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Unconventional spin order in the triangular lattice system $NaCrO_2$: A neutron scattering study

D. Hsieh[a,*], D. Qian[a], R.F. Berger[b], R.J. Cava[b], J.W. Lynn[c], Q. Huang[c], M.Z. Hasan[a]

[a]*Joseph Henry Laboratories of Physics, Princeton University, Princeton, NJ 08544, USA*
[b]*Department of Chemistry, Princeton University, Princeton, NJ 08544, USA*
[c]*NIST Center for Neutron Research, Gaithersburg, MD 20899, USA*

Abstract

We report high resolution neutron scattering measurements on the rhombohedrally stacked triangular antiferromagnet $NaCrO_2$ which has recently been shown to exhibit an unusually broad fluctuating cross-over regime extending far below the onset of spin freezing at T_c. Our results show that at T_c purely two-dimensional quasi-static spin correlations of the 120° type exist. Below some cross-over temperature ($T \sim 0.75T_c$) a small incommensuration develops which helps resolve the inter-layer spin frustration and drives *short-range* three-dimensional magnetic order. This incommensuration assisted dimensional cross-over suggests that inter-layer frustration is responsible for stabilizing the rare 2D correlated phase above $0.75T_c$.

Keywords: $NaCrO_2$; Triangular lattice; Low dimensional magnetism; Neutron scattering

The two-dimensional (2D) triangular lattice Heisenberg antiferromagnet (TLHAFM) has been theoretically shown to exhibit many novel topological phases [1]. However, in real layered triangular magnets, finite inter-layer coupling nearly always gives rise to three-dimensional (3D) antiferromagnetic long-range order near the expected topological transition temperature. In the rhombohedrally (ABCABC) stacked spin-3/2 TLHAFM $NaCrO_2$, 2D magnetic correlations develop below $T_c \sim 41$ K but no LRO is observed by neutron scattering down to lowest temperatures [2]. Recent μSR and NMR studies observe an onset of spin freezing at T_c [3], but unlike a conventional second order phase transition, this is followed by a 30 K-broad regime of slow spin fluctuations upon further cooling.

To understand this unconventional spin ordering process, we performed a detailed neutron scattering study of $NaCrO_2$. Measurements were made at the NIST Center for Neutron Research using the now decommissioned BT2 triple-axis spectrometer with a fixed incident and final energy of 14.7 meV. A polycrystalline sample of $NaCrO_2$ was prepared using the method described by Olariu et al. [3].

Fig. 1A shows the magnetic diffraction intensity (I) as a function of temperature (T) and momentum transfer (Q). Constant T scans were fit to a convolution of the instrument resolution with the spherical average of magnetic scattering from a quasi-2D magnet. This is described by a Warren function with an additional phase factor [4]:

$$I(Q) = C\frac{|F(Q)|^2}{Q}\sum_{\vec{\tau}}\int_{-\pi/2}^{\pi/2}(|\vec{m}_{\vec{q}}|^2 - |\hat{Q}.\vec{m}_{\vec{q}}|^2) \times e^{-(\xi_{ab}^2/4\pi)(Q\cos\phi - |\vec{\tau}\pm\vec{q}_\parallel|)^2} \times \left|\sum_{n=0}^{N-1} e^{in[\vec{\tau}.\vec{d}_\parallel + (Q\sin\phi \mp q_\perp)d_\perp]}\right|^2 d\phi. \quad (1)$$

Here C is an instrumental constant, $F(Q)$ is the magnetic form factor for Cr^{3+}, $\vec{q}_\parallel$ and $\vec{q}_\perp$ are the components of the ordering wave vector parallel and perpendicular to the triangular planes, respectively, and the sum is over 2D reciprocal lattice vectors $\vec{\tau}$. The ordered moment on a site $\vec{r}$ is given by $\vec{m}_{\vec{q}}e^{i\vec{q}.\vec{r}} + \vec{m}^*_{\vec{q}}e^{-i\vec{q}.\vec{r}}$, where $\vec{m}_{\vec{q}} = (im_{qx}, m_{qy}, m_{qz})$.

*Corresponding author. Tel.: +1 609 258 3044; fax: +1 609 258 1006.
E-mail addresses: dhsieh@princeton.edu (D. Hsieh), mzhasan@princeton.edu (M.Z. Hasan).

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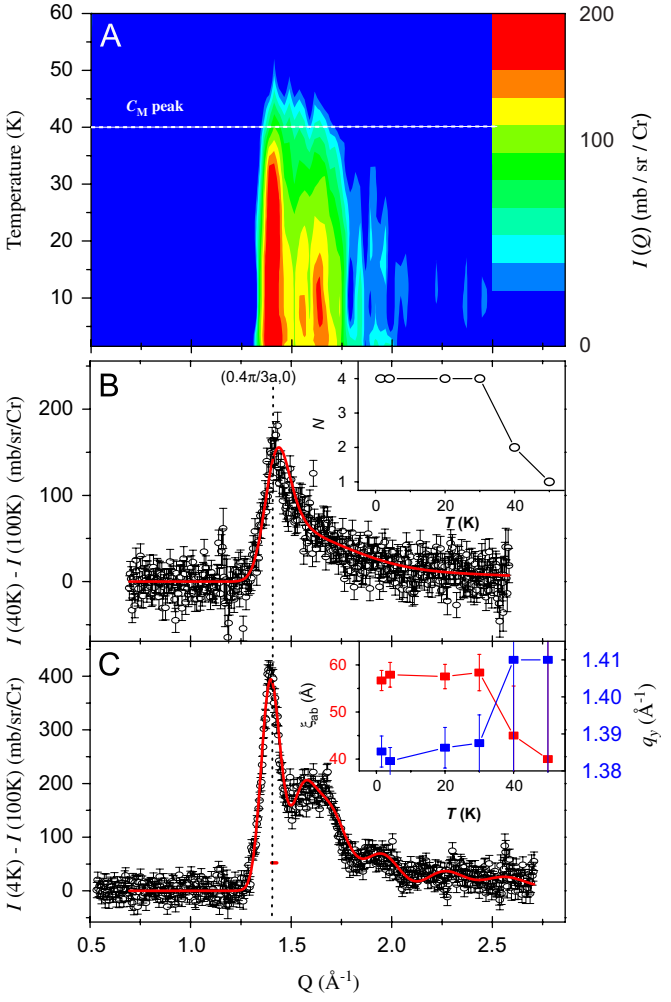

Fig. 1. Quasi-static spin correlations. (A) Neutron thermodiffractogram measured using DCS. Dashed line indicates peak position of magnetic specific heat C_M. Intensity is shown on a log scale. (B) 40 K and (C) 4 K magnetic diffraction obtained by subtracting 100 K background. Red (online) curves are fits using methods described in the text, and the red bar indicates the FWHM resolution width. Inset (B): value of N that minimized χ^2 at each temperature. Inset (C): temperature dependence of ξ_{ab} and q_y.

The number of correlated layers N was restricted to integer values, with $\vec{d} = (a/2\sqrt{3}, a/2, c/3)$ being a vector connecting adjacent layers.

For $T > 40$ K, $I(Q)$ has a sawtooth shape (Fig. 1B) indicating highly 2D spin correlations ($N \approx 1$). Weak inter-plane correlations, evidenced by intensity modulations along the high-Q tail of the sawtooth profile, develop below 40 K and N saturates to 4 (= 1 unit cell) from 30 K down to 1.5 K. All (0 0 l) nuclear Bragg peaks were resolution limited ($\Delta Q^{-1} > 2c$) which suggests that the short-ranged nature of ξ_c is not due to structural disorder. We can also rule out a spin glass freezing based on the absence of splitting between zero field cooled and field cooled susceptibility curves [5]. The in-plane correlation length ξ_{ab} also increases abruptly between 50 and 30 K, corresponding to 13 and 20 triangular lattice spacings, respectively, and remains constant upon further cooling. The data at all temperatures show spins confined to the xz-plane (i.e. $m_{qy} \ll m_{qx}, m_{qz}$), which is consistent with electron paramagnetic resonance (EPR) results supporting a small easy-axis anisotropy [6].

The development of weak c-axis correlations between 40 and 30 K is accompanied by a change in $\vec{q}$ (inset Fig. 1C) from $(0.0(0), 1.41(5), 0.0(0)) \approx (0, 1.411, 0) = (0, 4\pi/3a, 0)$ to $(0.03(1), 1.38(1), 0.15(2))$, respectively, and remains fairly constant down to 1.5 K. The ordering wave vector at 40 K corresponds to a 120° arrangement, and is consistent with the absence of a difference signal in the $Q \to 0$ limit. When such 120° ordered triangular layers are rhombohedrally stacked there is a cancellation of inter-layer Weiss fields which leads to a classical decoupling of spin layers. The slight incommensuration at 30 K represents a departure from 120° order which resolves the inter-layer frustration to some extent, and naturally explains the onset of inter-layer correlations. This supports recent μSR and NMR data [3] which point to a dynamical cross-over near 30 K.

The classical ground state $\vec{q}$ of a rhombohedrally stacked TLHAFM including only nearest neighbor in-plane J_1 and inter-plane J_1' exchange was shown in Ref. [7] to be infinitely degenerate along helices given by $((-2j'/\sqrt{3}a)\ \sin(cq_z/3), 4\pi/3a + (2j'/\sqrt{3}a)\ \cos(cq_z/3), q_z)$, where $j' \equiv J_1'/J_1$. Quantum fluctuations select the discrete set $q_z = (2n\pi/c)$ $((2n+1)\pi/c)$ for j' positive (negative), where n is an integer. The $\vec{q}$ extracted from data below 30 K lies on the degenerate helix if one takes $j' \sim -0.1$. Contrary to Ref. [3], this result together with $J_1 + J_1'$ extracted from the Curie–Weiss temperature of $NaCrO_2$ [8] suggests that inter-layer coupling is driven by exchange ($J_1' \sim 0.3$ meV) as opposed to Cr^{3+} dipolar energies $(3.8\mu_B)^2/d^3 \sim 0.06$ meV. The fitted q_z value 0.15(2) is close to $\pi/c = 0.197$ consistent with $j' < 0$. Thus quantum order-by-disorder may be responsible for the single-$\vec{q}$ low T magnetic structure, and may be related to the short-range nature of the c-axis correlations.

To conclude, our neutron scattering results clearly reveal a rare 2D AFM phase in $NaCrO_2$. The development of c-axis correlations at low temperature together with a weak incommensurate modulation strongly suggests inter-layer frustration as the mechanism for stabilizing this unusual 2D phase. Such a 2D magnetic phase of Mott insulators can be a host for exotic superconductors where pairing could be mediated by the naturally available spin fluctuations and makes an interesting variant of the recently discovered cobaltate superconductor, hydrated Na_xCoO_2 [9].

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