The electron-paramagnetic resonance linewidth of double-exchange interaction systems

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Abstract
We studied the electron-paramagnetic resonance linewidth of colossal
magnetoresistance manganites. Starting from the quantum Langevin equation,
we derived the transverse susceptibility of the double-exchange interaction
systems and identified the damping function to be the inverse of the relaxation
time T_2. We argued that the anisotropic energy caused by the crystal field
was the most probable relaxation mechanism. The spin correlation caused
by the double-exchange interaction was examined with the Schwinger boson
approach. Taking into account the constraints of Schwinger bosons, we found
that the linewidth is approximately proportional to the temperature. At the
low-temperature end (near the Curie temperature T_C), there is a minimum,
below which the linewidth increases sharply. At the high-temperature limit,
the linewidth is proportional to $\sqrt{T/T_C}$. Most importantly, the linewidth is a
universal function of T/T_C.

1. Introduction

It was found 50 years ago that perovskite manganites ($La_{1-x}A_xMnO_3$, where A is the divalent
element and x the doping concentration) show many interesting phenomena [1]. In the past 15
years, there has been a renewed interest in this kind of material due to the discovery of colossal
magnetoresistance (CMR) [2]. Later studies [3] revealed a complex phase diagram, e.g.
ferromagnetic insulator, ferromagnetic metal, paramagnetic insulator and antiferromagnetic
insulator and very rich physics, e.g. charge ordering, orbit ordering and the Jahn–Teller effect.
The fundamental mechanism in manganites is the double-exchange (DE) interaction, which
was first proposed by Zener [4] and later developed by Anderson and Kasegawa [5] and
de Gennes [6]. However, lattice distortion, orbital degeneracies and superexchange (SE)
interaction all play significant roles [7, 8]. The interplay of these interactions make these
systems even more complex and intriguing.

Among the studies on manganites, there is the electron-paramagnetic resonance (EPR)
experiment [9–15], which is very helpful for understanding the magnetism-related interac-
tion and spin correlation. Peak position and peak intensity provide information about the
g-factor and constituents. From the linewidth, one can find clues to the coupling strength,

spin-relaxation mechanism and susceptibility. Results of previous experiments showed that the linewidth of EPR spectra in the paramagnetic phase increases almost linearly with temperature [10–14]. The origin is not clear. It was suggested to be spin-lattice relaxation [12]. A more recent analysis of experimental data [13] proposed an expression, $C(T - \theta)/T$, for linewidth and achieved a good fit. However, there is obviously more work to be done. The most important task is a rigorous derivation of linewidth from the fundamental mechanism. This is the aim of the present study. First, we construct the relation between susceptibility and the relaxation time of spins using the Langevin equation. This is presented in section 2. In section 3, the DE interaction Hamiltonian is reduced to a simpler form. We calculate the spin-correlation function and susceptibility in section 4 and present the results with some discussion in section 5.

2. Formalism of transverse susceptibility

The EPR lineshape can be computed from the transverse susceptibility. Suppose that, in addition to a static magnetic field H_0 in the z-direction, one applied microwaves with magnetic field in the x-direction to the system. The real (imaginary) part of the transverse susceptibility is given by the magnetization in the $x(y)$-direction divided by the microwave magnetic field. By solving the Bloch equations [16], the transverse susceptibility is given by

$$\chi(\omega) = -\frac{\Omega \chi_0}{2(\omega - \Omega + i/T_2)}, \tag{1}$$

where $\Omega = g\mu_B H_0$ and $\chi_0 = M_z/H_0$, is the static susceptibility. The linewidth is usually denoted as $1/T_2$. There are many ways to calculate the transverse susceptibility and, hence, $1/T_2$. In this paper, we adopt the method of the quantum Langevin equation [17]. Using this method, we were able to treat a microscopic mechanism systematically. We start with the linear response theory. The negative imaginary part of the transverse susceptibility is given by [18]

$$\chi''(\omega) = \frac{(g\mu_B)^2}{4V} \text{Re} \int_0^\infty e^{i\omega t} \langle [S_-, S_+(t)] \rangle dt, \tag{2}$$

where $\mathbf{S}$ is the total spin of the system; $\langle x \rangle$ represents the thermal average of operator x and $\hbar$ was set to unity. Without loss of generality, the above equation can be written as

$$\chi''(\omega) = \frac{(g\mu_B)^2}{4V} \text{Re} \sum_{l,m} (S_-)_{ml} (S_+(\omega))_{lm} (e^{-\beta E_m} - e^{-\beta E_l}). \tag{3}$$

Equation (3) can be calculated with the Langevin equation of $S_+(t)$,

$$\frac{dS_+(t)}{dt} = -i\Omega S_+(t) - \int_0^t \Phi(t - s) S_+(s) ds + R(t), \tag{4}$$

where $R(t)$ is the random force ($\langle R(t) \rangle = 0$) and $\Phi(t)$ is the damping function. After Laplace–Fourier transform, equation (4) has the form

$$S_+(\omega) = \frac{i[S_+(t = 0) + r(\omega)]}{\omega - \Omega + i\phi(\omega)}, \tag{5}$$

where $r(\omega)$ and $\phi(\omega)$ are the Laplace–Fourier transforms of $R(t)$ and $\Phi(t)$, respectively. Substituting equation (5) into equation (3), one can calculate $\chi''(\omega)$,

$$\chi''(\omega) = \frac{(g\mu_B)^2}{4V}(1 - e^{-\beta\Omega})\text{Re}\frac{-i\langle S_+S_-\rangle}{\omega - \Omega + i\phi(\omega)}. \tag{6}$$

By comparing with equation (1), we identified $\text{Re}\,\phi(\omega)$ with $1/T_2$.

$\Phi(t)$ is a force–force correlation function. (Interested readers are referred to the works of Kubo [17] and Mori [19]),

$$\Phi(t) = \frac{k_BT}{\langle S_+S_-\rangle}\int_0^\beta \mathrm{d}\lambda\langle e^{\lambda\mathcal{H}}\dot{S}_+(t)e^{-\lambda\mathcal{H}}\dot{S}_-\rangle. \tag{7}$$

There are at least two ways of calculating $\phi(\omega)$. Mori [19] developed a method of continual fractions. We found that the fractions quickly become complicated and intractable in the double-exchange system. Therefore we adopted the approach proposed by Ting and Nee [20]. Inserting complete sets of states into equation (7), we obtain

$$\Phi(t) = \frac{1}{\langle S_+S_-\rangle}\sum_{n,m}\frac{e^{-\beta E_m} - e^{-\beta E_n}}{\beta E_{nm}}\dot{S}_{+nm}\dot{S}_{-mn}e^{iE_{nm}t}. \tag{8}$$

Hence,

$$\phi(\omega) = -\frac{1}{\langle S_+S_-\rangle}\sum_{n,m}\frac{e^{-\beta E_m} - e^{-\beta E_n}}{\beta E_{nm}}\frac{\dot{S}_{+nm}\dot{S}_{-mn}}{i\omega + iE_{nm} - \eta}. \tag{9}$$

Its real part is

$$\text{Re}\,\phi(\omega) = \frac{\pi}{\langle S_+S_-\rangle}\sum_{n,m}\frac{e^{-\beta E_m} - e^{-\beta E_n}}{\beta E_{nm}}\dot{S}_{+nm}\dot{S}_{-mn}\delta(\omega + E_{nm}). \tag{10}$$

Finally, we have

$$\text{Re}\,\phi(\omega) = \text{Im}\frac{\psi(\omega) - \psi(0)}{\omega}, \tag{11}$$

where

$$\psi(\omega) = \frac{ik_BT}{\langle S_+S_-\rangle}\int_0^\infty \mathrm{d}t\langle[\dot{S}_-, \dot{S}_+(t)]\rangle e^{i\omega t} \tag{12}$$

is the response function of $\dot{S}_+$. Equation (11) is readily obtained by inserting complete sets of states between the operators of equation (12). Now, we are ready to apply the above method to the physical systems we are interested in. Our strategy is to calculate the relevant terms of $\psi(\omega)$ with Matsubara formalism and then use analytical continuation. The result is substituted into equation (11) to evaluate the linewidth.

3. Double-exchange interaction

The fundamental mechanism in CMR manganites is the DE interaction. The Hamiltonian has the following form:

$$H = -t\sum_{\mathbf{k},\sigma}c_{\mathbf{i}\sigma}^\dagger c_{\mathbf{j}\sigma} + J\sum_{\mathbf{i}}\mathbf{S_i}\cdot\sigma_\mathbf{i} + g\mu_B H_0\sum_{\mathbf{i}}S_{\mathbf{i}z}. \tag{13}$$

The first term of equation (13) describes the hopping of the holes between Mn^{+3} and Mn^{+4} sites. The second term is Hund's coupling, when combined, they give the DE interaction. The third term is due to the applied field. In the hole picture, the manganese ions have a spin, $S = 2$. Whenever a hole is present, the local spin is coupled strongly with the spin of the hole. According to Hund's rule, the antiparallel state has the lower energy. The level spacing J is of the order of eV and, hence, is the largest energy in the system. To treat it correctly, we used the approach proposed by Kubo and Ohata [21]. They used projection operators to impose a restriction on holes. Hoppings can only occur at those sites where the local spins are antiparallel to those of holes. Thus, the first and second terms of equation (13) are replaced by

$$H_{DE} = -t \sum_{\mathbf{i},\mathbf{j},\alpha,\beta,\gamma} c_{\mathbf{i}\alpha}^{\dagger} P_{\mathbf{i}\alpha\gamma} P_{\mathbf{j}\gamma\beta} c_{\mathbf{j}\beta}, \tag{14}$$

where α, β and γ are spin indices and

$$P_{\mathbf{i}} = \frac{S - \mathbf{S_i} \cdot \sigma}{2S + 1}, \tag{15}$$

with $\sigma/2$ denoting the spins of the conduction holes. It has been shown that H_{DE} in equation (14) is equivalent to the following Hamiltonian [22–24]:

$$H_{DE} = -\frac{t}{2S + 1} \sum_{\mathbf{i},\mathbf{j}} f_{\mathbf{i}}^{\dagger} (A_{\mathbf{j}}^{\dagger} A_{\mathbf{i}} + B_{\mathbf{j}}^{\dagger} B_{\mathbf{i}}) f_{\mathbf{j}}, \tag{16}$$

where the slave-fermion method was applied:

$$c_{i\uparrow} = f_i \alpha_i, \tag{17a}$$

$$c_{i\downarrow} = f_i \beta_i \tag{17b}$$

and the spin-up and spin-down operators α and β combined with the localized spins were represented by the Schwinger boson operators A and B:

$$A^{\dagger}|S - 1/2, m - 1/2\rangle = \sqrt{S + m}|S, m\rangle, \tag{18a}$$

$$B^{\dagger}|S - 1/2, m + 1/2\rangle = \sqrt{S - m}|S, m\rangle. \tag{18b}$$

There is a constraint due to the slave fermion and Schwinger boson approaches [25]

$$A_{\mathbf{i}}^{\dagger} A_{\mathbf{i}} + B_{\mathbf{i}}^{\dagger} B_{\mathbf{i}} + f_{\mathbf{i}}^{\dagger} f_{\mathbf{i}} = 2S. \tag{19}$$

In the momentum space, we have

$$H_{DE} = -\frac{1}{N(2s + 1)} \sum_{\mathbf{k},\mathbf{p},\mathbf{q}} E_{\mathbf{k}-\mathbf{p}-\mathbf{q}} f_{\mathbf{k}-\mathbf{q}}^{\dagger} (A_{\mathbf{p}} A_{\mathbf{p}+\mathbf{q}}^{\dagger} + B_{\mathbf{p}} B_{\mathbf{p}+\mathbf{q}}^{\dagger}) f_{\mathbf{k}}, \tag{20}$$

where

$$E_{\mathbf{k}} = t \sum_{n.n.} e^{i\mathbf{k}\cdot\mathbf{R}_{nn}}, \tag{21}$$

with the summation being over the nearest neighbours. It is clear that the bosons and fermions are closely related to each other. To show the implications of the Hamiltonian in equation (20), we give the boson and fermion energies at the mean-field level, respectively, as follows:

$$\omega_{\mathbf{q}} = \frac{1}{N(2s + 1)} \sum_{\mathbf{k}} E_{\mathbf{k}+\mathbf{q}} \langle f_{\mathbf{k}}^{\dagger} f_{\mathbf{k}} \rangle \tag{22}$$

and

$$\varepsilon_{\mathbf{k}} = \frac{1}{N(2s+1)} \sum_{\mathbf{p}} E_{\mathbf{k+p}} \langle A_{\mathbf{p}}^{\dagger} A_{\mathbf{p}} + B_{\mathbf{p}}^{\dagger} B_{\mathbf{p}} \rangle. \tag{23}$$

Thus we have simplified the DE Hamiltonian considerably.

There exists an SE interaction between manganese ions. The manganites exhibit an antiferromagnetic phase whenever the DE interaction is diminished. The Néel temperature is about 200 K. This indicates that the SE interaction coupling J_{SE} is of the order 1 meV or $t/100$. In the temperature range 300–1000 K, the thermal fluctuation is more important than that caused by the SE interaction. The latter can be accounted for with random-phase approximation [26]. The interaction between manganese ions (indirect exchange through the DE interaction) is 'screened' by the factor, $1 + J_{SE}/(i\omega_n - \omega_0)$, where ω_0 is the spin-level splitting. This, in effect, decreases the spin stiffness and the T_C. In the following calculation, we more or less incorporate the effect of the SE interaction by using an effective spin stiffness. On the other hand, as the fluctuation caused by the SE interaction is more important near the T_C, we expect the slope of $1/T_2$ to decrease as the low-temperature end is lifted.

4. Relaxation

The reason for broadening in the EPR spectra is not clear. The dipolar interaction is too small in magnitude. It produces a linewidth of a few G. The Dzyaloshinsky–Moriya (DM) interaction [27] is a possible candidate. The ESR data of Alejandro $et\ al$ [28] on $La_{7/8}Sr_{1/8}MnO_3$ suggests that the DM interaction is of the order 0.4 K. The data of [29] gives the order of 1 K, the analysis of Huber $et\ al$ [30] gives 0.8 K and that of Deisenhofer $et\ al$ [31] gives 0.25 K. However, note that all the above studies are concerned with the insulating region, i.e. small doping. In the metallic region that we are interested in, the DM interaction will be much smaller for the following reasons. First, the lattice distortion is smaller. Secondly, the electron orbits are less localized and the orbital characteristics are less manifest. Lastly, the broadening should have been closely related to the Lande-g factor if indeed the DM interaction was the cause [27]. Experimental data [13] showed that $g \simeq 2.0$, an indication that the DM interaction is not robust.

Lattice distortion is another possibility; however, the phonon-related mechanism should have given a different temperature dependence. We have a more detailed discussion in the next section. The remaining candidate is the anisotropy energy of spins. In manganites, a manganese ion and neighbouring oxygen atoms formed an octahedron. Two octahedrons were rotated in the opposite direction and the bond angle of Mn–O–Mn deviated from π. Incidentally, experiments [32] showed that the bond angle affected the bandwidth, hence, it is an important parameter. For our purpose, the deviation of bond angle or bond bending broke significantly the cubic symmetry. In a system of cubic symmetry, the degeneracy of d-orbitals was lifted partially to e_g and t_{2g} orbitals. At systems with still lower symmetry, such as those in the presence of tetragonal distortion, an additional ligand field should be introduced into the Hamiltonian. It takes the form $B_2 O_2^0$, where O_2^0 is a spin operator and B_2 is the field strength [33]. Thus, we have the following general form of an additional term in our Hamiltonian:

$$H_{ani} = \sum_{\mathbf{i}} (F S_{\mathbf{i}x}^2 + G S_{\mathbf{i}y}^2), \tag{24}$$

where F and G are constants. Since the DE interaction conserves total spin, we found

$$\dot{S}_+ = i[H_{ani}, S_+]$$

$$= -\frac{i}{2} \sum_{\mathbf{i}} [(F-G)(S_{\mathbf{i}-}S_{\mathbf{i}z} + S_{\mathbf{i}z}S_{\mathbf{i}-}) + (F+G)(S_{\mathbf{i}+}S_{\mathbf{i}z} + S_{\mathbf{i}z}S_{\mathbf{i}+})]. \tag{25}$$

Hence,

$$
\langle [\dot{S}_+(t), \dot{S}_-] \rangle = \frac{1}{4} \sum_{\mathbf{i},\mathbf{j}} \{ (F - G)^2 \langle [S_{\mathbf{i}z}(t) S_{\mathbf{i}-}(t) + S_{\mathbf{i}z}(t) S_{\mathbf{i}-}(t), S_{\mathbf{j}z} S_{\mathbf{j}+} + S_{\mathbf{j}+} S_{\mathbf{j}z}] \rangle
$$

$$
+ (F + G)^2 \langle [S_{\mathbf{i}z}(t) S_{\mathbf{i}+}(t) + S_{\mathbf{i}+}(t) S_{\mathbf{i}z}(t), S_{\mathbf{j}z} S_{\mathbf{j}-} + S_{\mathbf{j}-} S_{\mathbf{j}z}] \rangle \}. \tag{26}
$$

Since the calculation was performed in the paramagnetic phase, $\mathbf{i}$ and $\mathbf{j}$ have to be equal. Furthermore, equation (26) can be simplified to the following form since the decouplings, such as $\langle S_{\mathbf{i}z}(t) S_{\mathbf{i}+}(t) S_{\mathbf{i}z} S_{\mathbf{i}-} \rangle \approx \langle S_{\mathbf{i}z}(t) S_{\mathbf{i}z} \rangle \langle S_{\mathbf{i}+}(t) S_{\mathbf{i}-} \rangle$, are good approximations in the paramagnetic phase:

$$
\langle [\dot{S}_+(t), \dot{S}_-] \rangle = \frac{\langle S_z^2 \rangle}{N} \sum_{\mathbf{i}} \{ (F - G)^2 \langle [S_{\mathbf{i}-}(t), S_{\mathbf{i}+}] \rangle + (F + G)^2 \langle [S_{\mathbf{i}+}(t), S_{\mathbf{i}-}] \rangle \}. \tag{27}
$$

To calculate $\psi(\omega)$, we replaced the spin operators by Schwinger boson operators

$$
S_{+\mathbf{i}} = A_{\mathbf{i}}^\dagger B_{\mathbf{i}}, \tag{28a}
$$

$$
S_{-\mathbf{i}} = B_{\mathbf{i}}^\dagger A_{\mathbf{i}}, \tag{28b}
$$

and introduced their respective Green functions

$$
\widetilde{D}_{iA}(\tau) = -\langle T A_i(\tau) A_i^\dagger \rangle, \tag{29a}
$$

$$
\widetilde{D}_{iB}(\tau) = -\langle T B_i(\tau) B_i^\dagger \rangle. \tag{29b}
$$

Substituting equations (27)–(29) into equation (12), we obtained

$$
\psi(\mathrm{i}\omega_n) = \frac{k_B T}{2N} \sum_{\mathbf{i}} \int_0^\beta \mathrm{d}\tau\, \mathrm{e}^{\mathrm{i}\omega_n \tau} [(F - G)^2 \widetilde{D}_{iA}(-\tau) \widetilde{D}_{iB}(\tau)
$$

$$
+ (F + G)^2 \widetilde{D}_{iA}(\tau) \widetilde{D}_{iB}(-\tau)]. \tag{30}
$$

In the paramagnetic phase, the zeroth-order spin propagators are

$$
D_A(\mathbf{q}, \mathrm{i}\omega_n) = D_B(\mathbf{q}, \mathrm{i}\omega_n) = \frac{1}{\mathrm{i}\omega_n - \omega_{\mathbf{q}}}. \tag{31}
$$

Applying Fourier transform and Matsubara summation, we obtain

$$
\psi(\mathrm{i}\omega_n) = \frac{k_B T}{N^2} (F^2 + G^2) \sum_{\mathbf{p},\mathbf{q}} \frac{n_{\mathbf{p}} - n_{\mathbf{q}}}{\mathrm{i}\omega_n - \omega_{\mathbf{p}} + \omega_{\mathbf{q}}}, \tag{32}
$$

where $n_{\mathbf{p(q)}}$ is the Bose–Einstein distribution function. Substituting equation (32) into equation (11), we obtained

$$
\mathrm{Re}\, \phi(\omega) = -\frac{\pi k_B T}{N^2} (F^2 + G^2) \sum_{\mathbf{p},\mathbf{q}} \frac{n_{\mathbf{q}} - n_{\mathbf{p}}}{\omega_{\mathbf{q}} - \omega_{\mathbf{p}}} \delta(\omega - \omega_{\mathbf{p}} + \omega_{\mathbf{q}}). \tag{33}
$$

If one writes

$$
\omega_{\mathbf{q}} = D q^2, \tag{34}
$$

where D is the spin stiffness, equation (33) can be reduced to

$$
\mathrm{Re}\, \phi(0) = -\frac{\pi k_B T V^2}{(2\pi)^6 N^2} \int \mathrm{d}^3 p \int \mathrm{d}^3 q\, \delta(\omega_{\mathbf{p}} - \omega_{\mathbf{q}}) \frac{\partial n_{\mathbf{p}}}{\partial \omega_{\mathbf{p}}}
$$

$$
= -\frac{(k_B T a^3)^2}{2(2\pi D)^3} (F^2 + G^2) \ln|1 - z|, \tag{35}
$$

with z being the fugacity and a the lattice constant. The fugacity is related to the constraint of the Schwinger bosons. In view of equation (19), the constraint in momentum space is

$$\sum_{\mathbf{q}} A_{\mathbf{q}}^{\dagger} A_{\mathbf{q}} = N(S - x/2), \tag{36}$$

where we have made the approximation of replacing local constraints with a global constraint and used the fact that in the paramagnetic phases A and B, bosons are equally populated. It is important to distinguish between D and the usual spin wave stiffness. The relation between spin operators and Schwinger bosons are, for example, $S_{+} = A^{\dagger} B$. Clearly, they are not the same. The spin wave propagator is a convolution of those of Schwinger bosons. Spin wave stiffness should be computed accordingly. Here, we are not concerned with spin waves. Equation (36) can be written as

$$\left(\frac{k_B T a^2}{4\pi D} \right)^{3/2} g_{3/2}(z) + \frac{z}{N(1-z)} = (S - x/2), \tag{37}$$

where the second term comes from the state at $q = 0$ [34], and

$$g_n(z) = \frac{1}{\Gamma(n)} \int_0^{\infty} \frac{x^{n-1}\, dx}{z^{-1} e^x - 1}, \tag{38}$$

with $\Gamma(n)$ being the gamma function. It is necessary for the ferromagnetic phase for the following reasons. If the dispersion relation of equation (22) were used, we would have $D \approx x t a^2 / (2S + 1)$. For a typical system of $t = 0.1\,\text{eV}$, $x = 0.3$ and $T = 200\,\text{K}$, the prefactor of $g_{3/2}(z)$ is approximately 0.13. Since $S = 2$ and the largest value of $g_{3/2}(z)$ is $g_{3/2}(1) \approx 2.6$, the presence of the second term of equation (37) is necessary. This is an indication that the spin stiffness given above needs to be revised. It is valid at a low temperature. As the temperature increases, the motion of holes will be impeded by spin fluctuations. As a result, the coupling between spins will be weakened and the spin stiffness is decreased. The computation of the actual value of D is very complicated and beyond the scope of this paper. Hence, we used a simple approach here. We assume that, at the Curie temperature, the second term on the left hand side of equation (37) vanishes, i.e.

$$\left(\frac{k_B T_C a^2}{4\pi D} \right)^{3/2} g_{3/2}(1) = (S - x/2). \tag{39}$$

We evaluated D in this way. Above T_C, the system is always insulating and the spin orientations are random. We can assume that D remains more or less temperature-independent in the paramagnetic phase. Now, the fugacity can be computed with the following equation:

$$\left(\frac{T}{T_C} \right)^{3/2} \frac{g_{3/2}(z)}{g_{3/2}(1)} = 1 \tag{40}$$

for temperatures greater than T_C. Substituting equation (39) into (35), we obtained

$$\frac{1}{T_2} = \text{Re}\, \phi(0) = -4 \frac{F^2 + G^2}{k_B T_C} \left(\frac{S - x/2}{g_{3/2}(1)} \right)^2 \left(\frac{T}{T_C} \right)^2 \ln|1 - z|. \tag{41}$$

Equations (40) and (41) are our final result.

In retrospect, the major mathematical structure is the factor $\partial n_{\mathbf{p}} / \partial \omega_{\mathbf{p}}$ in equation (35). It is quite robust. Its presence is not affected by taking into account the self-energy of boson propagators. We show this in the appendix. Hence, the expression in equation (41) seems to be universal.
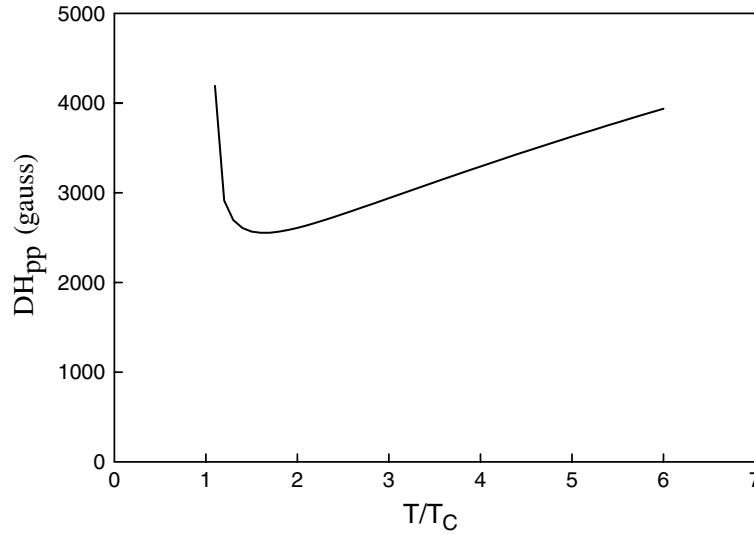
Figure 1. The linewidth, ΔH_{pp}, or the peak-to-peak magnetic field difference against T/T_C for $F = G = 2\,\text{K}$.

5. Results and discussion

All the experimental data showed that the EPR linewidth of manganites with doping between 0.2 and 0.4 has a quasi-linear dependence on temperature. Our calculation has a similar result (figure 1). At the low-temperature end (near Curie temperature T_C), there is a minimum. Below the minimum, the linewidth increases sharply. With increase in temperature, the linewidth increases almost linearly. It bends downward in the high-temperature region. The quasi-linear dependence on temperature has been attributed to various sources, notably the one-phonon spin-lattice relaxation proposed by Huber and Seehra (HS) [35] and Dormann–Jaccarino–Huber's (DJH) [36–38] consideration of dipolar interaction or any other temperature-independent interaction. In any case, spin-lattice relaxation cannot give the correct temperature dependence. For example, in equation (A-5) of [3], $\chi_T \, k_B T$ is almost constant in the high-temperature region. The phonon distribution function $n_{\mathbf{p},\mathbf{q}}$ gives a factor of $k_B T$. The spin correlation function introduces additional temperature dependence, except for, perhaps, in the high-temperature limit. Thus $1/T_2$ cannot be quasi-linear. As for DJH's explanation, we found in Huber's original derivation [37, 38] that $1/T_2$ is proportional to $k_B T \xi^{5/2}$. The correlation length ξ has to be temperature-independent for $1/T_2$ to be linearly proportional to T. This is unlikely and, hence, we concluded that neither can produce linear temperature dependence.

There is a sharp upturn at the low-temperature end. Experimentally, all the powder samples showed this kind of behaviour. Data from single-crystal samples are not conclusive. Therefore there was ambiguity as to whether it is a sample-dependent effect. Our calculation showed that it is intrinsic. The cause is the divergence of $\ln(1 - z)$ at T_C. It is universal. At temperatures close to T_C [39],

$$g_{3/2}(z) \simeq g_{3/2}(1) - 3.54\sqrt{\delta z}, \tag{42}$$

where $\delta z = 1 - z$. In view of equation (40), we obtained

$$\delta z \simeq 1.23\varepsilon^2, \tag{43}$$

where $\varepsilon = T/T_C - 1$. Thus we concluded that, near T_C, the divergence is of the form

$$\frac{1}{T_2} \propto -\ln \varepsilon. \tag{44}$$

The high-temperature behaviour of linewidth is straightforward. In view of equation (40), $g_{3/2}(z)$ and, hence, z is small. Since

$$g_{3/2}(z) = \sum_n \frac{z^n}{n^{3/2}}, \tag{45}$$

we have, for $T \to \infty$,

$$z \simeq g_{3/2}(1) \left(\frac{T_C}{T} \right)^{3/2}. \tag{46}$$

Hence,

$$\frac{1}{T_2} = \mathrm{Re}\, \phi(0) \longrightarrow 4g_{3/2}(1) \left(\frac{S - x/2}{g_{3/2}(1)} \right)^2 \frac{F^2 + G^2}{k_B T_C} \sqrt{\frac{T}{T_C}}. \tag{47}$$

This is compatible with the experimental evidence [13] showing deviation (bending downward) from linear temperature dependence.

In all the experiments, $1/T_2$ is a fraction of a Tesla. In view of equation (40), F and G being 1 K is enough to give the linewidth. A careful analysis of EPR data [29] indicated that this magnitude is reasonable. The only discrepancy between our calculation and experimental results is in the magnitude of the minimum $1/T_2$. This discrepancy may be due to our treatment of the constraints. We have replaced the constraint on each site (equation (19)) by a global one (equation (36)). If the local constraints were restored, the correlation between spins became stronger. The fluctuation ($1/T_2$) would be suppressed. Improvement on this point is one of our ongoing projects.

It has been proposed in [30, 37, 38] that linewidth has the following form over the high-temperature range:

$$\Delta H_{pp} = [\chi_0(T)/\chi(T)]\Delta H_{pp}(T \to \infty), \tag{48}$$

where $\chi(T)$ and $\chi_0(T)$ are, respectively, the susceptibilities of the spins with DE interaction and isolated spins. The argument is that, when the temperature is higher than all the energy scales, the response function $\langle [\dot{\mathbf{S}}(\tau), \dot{\mathbf{S}}] \rangle$ should be independent of temperature. Therefore it is interesting to examine whether double-exchange systems follow this relation. Equation (13) can be written in terms of Schwinger boson operators as follows:
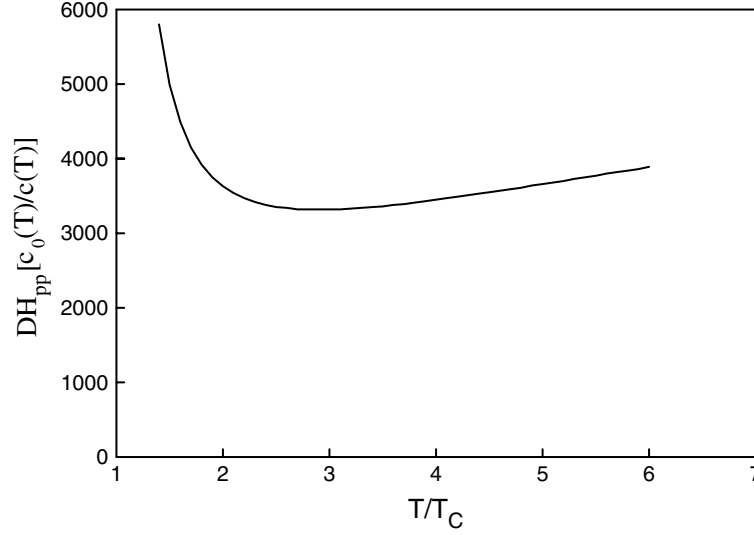
$$H = -\frac{1}{N(2s+1)} \sum_{\mathbf{k},\mathbf{p},\mathbf{q}} E_{\mathbf{k}-\mathbf{p}-\mathbf{q}} f_{\mathbf{k}-\mathbf{q}}^\dagger (A_\mathbf{p} A_{\mathbf{p}+\mathbf{q}}^\dagger + B_\mathbf{p} B_{\mathbf{p}+\mathbf{q}}^\dagger) f_\mathbf{k} + \frac{g\mu_B H_0}{2} \sum_\mathbf{q} (A_\mathbf{q}^\dagger A_\mathbf{q} - B_\mathbf{q}^\dagger B_\mathbf{q}). \tag{49}$$

The susceptibility has the form

$$\chi(T) = -\frac{1}{V}\frac{\partial^2 \mathcal{F}}{\partial H_0^2} = \frac{\beta}{V} \left(\frac{g\mu_B}{2} \right)^2 \left\{ \left\langle \left[\sum_\mathbf{q} (A_\mathbf{q}^\dagger A_\mathbf{q} - B_\mathbf{q}^\dagger B_\mathbf{q}) \right]^2 \right\rangle - \left\langle \sum_\mathbf{q} (A_\mathbf{q}^\dagger A_\mathbf{q} - B_\mathbf{q}^\dagger B_\mathbf{q}) \right\rangle^2 \right\}, \tag{50}$$

where $\mathcal{F}$ is the free energy. Note that

$$\langle A_\mathbf{q}^\dagger A_\mathbf{q} A_\mathbf{q}^\dagger A_\mathbf{q} \rangle - \langle A_\mathbf{q}^\dagger A_\mathbf{q} \rangle^2 = -\frac{1}{\beta}\frac{\partial n_\mathbf{p}}{\partial \omega_\mathbf{p}} \tag{51}$$

Figure 2. $\Delta H_{pp}[\chi(T)/\chi_0(T)]$ against T/T_C for $F = G = 2\,\mathrm{K}$, where $\chi(T)$ and $\chi_0(T)$ are the susceptibilities of the double-exchange system and isolated spins, respectively.

and

$$\langle A_{\mathbf{q}}^{\dagger} A_{\mathbf{q}} B_{\mathbf{q}}^{\dagger} B_{\mathbf{q}} \rangle = \langle A_{\mathbf{q}}^{\dagger} A_{\mathbf{q}} (2S - x - A_{\mathbf{q}}^{\dagger} A_{\mathbf{q}}) \rangle^2. \tag{52}$$

As a result, we obtained

$$\chi(T) = -(g\mu_{\mathrm{B}})^2 \sum_{\mathbf{q}} \frac{\partial n_{\mathbf{p}}}{\partial \omega_{\mathbf{p}}}$$

$$= \frac{(g\mu_{\mathrm{B}})^2}{8\pi^2 D} \sqrt{\frac{\pi k_B T}{D}} g_{1/2}(z), \tag{53}$$

where $g_{1/2}(z)$ was defined in equation (38) and z, the fugacity, can be evaluated with equation (40). Thus we can plot $\Delta H_{pp}[\chi(T)/\chi_0(T)]$ against T/T_C. In figure 2, we see that there is still reminiscent temperature dependence away from T_C, even though $\chi(T)/\chi_0(T)$ is a monotonically decreasing function of T. The reason, we think, is that the constraints (19) exist throughout the temperature range. Through equations (39) and (40), their presence affected the value of fugacity, which, in turn, affected the temperature dependence of ΔH_{pp}.

Perhaps our most important result is that $1/T_2$ is a universal function of T/T_C. That is to say, the linewidth multiplied by T_C plotted against T/T_C from different systems, lies on the same curve. The doping concentration x affected T_2 only through T_C. In practice, it may be more complicated. SE interaction and Jahn–Teller distortion will affect T_C and they are not accounted for by our calculation. However, there is experimental evidence for the universal behaviour. In [13], $\Delta H_{pp}(T)/\Delta H_{pp}(\infty)$ versus T/T_C from various systems were plotted and they lie on a same line. In our calculation, we do not have a $\Delta H_{pp}(\infty)$. However, if we divide $\Delta H_{pp}(T)$ or $1/T_2$ by a ΔH_{pp} at a sufficiently high temperature, the factor $(F^2 + G^2)/k_B T_C$ will be cancelled and a universal curve will appear.

Acknowledgment

Appendix

Here, we consider the contribution of the self-energy of boson propagators to the computation of equation (30). Applying the Matsubara formalism [40], we change the summation into integration:

$$\frac{1}{\beta} \sum_{m,\mathbf{p},\mathbf{q}} D(i\omega_m, \mathbf{p}) D(i\omega_n + i\omega_m, \mathbf{q}) = -\frac{1}{2\pi i} \sum_{\mathbf{p},\mathbf{q}} \int_C dz \, D(z, \mathbf{p}) D(z + i\omega_n, \mathbf{q}) n(z), \qquad (A.1)$$

where contour C encloses all the poles except those of $n(z)$. Owing to the self-energy, the propagator has a continuous distribution of poles. They form a branch cut. The contour can now be deformed so that the integration paths are along each side of the cuts:

$$\frac{1}{\beta} \sum_{m,\mathbf{p},\mathbf{q}} D(i\omega_m, \mathbf{p}) D(i\omega_n + i\omega_m, \mathbf{q})$$

$$= -\frac{1}{2\pi i} \sum_{\mathbf{p},\mathbf{q}} \int dz \, n(z) \{ D(z, \mathbf{p}) [D(z + i\omega_n + i\delta, \mathbf{q}) - D(z + i\omega_n - i\delta, \mathbf{q})]$$
$$+ D(z + i\omega_n, \mathbf{q}) [D(z + i\delta, \mathbf{p}) - D(z - i\delta, \mathbf{p})]. \qquad (A.2)$$

Using the properties of propagators, we have

$$\frac{1}{\beta} \sum_{m,\mathbf{p},\mathbf{q}} D(i\omega_m, \mathbf{p}) D(i\omega_n + i\omega_m, \mathbf{q})$$

$$= -\frac{1}{2} \sum_{\mathbf{p},\mathbf{q}} \int dz \, n(z) [D(z - i\omega_n, \mathbf{p}) + D(z + i\omega_n, \mathbf{p})] \operatorname{Im} \mathbf{D}(z, \mathbf{q}). \qquad (A.3)$$

When computing $\operatorname{Re} \phi$ from $\operatorname{Im} \psi$, we need to take the derivative with respect to ω_n and then set ω_n to 0 (see equation (11)),

$$\frac{1}{i\beta} \frac{\partial}{\partial \omega_n} \sum_{m,\mathbf{p},\mathbf{q}} D(i\omega_m, \mathbf{p}) D(i\omega_n + i\omega_m, \mathbf{q})$$

$$= \frac{1}{2} \int dz [D(z + i\omega_n, \mathbf{p}) - D(z - i\omega_n, \mathbf{p})] \frac{\partial}{\partial z} [\operatorname{Im} \mathbf{D}(z, \mathbf{q}) n(z)], \qquad (A.4)$$

where integration by parts and interchanging of $\mathbf{p}$ and $\mathbf{q}$ were made. By setting ω_n equal to zero, the right hand side of equation (A.4) becomes

$$i\pi \sum_{\mathbf{p},\mathbf{q}} \int dz \operatorname{Im} D(z, \mathbf{p}) \frac{\partial}{\partial z} [\operatorname{Im} \mathbf{D}(z, \mathbf{q}) n(z)]$$

$$= \frac{i\pi}{2} \sum_{\mathbf{p},\mathbf{q}} \int dz \operatorname{Im} D(z, \mathbf{p}) \operatorname{Im} \mathbf{D}(z, \mathbf{q}) \frac{\partial \mathbf{n(z)}}{\partial \mathbf{z}} \qquad (A.5)$$

and, thus, gives the factor $\partial n(z)/\partial z$. The vertex correction usually gives a multiplying factor and, hence, is unlikely to neutralize it.

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