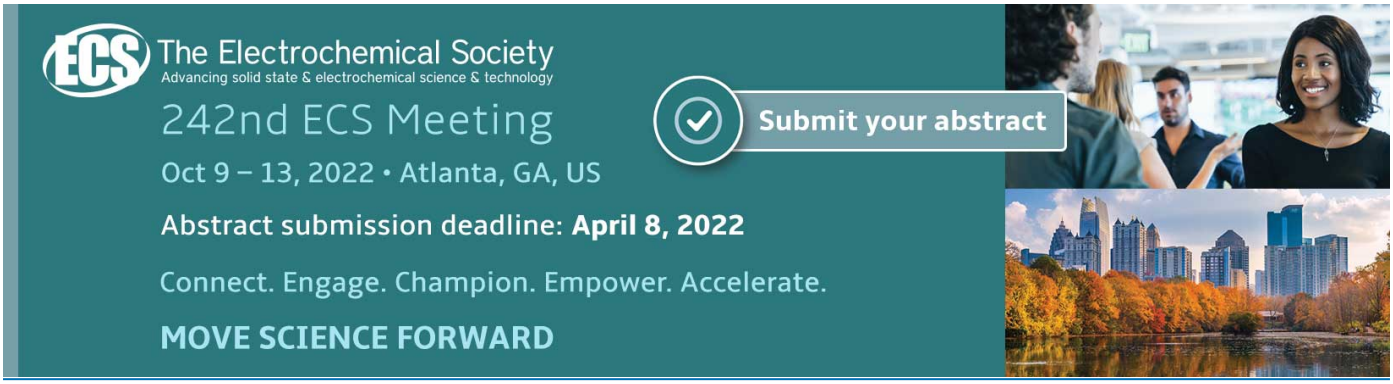
ESR in 2D triangular chromium lattices



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ESR in 2D Triangular Chromium Lattices

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Abstract. The spin dynamics in some two-dimensional (2D) triangular Cr-antiferromagnetic frustrated lattices, i.e. $HCrO_2$, $LiCrO_2$, and $NaCrO_2$ with ordered rock-salt structure as well as the delafossite compounds $CuCrO_2$ and $AgCrO_2$, has been investigated by Electron Spin Resonance (ESR). On approaching the Néel temperature T_N from above, the divergence of the temperature dependent linewidth is well described in terms of a Berezinskii-Kosterlitz-Thouless (BKT) like scenario due to magnetic vortex-antivortex pairing.

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1. Introduction

Recently, the discovery of superconductivity in $Na_xCoO_2{\cdot}yH_2O$ [1] as well as of multiferroicity in $CuCrO_2$ and $AgCrO_2$ [2] renewed the interest in triangular layered 2D antiferromagnetic oxides. These compounds are very interesting from the viewpoint of spin frustration and emerging exotic ground states [3, 4, 5]. Generally, an important aspect of 2D systems is the existence of topological defects. As early as 1971 Berezinskii [6] suggested vortices as such topological defects. Based on these ideas, Kosterlitz and Thouless [7, 8] formulated a theory of a vortex-mediated phase transition in the 2D XY model into a topological phase different from long-range magnetic order: Below a certain critical temperature – the Kosterlitz-Thouless temperature, T_{KT}, – vortices and antivortices are bound in pairs. Later on the 2D triangular Heisenberg antiferromagnet has been discussed to show a Kosterlitz-Thouless like transition in analogy to the 2D XY ferromagnet [9]. These findings encourage to search for vortices in the triangular chromium oxides, although they are of dominant Heisenberg character. In real 2D magnets, so called quasi-2D magnets, a residual interaction between neighboring magnetic layers always exists which together with the anisotropy of the intra-layer interaction can give rise to formation of 3D long-range order masking the Kosterlitz-Thouless transition. However, the tendency to undergo a vortex-pairing transition can be anticipated from the spin relaxation in the paramagnetic phase, which is influenced by the correlations of unbound vortices, as for example has recently been shown by means of ESR in the honeycomb-layer system $BaNi_2V_2O_8$ [10].

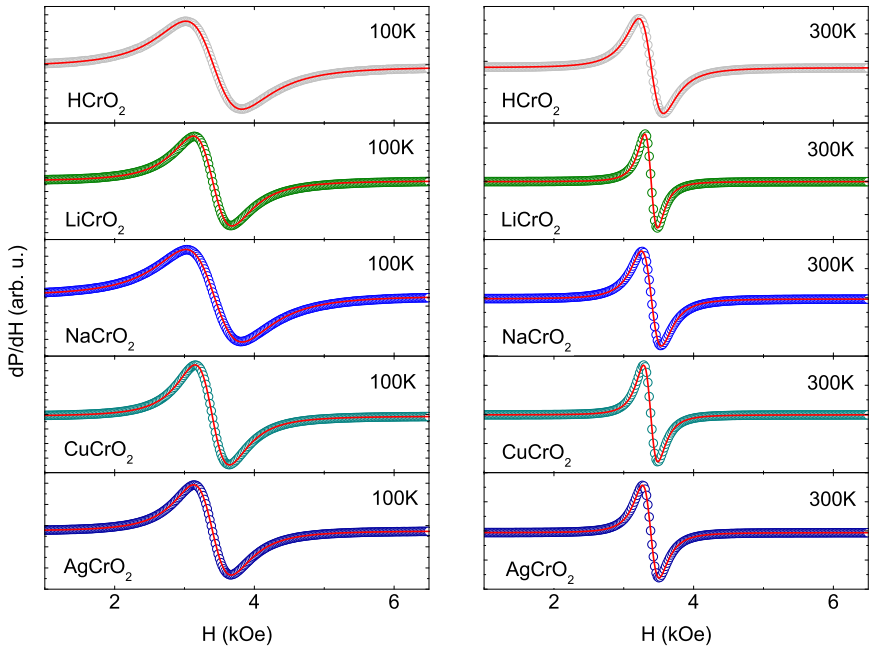

Figure 1. (Color online) ESR spectra of $ACrO_2$ in X-band for selected temperatures in the paramagnetic regime. Solid line: fit with the field derivative of a Lorentz line.

2. Experiment

Ceramic samples of $ACrO_2$ (A = Li, Na, Cu, Ag) were prepared by solid-state reactions as described in [2] and $HCrO_2$ was prepared using a hydrothermal method. For the ESR experiments the polycrystalline material was fixed in quartz tubes with paraffin. The ESR measurements were performed on a Bruker ELEXSYS E500-CW spectrometer at X-band (9.4 GHz) frequency equipped with a continuous He-gas flow cryostat (Oxford Instruments) working in the temperature range $4.2 \leq T \leq 300$ K. Due to the lock-in technique with field modulation the field derivative of the microwave–absorption signal is detected as a function of the static magnetic field. Resonance absorption occurs, if the incident microwave energy matches the energy of magnetic dipolar transitions between the electronic Zeeman levels.

Typical ESR spectra are depicted in Fig. 1. All compounds show a single exchange-narrowed resonance line, which is satisfactorily described by a Lorentz curve. At elevated temperatures the resonance field corresponds to a g-value of about $g = 2$ indicating the quenched orbital moment of the half-filled t_{2g}^3 state in Cr^{3+} (spin $S = 3/2$). With decreasing temperature, the spectra broaden rapidly (cf. Fig. 2), shift to higher resonance fields, and become undetectable on approaching the Néel temperature $T_N \approx 20$ K, 62 K, 41 K, 24 K, and 21 K for H-, Li-, Na-, Cu-, and $AgCrO_2$, respectively. The temperature dependence of the linewidth approximately follows a critical behavior $\Delta H \propto (T - T_N)^{-p}$ in the whole temperature range with an exponent of $p \approx 0.7 - 0.9$. Note that this value is rather small and theoretically unexpected for two dimensional magnets [11], where such low exponents usually appear only close to T_N. This motivates

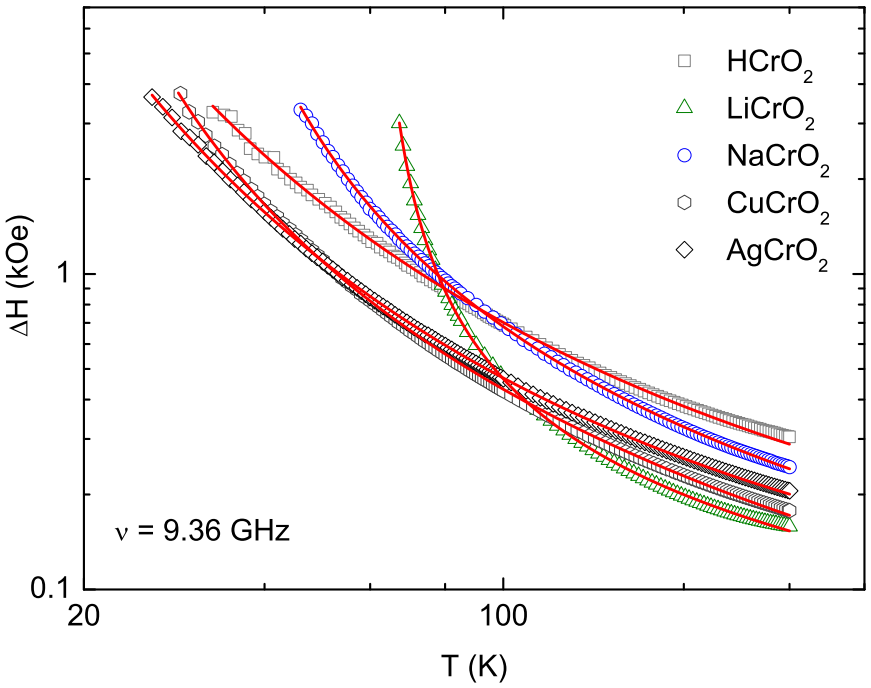

Figure 2. (Color online) Logarithmic plot of linewidth vs temperature for all compounds. Solid lines: fit in terms of a BKT-like scenario as described in the text.

a deeper analysis considering the influence of magnetic vortices on the spin relaxation.

3. Discussion and Conclusion

To understand the influence of the vortices on experimental observables one has to consider primarily the static part of the two-spin correlation function [12] which reads for $T > T_{\mathrm{KT}}$: $\langle S_x(0)S_x(r)\rangle \approx \exp[-r/\xi(T))]/\sqrt{r}$ where r denotes the distance between two spins S under consideration. In the XY model the correlation length $\xi(T)$ reveals an exponential divergence at T_{KT} following $\xi(T) = \xi_0 \exp(b/\tau^{\nu})$ with $\tau = (T/T_{\mathrm{KT}} - 1)$, $\nu = 0.5$, and $b \approx 1.5$ [8]; ξ_0 is of the order of the lattice constant. The correlation length $\xi(T)$ is related to the density of free vortices $n_{\mathrm{v}} \simeq (2\xi)^{-2}$ and, thus, can be interpreted as the average half distance between two free vortices [12]. The ESR linewidth ΔH is determined by four-spin correlation functions, which in independent-mode approximation can be factorized into products of two-spin correlation functions evaluated at the wave vector of magnetic order and $\omega \approx 0$ [11]. The influence of the vortex dynamics on the ESR linewidth was approximately derived by detailed calculations as $\Delta H = C \cdot \xi^3$ [13].

Taking C, b, T_{KT} and ν as fit parameters the linewidth data of $A\mathrm{CrO}_2$ are very well described by this expression in the whole temperature range without adding any residual linewidth contribution. One obtains the exponent $\nu \approx$ 0.43(7), 0.36(1), 0.39(3), 0.35(2), and 0.47(2) as well as the temperature $T_{\mathrm{KT}} \approx$ 4 K, 59 K, 23 K, 12 K, and 4 K for H-, Li-, Na-, Cu-, and AgCrO_2, respectively. The b parameter lies between 0.7 for Li- and 3.0

for $HCrO_2$. The most important outcome concerns the tendency of the exponent ν to attain values smaller than 0.5, i.e. around $\nu = 0.37$. Moreover, except for $LiCrO_2$, T_{KT} is far below T_N. Note that a broad peak around 25 K (close to $T_{KT} \approx 23$ K) was found in the μSR data for $NaCrO_2$, which indicates strong magnetic fluctuations even below T_N and was interpreted as a gradual freezing into the ground state [14]. Such a scenario is strongly reminiscent to the refined BKT model developed by Halperin and Nelson [15] and independently by Young [16]. This model extents the original ideas of Kosterlitz and Thouless for a Coulomb gas on a triangular lattice to describe the two-dimensional melting mechanism. The resulting correlation length resembles that of the BKT model with a modified exponent $\nu \approx 0.37$ and with an arbitrary b parameter value. Then the parameter T_{KT} corresponds to the temperature of the 2D melting transition.

Our ESR experiments in $ACrO_2$ establish the dominant role of magnetic vortices for the spin relaxation in these 2D triangular antiferromagnets. The applicability of the modified BKT model to frustrated two dimensional magnets motivates further theoretical investigations.

Acknowledgements

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