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Mn$^{2+}$ ear linewidth measurements have been carried out in exchange narrowing systems RbMnF$_3$, RhMnF$_3$ and MnF$_2$ and a systematic discrepancy between the experimental and the predictions of the theory in its simplest form has been reported. In this paper, we report a similar discrepancy for the exchange narrowed system TlMnCl$_3$, if the assumed line profile for the line shape function $I(\omega)$ is a truncated Lorentzian. However, better agreement with the experimental linewidth is obtained, if $I(\omega)$ is a truncated double Lorentzian.

TlMnCl$_3$ was prepared by heating (at 600$^\circ$C) a stoichiometric mixture of TlCl and MnCl$_2$$\cdot$4H$_2$O. ESR experiments for a powder sample of TlMnCl$_3$ were carried out on a conventional ear spectrometer operating in X band with 100 KHz modulation. An exchange narrowed signal of linewidth (half width at half maximum) $= 80 \pm 1$ gauss (Figure 1) is observed over a temperature range from 30$^\circ$C to -150$^\circ$C, suggesting that the transverse relaxation time $T_2 (= \text{longitudinal relaxation time } T_1)$ is independent of temperature. This exchange narrowed line broadens out and disappears at -156$^\circ$C, suggesting that the system undergoes an antiferromagnetic transition.

The ear linewidth for an exchange narrowed system is calculated using the moment method developed by Van Vleck and the linear response theory by Anderson and Weiss. Due to the
computational difficulties involved in calculating the moments beyond \( M_4 \), one assumes a line profile for \( I(\omega) \), for the experimentally observed lineshape whose linewidth is characterised by the available moments. We find that if \( I(\omega) \) is a truncated Lorentzian (TL), the linewidth calculated is much less than the experimental value. However, the linewidth can be calculated assuming different line profiles for \( I(\omega) \). For the gaussian Lorentzian (GL), the Truncated Double Lorentzian (TDL) and the Gaussian Double Lorentzian (GDL) line shapes, the linewidths (\( \delta \)) are given as follows:

\[
\delta_{\text{GL}} = \left( \frac{\pi}{2} \right)^{\frac{3}{2}} \frac{M_2}{M_4^{\frac{1}{2}}} \]  

(1)

\[
\delta_{\text{TDL}} = \frac{2\sqrt{\gamma}}{\pi} \frac{M_3^2 M_2}{M_4^{\frac{3}{2}}} \frac{B(\eta)^{\frac{3}{2}}}{C(\eta)^{\frac{1}{2}} A(\eta)^2} \]  

(2)

\[
\delta_{\text{GDL}} = \left( \frac{2}{\pi} \right)^{\frac{1}{2}} \frac{M_3 M_2}{M_4^{\frac{3}{2}}} \frac{B(\eta)^{\frac{3}{2}}}{C(\eta)^{\frac{1}{2}} A(\eta)^2} \]  

(3)

where the symbols have been defined by equations (3.1) to (3.10) of reference 1.

The moments \( M_2 \) and \( M_4 \) were calculated with an untruncated dipolar Hamiltonian and the isotropic exchange interaction, for the powder case. An approximate expression for \( M_6 \) was used from reference 1. \( M_2 = 1.6967 \times 10^{21} \) sec\(^{-2} \); \( M_4 = 9.8825 \times 10^{48} \) sec\(^{-4} \); \( M_6 = 1.1096 \times 10^{76} \) sec\(^{-6} \), with \( a = 5.02 \) Å; \( \omega_{\text{ex}} = 5.09 \times 10^{12} \) sec\(^{-1} \); \( T = 115^\circ \text{K} \) where the symbols have their usual meaning. The calculated linewidths for different line profiles and the deviation
from the experimental value are given in the table below:

<table>
<thead>
<tr>
<th>Line Shape</th>
<th>Calculated linewidth $\delta_{cal}$ (gauss)</th>
<th>Ratio $\frac{\delta_{expt}}{\delta_{cal}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>TL</td>
<td>3.63</td>
<td>5.52</td>
</tr>
<tr>
<td>GL</td>
<td>5.01</td>
<td>3.99</td>
</tr>
<tr>
<td>GDL</td>
<td>13.20</td>
<td>1.52</td>
</tr>
<tr>
<td>TDL</td>
<td>18.76</td>
<td>1.07</td>
</tr>
</tbody>
</table>

The Fourier transform of the lineshape function $I(\omega)$, is the relaxation function $\delta(t)$ and this is related to the correlation function of the full local field $\varphi(t)^1$. In the large exchange limit, $\varphi(t)$ decays in a time of the order of $t/J$. It can be shown that for $t \gg \delta/J$,

$$\delta(t) = \exp \left[ - \langle \omega(0)^2 \rangle t \int_0^\infty \varphi(\tau) d\tau \right]$$

where $J$ is the exchange constant. Since $\delta(t)$ is exponential except near $t = 0$, the central portion of $I(\omega)$ is Lorentzian with a width $\delta = \langle \omega(0)^2 \rangle \int \varphi(t) dt$ and so $\delta$ is proportional to the area under the curve $\varphi(t)$ vs $t$. $\varphi(t)$ can be evaluated for different line profiles, by taking the Fourier transform of $I(\omega)$ and using the expression (4). It can be shown that for different line profiles $I(\omega)$, the curve $\varphi(t)$ vs $t$ follows the gaussian shape for small values of $t$, whereas it deviates from gaussian form differently for large values of $t$. So, for the TDL line shape which predicts a linewidth closer to the experimental value and has the largest area under the curve $\varphi(t)$ vs $t$, $\varphi(t)$ falls off less rapidly than
the gaussian form especially for large values of $t$. Since $\psi(t)$
is the sum of both auto correlation functions and pair correlation
functions of the full local field, we feel that this deviation
of $\psi(t)$, for large values of $t$ arises due to an additional
contribution coming from pair correlation functions in the
presence of exchange interactions.

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