High-Temperature Electron Paramagnetic Resonance in Magnets with the Dzyaloshinskii-Moriya Interaction

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We analyze the high-temperature electron paramagnetic resonance (EPR) absorption in a weakly anisotropic Heisenberg magnet having two distinct types of anisotropy, represented, respectively, by a symmetric term and the Dzyaloshinskii-Moriya (DM) term. Contrary to the widespread opinion that the latter is responsible for the excessive linewidth observed in the EPR spectra of many oxides, we prove that its contribution to the linewidth is only of the same level as that of the symmetric anisotropy. This gives a solution to the long-standing controversial problem of the high-temperature magnetic relaxation in quantum-spin systems with the DM interaction.

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The electron paramagnetic resonance (EPR) technique is known to be a very powerful and useful tool in probing the magnetic interactions in solids. Recently, due to growing interest in quantum-spin systems, many of the well-known 1D compounds such as $CuGeO_3$ and NaV_2O_5 have been the subject of intensive investigations by this method [1–14]. However, a clear lack of a comprehensive EPR theory, as far as low-dimensional compounds are concerned, often makes the interpretation of the experimental data somewhat difficult. Paradoxically, the theoretical situation is more developed at low than at high temperatures. Very recently, Oshikawa and Affleck [15] proposed a new approach to the low-T EPR based on the bosonization and the Feynman-Dyson self-energy formalism. This paved the way to a new interpretation of the old EPR data obtained on 1D antiferromagnet (AFM) copper benzoate. As for the high-T EPR theory, the pioneering work of Kubo and Tomita [16], further developed by Mori [17], remains the most used at present.

In an EPR experiment, the magnetic field is applied in, say, the z direction, $H_0\mathbf{z}$, and the microwave field $h(t)\mathbf{x}$ is perpendicular to it. If the spin Hamiltonian $\mathcal{H}$ contains only two terms—the Zeeman interaction $\mathcal{H}_Z = \omega_0 S^z$ ($\omega_0 = H_0$) and the exchange energy $\mathcal{H}_{ex} = \Sigma J \mathbf{S}_l \mathbf{S}_{l+1}$—then, in the absence of anisotropy ($\mathcal{H}' = 0$), the spin dynamics would be described by only one mode $S^+ = (S^-)^+ : i\dot{S}^+ = -\omega_0 S^+$. The spins precess around H_0 with the Larmor frequency, and the uniform microwave field $h(t)$ acting on $S^+(S^-)$ causes a microwave absorption. It is easy to see that, in this particular case, the EPR signal is simply a sum of two nonshifted delta functions $\delta(\omega \pm \omega_0)$. This fact is often used as the starting point of an EPR theory. It shows also that the line broadening and the line shift are both due to the magnetic anisotropy. If now $\mathcal{H}' \neq 0$ and $\mathcal{H}' \ll \mathcal{H}_{ex}$, then the fluctuating torques are also nonzero, $[S^+, \mathcal{H}'] \neq 0$, and the S^+ mode gains a finite width. The Kubo theory predicts, in this case, a Lorentzian shape of the absorption, with a linewidth $\Delta\omega = \mathrm{Re}\int_0^\infty \psi(t)\,dt$, where $\psi(t) = \langle [\mathcal{H}', S^+](t), [S^-, \mathcal{H}'] \rangle / \langle S^+, S^- \rangle$ is a sum of *four-spin correlation functions* called the memory function. Generally, $\psi(t)$ is difficult to calculate, and, thus, one is forced to make some assumptions concerning its behavior. In the exchange narrowing regime, $\psi(t)$ is assumed to have a simple form, $\psi(t) \sim \mu_2 \delta(t)/J$ [16], resulting in a linewidth $\Delta\omega = \mu_2/J$ where μ_2 is the second moment of the absorption signal.

When the anisotropy is defined mostly by the symmetric quadratic form $dS_l^\alpha S_{l+1}^\alpha$, which represents the dipole-dipole or/and anisotropic exchange interactions, the situation has already been widely discussed in the case of low-dimensional magnets in the early 1970s. The experimental results on TMMC, CMC, and other 1D spin-5/2 Heisenberg AFM have been successfully interpreted in the framework of the Kubo approach modified by the spin-diffusion concept [18].

It appears, however, that the magnetic relaxation in new spin-1/2 oxides is a more complicated matter. One of the reasons is that these systems are not made of S ions, as in the case of TMMC. Thus, the terms such as the Dzyaloshinskii-Moriya (DM) interaction, $\mathbf{D} \cdot (\mathbf{S}_l \times \mathbf{S}_{l+1})$, should be taken into account.

Starting from the first EPR experiments on $CuGeO_3$ and NaV_2O_5, a considerable linewidth, $\Delta\omega$, much larger than one can expect from the Kubo formula supposing only the symmetric term $\Delta\omega \sim \mu_{2d}/J \sim d^2/J$, has been observed at high temperatures. This was then explained by Yamada *et al.* [14] who argued that, if one takes standard estimates for the DM term, $D \sim (\Delta g/g)J$, and the symmetric one, $d \sim (\Delta g/g)^2 J$ (where g and Δg are the g factor and its anisotropy, respectively), then the DM term gives a major contribution to the observed linewidth $\Delta\omega \sim \mu_{2D}/J \sim D^2/J$. Notice, however, that the existence of the DM interaction in the spin Hamiltonian of these compounds

is still the subject of considerable controversy (see, for example, [13]).

As we show in this Letter, the theory of exchange narrowing is utterly inadequate for describing the DM effects on the EPR line shape. Indeed, if one considers the most interesting case of a spin chain with a staggered DM term such that the orientation of the DM vector is site dependent, $\mathbf{D}_l = (-1)^l \mathbf{D}$, then, with the help of the relation

$$[S^a, \mathcal{H}_D] = -(1/2J) D^c \varepsilon^{acd} [S^d_\pi, \mathcal{H}_{ex}], \qquad (1)$$

it can be shown that $\psi(t)$ is driven exclusively by the staggered fluctuations, $\psi_D(t) \sim (d^2/dt^2)\langle S^+_\pi(t), S^-_\pi\rangle$, and the resulting linewidth is simply zero up to second order of D/J

$$\Delta\omega = \mathrm{Re}\int_0^\infty \psi_D(t)\, dt = 0, \qquad (2)$$

while the exchange narrowing formula gives $\Delta\omega \sim D^2/J$ [14]. Therefore, it remains a challenging problem to elucidate the role that the DM interaction plays in the EPR absorption without making any assumption about the memory function behavior.

In this Letter we realize this program. First, using a unitary transformation, we map the initial chain onto an anisotropic Heisenberg *XXZ* chain. Then, after evaluating the EPR absorption, we conclude that the effective magnetic anisotropy can always be presented in the form $\tilde{d} \backsimeq d - D^2/(2J)$. Hence, the DM interaction contributes to $\Delta\omega$ at a level of the symmetric anisotropy. On the other hand, computing the linewidth perturbatively up to second order of D/J, we show that the width appears only in the next order of the perturbation, $\Delta\omega \sim (D^2/J)(D^2/J^2)$.

Let us start to treat the DM problem by performing a unitary transformation in such a way that the transformed Hamiltonian contains only a *symmetric anisotropy of order* D^2/J [19]. We shall see that the DM contribution to the linewidth is then much simpler to analyze.

We consider the following spin chain Hamiltonian:

$$\mathcal{H} = \sum_l [J\mathbf{S}_l\mathbf{S}_{l+1} + (-1)^l \mathbf{S}_l \cdot \hat{\mathbf{T}}_1 \cdot \mathbf{S}_{l+1} + \mathbf{S}_l \cdot \hat{\mathbf{T}}_2 \cdot \mathbf{S}_{l+1}], \qquad (3)$$

where $T^{ab}_1 = D\varepsilon^{abc}v^c$ corresponds to the DM interaction (with $a, b, c = x, y, z$), while $T^{ab}_2 = d(v^a v^b - \frac{1}{3}\delta^{ab})$ is a symmetric anisotropy of strength d, the anisotropy direction being given by the unit vector $\mathbf{v}$. The Hamiltonian can be cast under the form

$$\mathcal{H} = \sum_{l \text{ even}} \mathbf{S}_l \cdot \hat{\mathbf{T}} \cdot (\mathbf{S}_{l+1} + \mathbf{S}_{l-1}), \qquad (4)$$

with $\hat{\mathbf{T}} = \hat{\mathbf{1}} \cdot J + \hat{\mathbf{T}}_1 + \hat{\mathbf{T}}_2$. Such a T matrix is decomposed in its polar form,

$$\hat{\mathbf{T}} = R\hat{\mathbf{M}}, \qquad (5)$$

where R is a rotation given by

$$R^{ab} = v^a v^b + \cos\varphi(\delta^{ab} - v^a v^b) + \sin\varphi\, \varepsilon^{abc} v^c,$$

with $\cos\varphi = 1/\sqrt{1 + D^{*2}}$ and $D^* = D/(J - d/3)$, while $\hat{\mathbf{M}}$ is the symmetric matrix

$$M^{ab} = \left(J + \frac{2d}{3}\right) v^a v^b + \sqrt{\left(J - \frac{d}{3}\right)^2 + D^2} \times (\delta^{ab} - v^a v^b).$$

Inserting the decomposition (5) into (4) and performing the canonical transformation $\mathbf{U}$ defined by

$$\begin{aligned} \mathbf{U}\mathbf{S}_l\mathbf{U}^+ &= R\mathbf{S}_l, \qquad \text{for } l \text{ even}, \\ \mathbf{U}\mathbf{S}_l\mathbf{U}^+ &= \mathbf{S}_l, \qquad \text{for } l \text{ odd}, \end{aligned} \qquad (6)$$

one then obtains a transformed Hamiltonian $\tilde{\mathcal{H}} = \mathbf{U}\mathcal{H}\mathbf{U}^+$ given by

$$\tilde{\mathcal{H}} = \sum_l \mathbf{S}_l \cdot \hat{\mathbf{M}} \cdot \mathbf{S}_{l+1}.$$

This Hamiltonian corresponds to a system with a symmetric anisotropy of strength $\tilde{d}$. In the particular case $\tilde{d} = 0$, we end up with a transformed Hamiltonian without any anisotropy. Notice that, when defining (6), we supposed that $\mathbf{v}$ is arbitrarily oriented with respect to the magnetic field H_0. Therefore, the unitary transformation (6) is an extension of the one presented in [19] and appears to be very useful in the angular dependence analysis of EPR absorption spectra.

The EPR absorption, within the Mori formalism [17], is given by

$$I(\omega) \propto \beta\omega^2 \mathrm{Im}\left\langle S^x, \frac{1}{\omega - L_{\mathcal{H}} - L_Z} S^x \right\rangle_{\mathcal{H}+\mathcal{H}_z}, \qquad (7)$$

$L_{\mathcal{H}}$ and L_Z being the Liouville operators associated with $\mathcal{H}$ and $\mathcal{H}_Z$, respectively, and $\langle A, B\rangle = \int_0^1 dx \times \langle Ae^{-x\beta H} B e^{x\beta H}\rangle_{\mathcal{H}}$. In order to simplify the discussion, we assume that the applied magnetic field H_0 is in the anisotropy direction $\mathbf{v}$, say the z direction. So, by introducing the canonical transformation and noticing that the Zeeman Hamiltonian is invariant under this transformation, one is led to

$$I(\omega) = \frac{1 + \cos\varphi}{2} \tilde{I}(0, \omega) + \frac{1 - \cos\varphi}{2} \tilde{I}(\pi, \omega), \qquad (8)$$

where $\tilde{I}(q, \omega)$ is defined as

$$\tilde{I}(q, \omega) \propto \beta\omega^2 \mathrm{Im}\left\langle S^x_q, \frac{1}{\omega - L_{\tilde{\mathcal{H}}} - L_Z} S^x_{-q} \right\rangle_{\tilde{\mathcal{H}}+\mathcal{H}_z}.$$

We readily see that the EPR absorption we are computing is essentially given by the absorption at $q = 0$ of the transformed system which contains a symmetric anisotropy $\tilde{d}$. To be more specific, let us consider the particular case $\tilde{d} = 0$. Then the transformed system is isotropic ($\tilde{\mathcal{H}} = \mathcal{H}_{ex}$), and we know that $\mathcal{H}_{ex} + \mathcal{H}_Z$ possesses a proper mode

S^+. By the inverse transformation, we deduce that the original system ($\mathcal{H} + \mathcal{H}_Z$) has also a proper mode S_n:

$$S_n = \tfrac{1}{2}(e^{i\varphi} + 1)S^+ - \tfrac{1}{2}(e^{i\varphi} - 1)S^+_\pi,$$

which is, of course, mainly the S^+ mode. Therefore, one has also to introduce another mode,

$$S_b = \tfrac{1}{2}(e^{i\varphi} - 1)S^+ + \tfrac{1}{2}(e^{i\varphi} + 1)S^+_\pi,$$

which is mainly the S^+_π mode. Since the S_n mode is a proper one, it will give a Dirac peak in the EPR signal, while the S_b mode, which is not a proper one, will give a signal which is strongly temperature dependent. That explains why the EPR absorption (8) is given by

$$I(\omega) \propto \beta\omega^2\left\{\frac{1+\cos\varphi}{2}\,\frac{\pi}{4}\,[\delta(\omega-\omega_0)+\delta(\omega+\omega_0)]\sigma^{+-} - \frac{1-\cos\varphi}{8}\,\mathrm{Im}[\Sigma^{+-}(\pi,\omega-\omega_0)+\Sigma^{-+}(\pi,\omega+\omega_0)]\right\}, \tag{9}$$

where $\sigma^{+-} = \langle S^+, S^-\rangle_{\mathcal{H}_{ex}+\mathcal{H}_Z}$ is the static correlation function and $\Sigma^{+-}(\pi,\omega)$ is the dynamic correlation function governed by the sum $\mathcal{H}_{ex} + \mathcal{H}_Z$. We see that the main term is the first one, corresponding to the response of an isotropic system, while the second one, with the weight $\backsim(D/J)^2$, is essentially constant for $\omega \pm \omega_0 \ll J$, as it has been emphasized in [20]. As a result, the system behaves then essentially as an isotropic one. In the more general case ($\tilde{d} \sim D^2/J$), again the EPR absorption $I(\omega)$ is essentially driven by the absorption $\tilde{I}(\omega)$ of a system with a symmetric anisotropy of strength D^2/J. Hence, we see that the Dzyaloshinskii-Moriya interaction cannot lead to a linewidth much larger than the linewidth one would get with a symmetric anisotropy. On the contrary, we have proven that they are of the same order of magnitude.

One also should note that Eq. (9) illustrates a remarkable difference between the symmetric and the DM anisotropies as regards the spin dynamics. It is clear from (9) that in the presence of the DM interaction the spin dynamics of a weakly anisotropic Heisenberg magnet is governed by two modes. The uniform microwave field $h(t)$ acts on a uniform variable $S^+(t)$ but, through the DM term, it appears to be coupled to the staggered one $S^+_\pi(t)$. If now one would like to estimate the linewidth of the above two modes by the method of moments, the S_n and S_b operators should be used instead of S^+ and S^+_π. Specifically, the second moment of the S_n mode is given by $\mu_{2S_n} = \mathrm{Tr}[S_n, \mathcal{H}]^2/\mathrm{Tr}S_n^2 \backsimeq \tilde{d}^2$ and, quite in a similar manner, $\mu_{2S_b} = \mathrm{Tr}[S_b, \mathcal{H}]^2/\mathrm{Tr}S_b^2 \backsimeq J^2$. These results are in agreement with the above made estimates.

It is then worthwhile comparing the exact results with the usual approach for a pure DM interaction. Let us recall that the EPR absorption (7) at high temperature takes the form

$$I(\omega) \propto \beta\omega^2\,\mathrm{Im}\left\{\frac{1}{\omega+\omega_0-\Gamma^{-+}(\omega)} + \frac{1}{\omega-\omega_0-\Gamma^{+-}(\omega)}\right\},$$

where $\Gamma^{+-}(\omega)$ ($\Gamma^{-+}(\omega)$) is the self-energy (or the memory spectrum). Again, for the sake of simplicity, we consider the case where H_0 and D are both in the z direction and we neglect a small shift of the resonance frequency ω_0. As usual, the self-energy, $\Gamma^{+-}(\omega) = \gamma^{+-}(\omega)/\sigma^{+-}$, which is the Laplace transform of the memory function $i\psi(t)$, is evaluated up to second order of perturbation theory, leading to

$$\gamma^{+-}(\omega) = \left\langle A^+, \frac{1}{\omega - L_{\mathcal{H}_{ex}} - L_Z + i\varepsilon}\,A^-\right\rangle_{\mathcal{H}_{ex}}, \tag{10}$$

where $A^a = -i[S^a, \mathcal{H}_D]$ is quadratic in the spin operators, making $\gamma^{+-}(\omega)$ a four-spin correlation function. The standard evaluation of such a function consists of a decoupling procedure expressing $\gamma^{+-}(t)$ as the product of two-spin correlation functions for which some exponential decay behavior is made. Computing A^a by the use of (1) and inserting this result into (10), we arrive at one of the most significant of our conclusions, that the self-energy $\Gamma^{+-}(\omega)$, in the case of the DM interaction, depends on *the unique dynamic two-spin correlation function,*

$$G(\omega) = \left\langle S^+_\pi, \frac{L^2_{\mathcal{H}_{ex}}}{\omega - L_{\mathcal{H}_{ex}} + i\varepsilon}\,S^-_\pi\right\rangle_{\mathcal{H}_{ex}},$$

which reduces to $G(\omega) = -\omega\sigma^{+-} + \omega^2\Sigma^{+-}(\pi,\omega)$. The "static part" of $\gamma^{+-}(\omega) = (D/2J)^2G(\omega)/2$ will produce a shift in the resonance frequency, while the dynamic part is essentially purely imaginary and therefore responsible for the linewidth. If the EPR signal is supposed to be a Lorentzian, then, in accordance with (2), one obtains $\Delta\omega = \mathrm{Im}\Gamma^{+-}(0) = 0$ up to second order of D/J. However, the linewidth can be approximately defined as $(D^2/J)\cdot(D^2/J^2)$, which is of the order of a symmetric anisotropy, in agreement with the exact result (9). This result is worthy of further comments. First, it clearly shows how dangerous is the above-mentioned decoupling procedure for $\psi(t)$, since it leads to a finite density of the memory spectrum $\Gamma^{+-}(\omega \to 0) \sim \mu_2/J$. Second, it rules out completely the possibility of spin diffusion, or more generally the possibility of the power-law long-time behavior of $\psi(t)$, when the magnetic relaxation is purely governed by the DM interaction. As we have already pointed out, $\Gamma^{+-}(\omega)$, in this case, is driven exclusively by the staggered spin fluctuations, so that $\mathrm{Im}\Gamma^{+-}(\omega) \sim G(\omega) \sim \omega^2\Sigma^{+-}(\pi,\omega)$. Using the results of [20], one can safely conclude that $\Sigma^{+-}(\pi,\omega \to 0)$ is finite at high T, indicating that $\psi(t)$ decays quite rapidly, on the scale of $t \sim 1/J$. This differs fundamentally

from the long-time behavior of $\psi(t) \sim t^{-\alpha}$ suggested in the case of the symmetric anisotropy [18]. Third, although our results, obtained by a perturbation theory, concern a pure DM anisotropy, it is easy to check that, in a more general case (when $d \neq 0$), we again arrive at the conclusion that if $d \approx D^2/(2J)$ then $\tilde{d} = 0$ and the system behaves as an isotropic one with $\Delta\omega = 0$. This situation ($\tilde{d} = 0$) which, at first sight, seems totally artificial, holds, in fact, as it was pointed out by Kaplan, Shekhtman, Entin-Wohlman, and Aharony (KSEA) [21], for a very wide class of spin-1/2 1D and 2D cuprates. KSEA have shown, on rather general grounds, that in these materials the symmetric term d is always accompanied by the DM anisotropy D leading to an isotropic behavior $\tilde{d} = 0$. Recently, the KSEA predictions have been established experimentally in various studies, including the ESR measurements on the spin-1/2 2D tetragonal cuprate $Sr_2CuO_2Cl_2$ [22]. Since our conclusions for $\tilde{d} = 0$ remain also valid in 2D magnets at moderate temperatures, it is interesting to note that the experimental linewidth in $Sr_2CuO_2Cl_2$ at $T = 70$ K is surprisingly small, $\Delta\omega/J \approx 2 \times 10^{-5}$, as compared with the estimate $\Delta\omega/J \sim (D/J)^2 \approx 10^{-2}$. We speculate therefore that this is a first but clear experimental evidence in favor of our predictions.

In conclusion, we have shown that, contrary to what is usually suggested in the literature, the DM interaction cannot be at the origin of the excessive linewidth observed in the ESR spectra of many oxides. Its contribution is of the same order as that of the symmetric anisotropy. In the case of an exact compensation ($\tilde{d} = 0$), the system behaves as an isotropic one. Although we do not offer an explanation for the large linewidth in $CuGeO_3$ and NaV_2O_5, our results clearly suggest that a revision of the previous experimental studies is needed. Finally, the memory function can be expressed in terms of a unique two-spin correlation function. This rules out the spin diffusion scenario in the case of a pure DM anisotropy.

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