not have to introduce the frequency ν_0 but can use the second and fourth moments to estimate δ; this, however, without leading to qualitative changes in the results given below, makes the formulas much more unwieldy. Using (18) and performing straightforward manipulations, we obtain, in accordance with (2),

$$\chi_S''HTA_D(\omega)=-\frac{\pi}{2\Delta}\gamma^2\frac{\omega_1^2}{\omega}N\nu_0M_2^0(1-p^2)\,\mathrm{arc\,tanh}\,p$$
$$\times\{(1-p^2)[1/f_+(\Delta+\omega)+1/f_+(\Delta-\omega)]$$
$$+2p^2[f_-(\Delta+\omega)/f_+^2(\Delta+\omega)+f_-(\Delta-\omega)/f_+^2(\Delta-\omega)]\}, \quad (21)$$

where

$$f_\pm(\nu)=\pi^2M_2^{02}(1-p^2)^2\pm4\nu_0^2\nu^2.$$

All the temperature dependence in (21) has been reduced to a dependence on the polarization, which is considerably more convenient for using this formula. At large values of p, it follows from (21) that the dependence on p is carried by the function $(1-p)^2\,\mathrm{arctanh}\,p$, which goes to zero as $p\to1$. This result can be interpreted in the following way: when the spins are completely "frozen" in a strong field, the spin system cannot respond to the field $H_2(t)$ by a change in magnetization. However, our reasoning that p is close to unity at the same time the dipole-dipole reservoir remains high-temperature can be justified only for such large values of Δ that the enhanced-susceptibility effect loses meaning; therefore, expression (21) in actuality only describes the tendency of the susceptibility to fall off with increasing polarization at sufficiently high values of p.

The question of just what the value of p actually is should be solved with the aid of a low-temperature theory of saturation. Such a theory in HTA_D shows[9] that for strong saturation on the remote wing of the line, the Zeeman subsystem remains almost in equilibrium with the lattice, and after completion of the mixing process one has $\beta\approx\beta_L\omega_0/\Delta$, where β_L is the inverse lattice temperature. Here the quantity which determines the applicability of HTA_D, $\beta D=\beta_L\omega_0D/\Delta$, can be much smaller than unity even for $\beta_L\omega_0\gtrsim1$. In such a case $p=-\tanh(\beta_L\omega_0/2)$, and the conclusions reached above regarding the p dependence of $\chi_S''(\omega)$ is easily carried over to the dependence on β_L.

The frequency dependence of χ_S'' as given by formula (21) is qualitatively the same as in HTA_D (Ref. 10): it increases with increasing ω, has "resonance" peaks at frequencies $\omega = \pm\Delta$, and falls off rapidly with further increase in ω.

Let us track the trends of $\chi_S''(\omega)$ as the dipole-dipole reservoir cools. For this purpose we evaluate the moments M_1 and M_2 of function (20) to first order in the nonlinear-spin-temperature-effects approximation,[3] which has been used previously to describe the thermodynamic properties of spin systems on dynamic cooling. In the first order of this approximation the averages over the quasi-equilibrium density matrix (10) assume the form

$$\langle A\rangle_q \approx \langle A\rangle_0 - \beta(\langle A\mathscr{H}_d'\rangle_0 - \langle A\rangle_0\langle\mathscr{H}_d'\rangle_0), \tag{22}$$

where $\langle A\rangle_0$ is evaluated with the density matrix

$$\rho_0 = \exp(-\alpha\tilde{\mathscr{H}}_0)/\mathrm{Sp}\exp(-\alpha\tilde{\mathscr{H}}_0).$$

In the case of a spherical sample with a simple cubic spin lattice we have $\langle\mathscr{H}_d'\rangle_0 = 0$, and the formulas for M_1 and M_2 (obtained in the usual way[12]) reduce to

$$M_1 = (\langle[\mathscr{H}_d', S_+]S_-\rangle_0 - \beta\langle[\mathscr{H}_d', S_+]S_-\mathscr{H}_d'\rangle_0)/\langle S_+S_-\rangle_0, \tag{23}$$

$$M_2 = (\langle[\mathscr{H}_d', [\mathscr{H}_d', S_+]S_-]\rangle_0 - \beta\langle[\mathscr{H}_d', [\mathscr{H}_d', S_+]]S_-\mathscr{H}_d'\rangle_0)/\langle S_+S_-\rangle_0. \tag{24}$$

The first terms in (23) and (24) are familiar[3]: $M_1(\beta=0)=0$, $M_2(\beta=0)=M_2^0(1-p^2)$. After some rather awkward manipulations we get

$$M_1 = \beta\varphi_1(p), \quad M_2 = \varphi_2(p) + \beta\varphi_3(p),$$
$$\varphi_1(p) = -{}^1/_6 M_2^0(1-p^2)(3-2p), \quad \varphi_2(p) = M_2^0(1-p^2), \tag{25}$$

$$\varphi_3(p) = -\frac{3}{16}\sum_{i=0}^{4} c_i p^i,$$

where M_2^0 and c_i can be expressed in terms of the lattice sums

$$M_2^0 = 9\sigma_0/4, \quad c_0 = -4\sigma_1 + 3\sigma_2,$$
$$c_1 = -15\sigma_1 + 2\sigma_2, \quad c_2 = 8(2\sigma_1 - \sigma_2),$$
$$c_3 = 3(5\sigma_1 - \sigma_2), \quad c_4 = 6(\sigma_2 - 2\sigma_1),$$

where

$$\sigma_0 = \sum_k A_{jk}^2, \quad \sigma_1 = \sum_k A_{jk}^3, \quad \sigma_2 = \sum_{k,m} A_{jk}A_{km}A_{mj}.$$

For fields parallel to the axis of the lattice we have $\sigma_0 = 1.33D^2$, $\sigma_1 = -0.350D^3$, $\sigma_2 = 0.737D^3$ (Ref. 3), where $D = 1.58\gamma^2/a^3$. The above expressions imply, in particular, that the shift of the resonance frequency on cooling of the dipole-dipole reservoir appears even for spherical samples and is opposite in sign to β. Since this shift appears in the same order as the additional contribution to M_2, it must be taken into account in approximating the line shape function. For qualitative estimates we use the asymmetric normalized Lorentzian

$$g(\nu) = \delta(1+b\nu)/[\pi(\nu^2+\delta^2)],$$

with parameters expressed in terms of the moments as

$$b = M_1/M_2, \quad \delta = \pi M_2/2\nu_0.$$

We use this function to evaluate the correction to (21) due to deep cooling beyond the limits of HTA_D [corresponding to formula (17)]. Let us give only the result for $\omega \ll |\Delta|$:

$$\chi_S''(\omega) = \chi_S''^{\mathrm{HTA}_D}(\text{Eq. (21)}) + \beta\pi\gamma^2\frac{\omega_1^2}{\Delta\omega}\nu_0(1-p^2)\,\mathrm{arc\,tanh}\,p \times\frac{1}{f_+}\Big\{\frac{\Delta}{2}M_2^0(1-p^2)\Big(1+p-2p\frac{f_-}{f_+}\Big) - \Big(1+\frac{4}{f_+}\pi^2p^2M_2^0\varphi_2\Big)\Big(4\Delta\varphi_1 + 3\varphi_2 - \frac{2}{f_+}\pi^2\varphi_2^2\varphi_3\Big) - p\Big[\varphi_3' + 4\Delta M_2^0\Big(\frac{1}{3}+p-p^2\Big) - \frac{2}{f_+}\pi^2\varphi_2^2\varphi_3'\Big]\Big\}, \tag{26}$$

where $\varphi_3' = \partial\varphi_3(p)/\partial p$. At large values of p the factor in the β term in (26) takes on negative values, i.e., going outside of HTA_D leads to a decrease in χ_S''.

4. MAGNETICALLY DILUTE SYSTEMS

For magnetically dilute systems with an uncorrelated random distribution of spins over lattice sites we use the statistical theory of the line shape,[14] after first extending it to the low-temperature case, to evaluate the correlator $\langle[S_+(t), S_-]\rangle_q$ to second order in the expansion for the dipole subsystem [under certain circumstances the contribution to $\chi_S''(\omega)$ from the first order terms is equal to zero, while the complexity of the calculations increases rapidly with increasing order of the expansion]:

$$\langle K(t)\rangle_q \approx \langle K(t)\rangle_0 - \beta(\langle K(t)\mathscr{H}_d'\rangle_0 - \langle K(t)\rangle_0\langle\mathscr{H}_d'\rangle_0) + \frac{\beta^2}{2}(\langle K(t)\mathscr{H}_d'^2\rangle_0 - 2\langle K(t)\mathscr{H}_d'\rangle_0\langle\mathscr{H}_d'\rangle_0 - \langle\mathscr{H}_d'^2\rangle_0\langle K(t)\rangle_0 + 2\langle\mathscr{H}_d'\rangle_0^2\langle K(t)\rangle_0), \tag{27}$$

where $K(t) \equiv [S_+(t), S_-]$. Here, as above, we restrict discussion to the case of a spherical sample with a simple cubic lattice of spins $S = 1/2$. As usual in the statistical theory[14] we keep in $\mathscr{H}_d'$ only the anisotropic part of the secular dipole-dipole interaction

$$\mathscr{H}' = \frac{1}{2}\sum_{j,k} A_{jk}S_z^jS_z^k, \quad A_{jk} = \frac{3}{2}\frac{\gamma^2}{r_{jk}^3}(1-3\cos^2\theta_{jk}). \tag{28}$$

The details of the calculations in the statistical method will be reported elsewhere. All the correlators of interest, $\langle K(t)\rangle_0$, $\langle K(t)\mathscr{H}'\rangle_0$, $\langle K(t)\mathscr{H}'^2\rangle_0$ are found to be exponentially damped for $\gamma^1|t|/r_{\min}^3 \gg 1$, i.e., under conditions of strong magnetic dilution the central part of the line and that part of the wings which is of practical interest are Lorentzian. The damping rates turn out to be the same for all these correlators and do not depend on the temperature of the Zeeman system [this latter fact was noted previously[13] for a function close to $\langle K(t)\rangle_0$]. We have

$$\langle K(t)\rangle_0 = pNe(t), \quad \langle K(t)\mathscr{H}'\rangle_0 = i\delta\,\mathrm{sign}\,t\,e(t),$$
$$\langle K(t)\mathscr{H}'^2\rangle_0 = -\frac{1}{4}pN\delta^2 e(t), \tag{29}$$

$$e(t) = \exp(-\delta|t|), \quad \delta = C\frac{2\pi^2\gamma^2}{3^{3/2}},$$

where δ is the half-width of the line and C is the concentration.

Using (29), we reduce $\Phi_{zz}(\omega)$ from (13) to the form [with allowance for (14)]:

$$\Phi_{zz}(\omega)=-\frac{\gamma^2}{8\pi}\alpha\frac{\omega_1^2}{\omega^2\Delta}N\int_{-\infty}^{\infty}d\tau[e^{i(\Delta+\omega)\tau}+e^{i(\Delta-\omega)\tau}]$$

$$\times\frac{\partial}{\partial\alpha}\left[p-i\frac{\beta}{2}\delta\,\mathrm{sign}\,t-\frac{\beta^2}{8}p\delta^2\right]\exp(-\delta|t|). \quad (30)$$

Using (2), we find

$$\chi_S''(\omega)=-\frac{N}{2}\gamma^2\frac{\omega_1^2}{\omega\Delta^2}(1-p^2)\,\mathrm{arctanh}\,p\left(1-\frac{\beta^2}{8}\delta^2\right)$$

$$\times\left[\frac{\delta}{(\Delta+\omega)^2+\delta^2}+\frac{\delta}{(\Delta-\omega)^2+\delta^2}\right]. \quad (31)$$

From this expression we can clearly see how cooling both subsystems affects the behavior of $\chi_S''(\omega)$, which grows and then decays $\propto(1-p^2)\mathrm{arctanh}\,p$ with increasing p, and decays $\propto(1-\beta^2\delta^2/8)$ as the "cooling factor" $|\beta|\delta$ of the dipole-dipole reservoir, increases.

We note that in the case under discussion the high-temperature expansion for a system of interactions is simultaneously a concentration expansion (the expansion parameter is $\beta\delta$, and δ is proportional to C). This circumstance is nontrivial and even somewhat unexpected, since the most important characteristic of a dipole-dipole reservoir is the local-field frequency D, which is proportional to $C^{1/2}$. At low concentrations one has $D\gg\delta$. In particular,[15] at a temperature of the dipole-dipole reservoir of order D/k the dynamic cooling of the spin system gives rise to a nonzero order parameter (of the Edwards-Anderson type), while at a temperature of order δ/k the "freezing" of the spins in the local fields is already complete. Thus, in spite of the use of the high-temperature expansion, formula (31) encompasses a rather wide interval of β. It must be kept in mind, however, that outside of the HTA the quantities α and β are no longer related in a simple way to the observable quantities, i.e., the polarization P and the average dipole energy $\langle\mathscr{H}_d'\rangle_q$, and if we transform from p and β to P and $\langle\mathscr{H}_d'\rangle_q$, the high-temperature expansion will no longer be an expansion in integral powers of the concentration, but will assume a more complicated structure.

5. CONCLUSION

The results given above permit the assertion that the enhanced-susceptibility effect also exists at low spin temperatures and that measurements of the frequency dependence of the enhanced susceptibility and the dependence on the initial polarization and average energy of the dipole-dipole reservoir yield important information on the dynamics of both regular and magnetically dilute spin systems. The theory set forth above, however, will remain incomplete until the development of a theory of saturation capable of describing the evolution of spin subsystems at a low temperature of the dipole-dipole reservoir. Such a theory should enable one to evaluate the temperatures of the subsystems (or their average energies) on completion of a certain process (e.g., demagnetization in a rotating coordinate system) for given initial temperature. Nevertheless, the relations given above permit estimation of the temperature dependence of the longitudinal susceptibility, or, on the other hand, if one has the experimental data, these relations can be used to estimate the subsystem temperatures.

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