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Title: MAGNETIC PROPERTIES OF THE FRUSTRATED ANTIFERROMAGNET LiCrO2

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Magnetic properties of the frustrated antiferromagnet $LiCrO_2$

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Abstract

We report Electron Paramagnetic Resonance (EPR) and dc susceptibility (χ) measurements on the two-dimensional (2D) Heisenberg triangular-lattice antiferromagnet $LiCrO_2$. From 150 to 615 K, the linewidth and the g-value are temperature independent, but below 150 K the linewidth broadens and g deviates from its high temperature value of 1.98, suggesting the presence of short-range antiferromagnetic correlations in the paramagnetic phase. $\chi(T)$ for $T > T_N \approx 62$ K agrees with the predictions of the quantum-generalized constant-coupling approximation.

Key words:
$LiCrO_2$, geometrical frustration, Cr^{3+}, Electron paramagnetic resonance

Compounds forming rhombohedrally stacked triangular-lattice antiferromagnets (TAl-AF) have been widely studied because they could exhibit magnetic ordering which is against the Néel state. In $LiCrO_2$, the Cr^{3+} ions are located in edge-sharing distorted octahedra forming a plane triangular array separated by two layers of Li^+ and O^{2-}, making it a promising candidate for a 2D Heisenberg TAl-AF.

$LiCrO_2$ was prepared by heating a mixture of Li_2CO_3 and Cr_2O_3 at 1123 K in air for 1 day. The room temperature rhombohedral lattice parameters are $a = 2.891(1)$ Å and $c = 14.363(2)$ Å in agreement with reported values [1]. The dc magnetic susceptibility $\chi(T) = M/H$ was measured by a SQUID magnetometer (Quantum Design) mated with a high-temperature oven. The electronic paramagnetic resonance (EPR) spectra were taken in powder over 55 – 615 K at 9.3 GHz, in a Varian E-line spectrometer using a TE_{102} room temperature cavity and a N_2 flow temperature control system.

EPR powder spectra arising from Cr^{3+} ($S = 3/2$) show in the paramagnetic phase a single absorption line with Lorentzian shape (see inset Fig. 1). The T–dependence of the linewidth (ΔH), calculated by fitting the experimental data with a derivative of a Lorentzian, is displayed in Fig. 1. Above 150 K, ΔH and the g-value are temperature independent, but at lower temperatures ΔH broadens and g deviates from the constant value of 1.98, suggesting the existence of short-range an-

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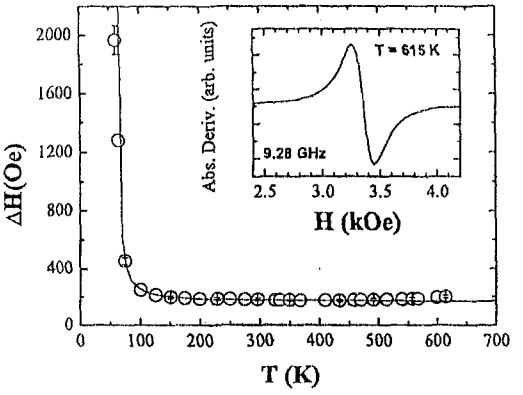

Fig. 1. EPR linewidth vs T. The solid line is the fit. The inset shows the T-dependence of the g factor.

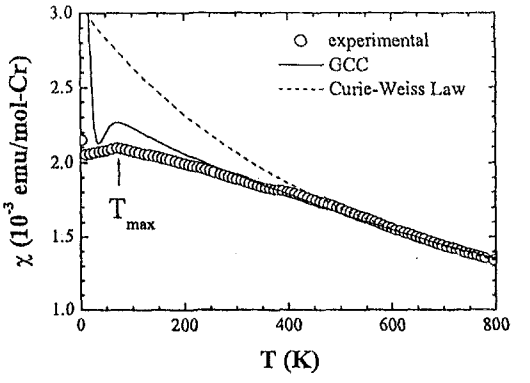

Fig. 2. $\chi(T)$ of $LiCrO_2$ measured at H = 1 kOe. The data are open circles. The solid line is the fit obtained using the quantum GCC approximation. The dashed line is the prediction of the Curie–Weiss law.

tiferromagnetic correlations in the paramagnetic phase. The EPR ΔH in $LiCrO_2$ resembles that of the 3D frustrated antiferromagnet $ZnCr_2O_4$ [2]. The temperature dependence of the linewidth was fit to $\Delta H(T) = \Delta H(\infty) + R|T - T_N|^{-x}$. The solid line in Fig. 1 shows the fitting with $\Delta H(\infty) = 165$ K, $R = 1934(20)$ Oe K^x, $T_N = 62(1)$ K, and $x = 0.78(1)$. This value of T_N agrees with the value of 62 K determined by Mössbauer spectroscopy [3].

Figure 2 shows the magnetic susceptibility of the powder sample of $LiCrO_2$. The susceptibility below 300 K is almost temperature independent displaying a weak broad maximum at $T_{max} = 75$ K, which is in agreement with the reported one [3].

The observed temperature dependent of $\chi(T)$ is analyzed in terms of the the quantum-generalized constant-coupling (GCC) method [4]. Including both nearest-(NN) and next-nearest-neighbor (NNN) interactions, the magnetic susceptibility is

$$\chi^{gcc}(T) = \frac{C}{3T} \frac{1 + \varepsilon_3(T)}{1 - (2 + 3\lambda)\, \varepsilon_3(T)},$$

where C is the Curie constant, J_1 and J_2 are the NN and NNN exchange interactions, respectively, $\lambda = \frac{J_1}{J_2}$, and expressions for $\varepsilon_3(T)$ are easily obtained from those of $\varepsilon_p(T)$ in Ref. [4] taking $p = 3$.

In Fig. 2, we show the $\chi(T)$ data in the paramagnetic phase together with the fit obtained using the quantum GCC approximation with $J_1 = 83$ K and $J_2 = -12.45$ K. We have also included a fit using the Curie–Weiss law ($450 \leq T \leq 800$ K) for $\Theta = -620$ K and $C = 1.9$ emu K/mol. It is important to stress that the C and J_1 values used in the fit to the GCC expression were those extracted from the fit to the Curie–Weiss law. Therefore, the only free parameter of the GCC model is λ. Still, agreement between the data and the quantum GCC model is quite good; the calculated susceptibility reproduces the peak at T_{max}, even though the theoretical peak is sharper. For $S = 3/2$, the GCC model predicts that the uniform susceptibility will diverge as $T \to 0$, as suggested by the lowest temperature data in Fig. 2. In contrast, the susceptibility calculated in the Curie–Weiss law shows no peak and increases monotonically.

In summary, $LiCrO_2$ displays a Cr^{3+} EPR linewidth consistent with the presence of short-range magnetic correlations in the paramagnetic phase similar to what has been found in $ZnCr_2O_4$. $\chi(T)$ is well reproduced by the quantum GCC model revealing strong signs of frustration.

References

[1] V.R. Galakhov *et al.*, Solid. State Comm. 95 (1995) 347.

[2] H. Martinho *et al.*, J. Appl. Phys. 89 (2001) 7050.

[3] C. Delmas, G. Le Flem, G. Fouassier and P. Hagenmuller, J. Phys. Chem. Solids 39 (1978) 55.

[4] A.J. Garcia-Adeva and D.L. Huber, Phys. Rev. B 63 (2001) 174433; *ibid.* 65 (2002) 184418.