Analysis of the temperature dependence of the electron spin resonance linewidth in exchange-coupled magnetic insulators

M. Acikgoz $^a$ and D. L. Huber $^b$

$^a$ Department of Chemistry, Rutgers University-Newark, Newark, NJ 07102, USA. E-mail address: muhammed.acikgoz@rutgers.edu

$^b$ Department of Physics, University of Wisconsin-Madison, Madison, WI 53706, USA. E-mail address: dhuber@wisc.edu

Abstract

We analyze the temperature dependence of the electron spin resonance linewidth in exchange-coupled magnetic insulators using results from Co$_3$O$_4$ as an example. The focus is on separating the contributions from spin-spin interactions, spin-one-phonon interactions and spin-two-phonon interactions. Expressing the linewidth as a sum of the three contributions, varying as const., $BT$, and $CT^2$, respectively, we use a least-squares fit over the temperature range $50 K \leq T \leq 500 K$ to obtain values of the three components. It is found that the spin-spin mechanism is dominant below 100 $K$, while the two-phonon mechanism is most important above 250 $K$. In the intermediate region, all three mechanisms make significant contributions. The success of the high temperature approximations for the one and two-phonon terms, which occurs well below the Debye temperature of 525 $K$ in Co$_3$O$_4$, is attributed to extreme exchange narrowing of the bandwidth of phonons contributing to the linewidth.

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1. Introduction

Studies of the electron spin resonance (ESR) linewidth in the magnetic insulator CrBr₃ revealed a linear temperature dependence well above the critical region [1]. Subsequent analysis show that the linear temperature dependence was associated with the spin-phonon interaction [2,3]. Since Cr³⁺ is not an S-state ion, the interactions between the spins and phonons are comparatively strong in contrast to S-state systems such as MnF₂ where the linewidth approaches a constant value at high temperatures. Recently, results have been reported which indicate that in Co₃O₄, the linewidth begins to vary quadratically with increasing temperature [4,5] suggesting the growing importance of two-phonon processes. In this note we demonstrate how one can separate the contributions to the linewidth from the three processes.

2. Analysis

As pointed out in [3], above the critical region, the ESR linewidth in magnetic insulators can be expressed in the form

$$\Delta H(T) = \left( \frac{\chi_0(T)}{\chi(T)} \right) [A + BT + CT^2]$$

where $\chi_0(T)$ denotes the Curie susceptibility and $\chi(T)$ is the static susceptibility. The letters $A$, $B$ and $C$ refer to the spin-spin, spin-one-phonon, and spin-two-phonon contributions, respectively. In our analysis of Co₃O₄ we can use the Curie-Weiss approximation for the susceptibility so that the first factor in (1) takes the form

$$\frac{\chi_0(T)}{\chi(T)} = \frac{T - \theta}{T}$$

We analyze the data for Co₃O₄ from [5] over the range $50 \text{ K} \leq T \leq 500 \text{ K}$ with $\theta = -110 \text{ K}$ [5]. We assume that $A$, $B$ and $C$ are temperature-independent above $50 \text{ K}$, a high-temperature approximation consistent with the Néel temperature, 39 K. In Fig. 1 we show the results obtained from fitting the X-band linewidth data shown in Fig. 4 of [5]. The corresponding fitting parameters are

$$A = 3.607 \times 10^2 \text{ Oe}$$

$$B = 1.225 \text{ OeK}^{-1}$$

$$C = 8.570 \times 10^{-3} \text{ OeK}^{-2}$$

We discuss these results in the following section.

3. Discussion

As noted, the temperature dependence of the ESR linewidth of Co₃O₄ above the critical region reflects the interplay of spin-spin and spin-lattice interactions. In Fig. 2, we plot the temperature dependence of the three terms contributing to the product $[T / (T + 110)]\Delta H(T)$. It is apparent that below $100 \text{ K}$, the linewidth is dominated by the contribution from spin-spin interactions, while the two-phonon processes are dominant above $250 \text{ K}$ [4,5]. The range $100 \text{ K}$
< \ T \ < \ 250 \ K \ is \ a \ cross-over \ region \ where \ all \ three \ processes \ are \ making \ a \ significant \ contribution \ to \ the \ width.

It \ should \ be \ noted \ that \ the \ functional \ forms \ \( BT \) \ and \ \( CT^2 \) \ are \ high-temperature \ approximations \ to \ the \ expressions \ for \ the \ one-phonon \ and \ two-phonon \ processes. \ The \ explanation \ for \ the \ high \ temperature \ form \ is \ discussed \ in \ [3] \ where \ it \ is \ shown \ that \ exchange \ interactions \ between \ the \ magnetic \ ions \ limit \ the \ frequencies \ of \ the \ phonons \ contributing \ to \ the \ linewidth \ in \ a \ one-phonon \ process. \ Similar \ arguments \ apply \ to \ two-phonon \ processes \ as \ well. \ An \ estimate \ of \ the \ cut-off \ energy \ of \ the \ phonons \ in \ a \ system \ with \ nearest-neighbor \ exchange \ interactions \ is \ given \ in \ [3] \ and \ takes \ the \ form

\[
E_{\text{cut-off}}^{\text{phonon}} = (2S(S + 1)n_{nn}J_{nn}^2)^{1/2}
\]

where \( J_{nn} \) \ is \ the \ nearest-neighbor \ exchange \ interaction \ and \ \( n_{nn} \) \ is \ the \ number \ of \ nearest \ neighbors. \ With \ \( S = 3/2, n_{nn} = 4 \) \ and \ \( J_{nn} = 11.7 \ K \) \ [4], \ we \ obtain \ a \ cut-off \ temperature \ equal \ to \ 64 \ K. \ As \ a \ result, \ we \ expect \ the \ high \ temperature \ form \ for \ the \ spin-phonon \ contributions \ is \ appropriate \ for \ \( T > 60 – 70 \ K \). \ For \ comparison, \ we \ note \ that \ the \ Debye \ temperature \ is \ the \ nominal \ boundary \ of \ the \ high \ temperature \ regime \ for \ acoustic \ phonons. \ In \ the \ case \ of \ Co3O4 \ the \ experimental \ Debye \ temperature \ is \ 525 \ K \ [6], \ eight \ times \ larger \ than \ the \ exchange \ cut-off, \ indicating \ an \ extreme \ exchange \ narrowing \ of \ the \ width \ of \ the \ phonon \ band \ contributing \ to \ the \ linewidth.

4. Summary

We \ have \ outlined \ an \ approach \ for \ separating \ the \ contributions \ to \ the \ high-temperature \ ESR \ linewidth \ in \ magnetic \ insulators \ that \ are \ associated \ with \ spin-spin \ and \ one- \ and \ two-phonon \ processes. \ The \ analysis \ is \ applicable \ above \ the \ critical \ regime, \ which \ we \ loosely \ identify \ as \ the \ region \ where \ the \ Curie-Weiss \ approximation \ is \ useful, \ provided \ the \ temperature \ exceeds \ a \ cutoff \ temperature \ of \ the \ band \ of \ contributing \ phonons. \ ESR \ studies \ of \ Co3O4 \ show \ evidence \ of \ an \ extreme \ exchange \ narrowing \ of \ the \ spectrum \ of \ contributing \ phonons.

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Figure captions

Fig. 1. Co$_3$O$_4$. ESR linewidth vs $T$. The data points are from [5]. The solid curve is a three-parameter least squares fit described in the text.

Fig. 2. Co$_3$O$_4$. Contributions to $[T/(T+110)] \Delta H(T)$ from spin-spin interactions, $A$; one-phonon processes, $BT$; two-phonon processes, $CT^2$
Fig. 1
Fig. 2