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Temperature dependence of the width and the frequency of the EPR lines in a one-dimensional Heisenberg antiferromagnet with a Dzyaloshinsky–Moriya antisymmetric exchange interaction as a main perturbation term

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Abstract. On the basis of a classical model of spins, we discuss the temperature dependence of both the width and the resonance frequency of the EPR lines in a one-dimensional Heisenberg antiferromagnet with a Dzyaloshinsky–Moriya antisymmetric exchange interaction as a main perturbation term. We find that the width does not grow with decreasing temperature toward T_N, while the resonance frequency is independent of temperature. These results are completely different from those for a system with symmetric perturbation terms, such as the dipolar, anisotropic exchange and single-ion anisotropy terms; in such systems, with decreasing temperature toward T_N the linewidth broadens and diverges, while the resonance frequency goes up or down according to the direction of an external field with respect to the chain axis.

1. Introduction

Electron paramagnetic resonance (EPR) is a useful method for the investigation of spin dynamics, especially for low-dimensional Heisenberg magnets. As is well known, the width and resonance frequency of the EPR lines in most one-dimensional Heisenberg antiferromagnets (1DHAFs) show remarkable changes with the development of short-range order over a wide range of temperature T above T_N. That is, the linewidth $\Delta\omega$ increases over the short-range-order region and diverges when T approaches T_N, which has been found in many 1DHAFs as reviewed by Drumheller [1], while the resonance frequency ω goes up or down according to the direction of the external field $\boldsymbol{H}$ with respect to the chain axis. That is, ω for $\boldsymbol{H}$ parallel to the chain axis (denoted $\omega_\parallel$) increases with decreasing T toward T_N, while ω for $\boldsymbol{H}$ perpendicular to the chain axis (denoted $\omega_\perp$) decreases; these two frequencies have a relation $(\omega_\parallel \cdot \omega_\perp^2)^{1/3}$ = constant. Such a dependence of ω on T was pointed out theoretically [2, 3] and was confirmed in several 1DHAFs such as TMMC and $CsMnCl_3{\cdot}2H_2O$ [2, 4]. This behaviour of $\Delta\omega(T)$ and $\omega(T)$ is common in systems with symmetric perturbation terms, such as the dipolar, anisotropic exchange interaction terms, and has been established both theoretically and experimentally.

When an antisymmetric perturbation term such as a Dzyaloshinsky–Moriya (DM) interaction overwhelms the symmetric terms mentioned above, how will $\Delta\omega$ and ω behave with the change of T toward T_N? The purpose of the present study is to clarify this issue for a 1DHAF. The DM perturbation has been shown to play a unique role in both static and dynamic magnetic properties [5–7]. Its role in EPR lines has also been investigated intensively [8–11]. The results which will be developed in the successive sections will reveal a new aspect of the role of the DM perturbation in EPR lines of 1DHAFs. We have treated the classical Heisenberg model and have clarified that the dependence of both $\Delta\omega$ and ω on T over the short-range-order region is completely different from that in a system with the symmetric perturbation term mentioned above.

2. The EPR linewidth

We take the Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{Z}} + \mathcal{H}' \tag{1}$$

which consists of the exchange between the nearest-neighbour spins, the Zeeman and the perturbation terms, respectively. For a 1DHAF with the DM interaction, we have

$$\mathcal{H}_{\text{ex}} = -2J \sum_n \boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1} \tag{2}$$

$$\mathcal{H}_{\text{Z}} = -\mu_{\text{B}} \sum_n \boldsymbol{S}_n \cdot \mathbf{g} \cdot \boldsymbol{H} \tag{3}$$

and for $\mathcal{H}'$ we take

$$\mathcal{H}' = \sum_n \boldsymbol{d}_{n,n+1} \cdot (\boldsymbol{S} \times \boldsymbol{S}_{n+1}) \tag{4a}$$

or

$$\mathcal{H}' = \sum_n (-1)^n \boldsymbol{d}_{n,n+1} \cdot (\boldsymbol{S}_n \times \boldsymbol{S}_{n+1}). \tag{4b}$$

The expression (4a) for $\mathcal{H}'$ indicates that all the DM vectors $\boldsymbol{d}_{n,n+1}$ are in the same direction, i.e. $\boldsymbol{d}_{n-1,n} = \boldsymbol{d}_{n,n+1}$, while that of (4$b$) indicates that the DM vectors change their direction alternately, i.e. $\boldsymbol{d}_{n-1,n} = -\boldsymbol{d}_{n,n+1}$. Let us take the coordinates $[x, y, z]$ as $z \|$ chain axis and we treat both $\boldsymbol{d}_{n,n+1} \| z$ and $\boldsymbol{d}_{n,n+1} \perp z$.

When the decay of the spin correlation is rapid such as for the Gaussian process, we can treat the linewidth $\Delta\omega$ following Van Vleck's theory [12] as

$$\Delta\omega = \sqrt{\pi/2}\sqrt{(M_2)^3/M_4} \tag{5}$$

in which M_2 and M_4 are the second and the fourth moments, respectively. They are expressed as

$$M_2 = \langle [\mathcal{H}', S^+][S^-, \mathcal{H}'] \rangle / \hbar^2 \langle S^+ S^- \rangle \tag{6}$$

and

$$M_4 = \langle [\mathcal{H}_{\text{ex}}, [\mathcal{H}', S^+]][[S^-, \mathcal{H}'], \mathcal{H}_{\text{ex}}] \rangle / \hbar^4 \langle S^+ S^- \rangle \tag{7}$$

where $S^{\pm}$ are the transverse components of the total spin $\boldsymbol{S} = \Sigma_n \boldsymbol{S}_n$. To calculate these two moments, we assume the classical model of spins, i.e. we replace a spin $\boldsymbol{S}$ by a classical vector $\boldsymbol{s} = \boldsymbol{S}/\sqrt{S(S+1)}$. In the calculation of the thermal average $\langle \ldots \rangle$ we used an approximation $\mathrm{Tr}(\exp(-\mathcal{H}/k_{\mathrm{B}}T)(\ldots)) \simeq \mathrm{Tr}(\exp(-\mathcal{H}_{\mathrm{ex}}/k_{\mathrm{B}}T)(\ldots))$. Following Fisher's classical spin model [13] which was used for the case of symmetric perturbation [2, 3, 14], we obtain

$$M_2 = (1/3\hbar^2)d^2S(S+1)A(\theta)f^{\mathrm{DM}}(K) \tag{8}$$

for both (4*a*) and (4*b*), while

$$M_4 = (16/9\hbar^4)d^2J^2[S(S+1)]^2A(\theta)g_{\mathrm{a}}^{\mathrm{DM}}(K) \qquad \text{for equation (4}a\text{)} \tag{9a}$$

and

$$M_4 = (64/9\hbar^4)d^2J^2[S(S+1)]^2A(\theta)g_{\mathrm{b}}^{\mathrm{DM}}(K) \qquad \text{for equation (4}b\text{)} \tag{9b}$$

where $A(\theta)$ is the angular contribution arising from the angle θ between $\boldsymbol{H}$ and the chain axis and is expressed as

$$A(\theta) = \begin{cases} 1 + \cos^2\theta & \text{for } \boldsymbol{d}_{n,n+1} \parallel z \\ 1 + \frac{1}{2}\sin^2\theta & \text{for } \boldsymbol{d}_{n,n+1} \perp z. \end{cases} \tag{10}$$

The functions $f^{\mathrm{DM}}(K)$, $g_{\mathrm{a}}^{\mathrm{DM}}(K)$ and $g_{\mathrm{b}}^{\mathrm{DM}}(K)$ represent the temperature-dependent parts normalised at $T \to \infty$; Fisher's classical spin model [13] leads to the following expression for $f^{\mathrm{DM}}(K)$:

$$f^{\mathrm{DM}}(K) = (3u(K)/K)(1 - u(K))/(1 + u(K)) \tag{11}$$

while for $g_{\mathrm{a}}^{\mathrm{DM}}(K)$ and $g_{\mathrm{b}}^{\mathrm{DM}}(K)$ it leads to

$$\begin{aligned} g_{\mathrm{a}}^{\mathrm{DM}}(K) = {} & [(1 - u(K))/(1 + u(K))][(3v(K)/2K)(1 + u(K))(3 + u(K)) \\ & + (3u(K)/K)(1 - 2u(K) - 3v(K))] \end{aligned} \tag{12a}$$

and

$$\begin{aligned} g_{\mathrm{b}}^{\mathrm{DM}}(K) = {} & [(1 - u(K))/(1 + u(K))][(3v(K)/8K)(1 + u(K))\,(3 + u(K)) \\ & + (3u(K)/K)(1 + u(K))] \end{aligned} \tag{12b}$$

in which

$$K = 2JS(S+1)/k_{\mathrm{B}}T \qquad u(K) = \coth K - 1/K \qquad v(K) = 1 - 3u(K)/K.$$

We show in figure 1 the normalised linewidth $\Delta\omega^{\mathrm{DM}}(T)/\Delta\omega^{\mathrm{DM}}(T \to \infty)$. To compare the present results with that of the dipolar perturbation, we have also calculated $\Delta\omega^{\mathrm{dd}}(T)/\Delta\omega^{\mathrm{dd}}(T \to \infty)$ using the formula given in [14]. We find that $\Delta\omega^{\mathrm{DM}}(T)$ does not increase with decreasing T toward T_{N}.

3. The resonance frequency

When the Zeeman energy is much larger than that of the perturbation term, the resonance frequency ω is given [1] as

$$\hbar\omega = \langle [S^-, [S^+, \mathcal{H}]] \rangle / 2\langle S^z \rangle \tag{13}$$

where $S^{\pm}$ and S^z are the transverse and z components of the total spin $\boldsymbol{S} = \Sigma_n \boldsymbol{S}_n$

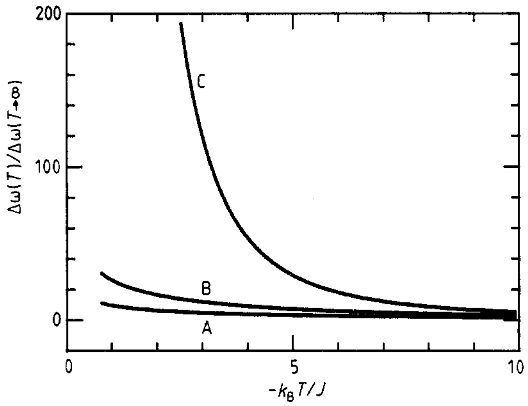

Figure 1. The EPR linewidth normalised at high temperatures is shown as a function of the normalised temperature $-k_BT/J$ $(J<0)$ for $S=\frac{5}{2}$. Curves A and B are for $d_{n-1,n}=d_{n,n+1}$ and $d_{n-1,n}=-d_{n,n+1}$, respectively. To make a comparison with a symmetric perturbation case, the linewidth in a one-dimensional Heisenberg antiferromagnet in which the dipolar interaction plays a role in the main perturbation term is shown as curve C. The antiferromagnetic transition temperature T_N is tentatively taken as 0 K.

and $\langle \ldots \rangle$ indicates the statistical average. When $d_{n,n+1} \parallel z$, we obtain the resonance frequencies for both $H \parallel z$ and $H \parallel x$ as

$$\hbar\omega_{\parallel} = g_{\parallel}\mu_B H - \frac{1}{\langle S^z \rangle}\sum_n d^z_{n,n+1}\langle S^x_n S^y_{n+1} - S^y_n S^x_{n+1}\rangle \tag{14a}$$

and

$$\hbar\omega_{\perp} = g_{\perp}\mu_B H + \frac{1}{2\langle S^x \rangle}\sum_n d^z_{n,n+1}\langle (S^x_n S^y_{n+1} - S^y_n S^x_{n+1}) + \mathrm{i}(S^z_n S^x_{n+1} - S^x_n S^z_{n+1})\rangle. \tag{14b}$$

The T-dependence of both $\omega_{\parallel}$ and $\omega_{\perp}$, if it exists, should arise from the correlation function $\langle \ldots \rangle$. Similar expressions involving $\langle S^\alpha_n S^\beta_{n+1} - S^\beta_n S^\alpha_{n+1}\rangle$ with $\alpha \neq \beta$ $(\alpha, \beta = x, y, z)$ are obtained for $d_{n,n+1} \perp z$. When a perturbation term consists of the dipolar interaction, the correlation function which appears in the expression for the resonance frequency is $\langle S^\alpha_n S^\alpha_{n+1} - S^\beta_n S^\beta_{n+1}\rangle$ [2] and hence it produces a T-dependent part when the Zeeman interaction is taken into account besides the exchange interaction for the calculation of the average $\langle \ldots \rangle$. The present antisymmetric correlation functions $\langle S^\alpha_n S^\beta_{n+1} - S^\beta_n S^\alpha_{n+1}\rangle$ with $\alpha \neq \beta$ vanish even when the Zeeman term is taken into account for the calculation of the average $\langle \ldots \rangle$. As a result, we conclude that the DM perturbation brings about no change of resonance frequency with T.

4. Discussion

As shown above, the antisymmetric perturbation

$$\sum_n d_{n,n+1} \cdot (S_n \times S_{n+1})$$

brings about characteristic temperature behaviours in both the linewidth and the resonance frequency—behaviours that are completely different from those in the symmetric perturbation systems. They solely orginate in the antisymmetric nature of the correlation functions such as $\langle S^\alpha_n S^\beta_{n+1} - S^\beta_n S^\alpha_{n+1}\rangle$ for the resonance frequency and

$\langle(S_n^\alpha S_{n+1}^\beta - S_n^\beta S_{n+1}^\alpha)^2\rangle$ for the linewidth with $\alpha\neq\beta$. In contrast, the correlation functions appear as $\langle S_n^\alpha S_{n+1}^\alpha - S_n^\beta S_{n+1}^\beta\rangle$ and $\langle S_n^\alpha S_{n+1}^\beta + S_n^\beta S_{n+1}^\alpha)^2\rangle$ in a 1DHAF with symmetric perturbation terms.

As far as we know, there has been no report on 1DHAFs in which the DM interaction is much larger than the symmetric terms. There is an exceptional example reported to date, i.e. $KCuF_3$ [15]; it has an extraordinarily strong DM interaction compared with the dipolar interaction, and hence its EPR linewidth and resonance frequency have been found to be governed by the DM perturbation. That is, the linewidth does not show an increase over the short-range-order region [15], while its resonance field measured at a fixed frequency is almost independent of T [16]. The linewidth of this compound has been interpreted following the theory for $S=\frac{1}{2}$ given by Soos *et al* [11]. Since the spin of $KCuF_3$ is $\frac{1}{2}$, our present results on the classical model of spins cannot be applicable. We think, however, that the essential temperature behaviour of the EPR lines observed in $KCuF_3$ supports the present results.

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References

[1] Drumheller J E 1982 *Magn. Reson. Rev.* **7** 123
[2] Nagata K and Tazuke Y 1972 *J. Phys. Soc. Japan* **32** 337
[3] Okamoto H and Karasudani T 1977 *J. Phys. Soc. Japan* **42** 717, 1131
[4] Tuchendler J, Magarino J and Renard J P 1979 *Phys. Rev.* B **20** 2637
[5] Zaspel C E 1985 *Phys. Rev.* B **31** 1621; 1987 *Phys. Rev.* B **36** 3953
[6] Xia Q and Riseborough P S 1988 *J. Appl. Phys.* **63** 4141
[7] Siskens Th J, Capel H W and Gaemers K J F 1975 *Physica* A **79** 259
[8] Zaspel C E 1984 *J. Chem. Phys.* **80** 3978
[9] Castner T G Jr and Seehra M S 1971 *Phys. Rev.* B **4** 38
[10] Drumheller J E, Dickey D H, Reklis R P and Zaspel C E 1972 *Phys. Rev.* B **5** 4631
[11] Soos Z G, McGregor K T, Cheung T T P and Silverstein A J 1977 *Phys. Rev.* B **16** 3036
[12] Van Vleck J H 1948 *Phys. Rev.* **74** 1168
[13] Fisher M E 1964 *Am. J. Phys.* **32** 343
[14] Tazuke Y and Nagata K 1975 *J. Phys. Soc. Japan* **38** 1003
[15] Yamada I, Fujii H and Hidaka M 1989 *J. Phys.: Condens. Matter* **1** 3397
[16] Ishii T and Yamada I 1990 *J. Phys.: Condens. Matter* **2** 5771