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# Na<sub>x</sub>CoO<sub>2</sub> : Enhanced low-energy excitations of electrons on a 2D triangular lattice

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## Abstract

To elucidate the low-energy excitation spectrum of correlated electrons on a 2D triangular lattice, we have studied the electrical resistance and specific heat down to 0.5 K and in magnetic fields up to 14 T, in Na<sub>x</sub>CoO<sub>2</sub> samples with a Na content ranging from  $x \approx 0.5$  to 0.82. Two distinct regimes are observed: for  $x$  from about 0.6 to  $x \approx 0.75$  the specific heat is strongly enhanced, with a pronounced upturn of  $C_p/T$  below about 10 K, reaching 47 mJ/(mol K<sup>2</sup>). This enhancement is suppressed in a magnetic field indicative of strong low-energy spin fluctuations. At higher Na content the fluctuations are reduced and  $\mu$ SR data confirm the SDW ground state below 22 K and the much reduced heat capacity is field independent.

*Key words:* Na<sub>x</sub>CoO<sub>2</sub>, spin fluctuations, thermodynamic properties, triangular lattice, SDW, muon spin rotation

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In light of the complexity of the ground state due to geometric frustration among localized magnetic moments residing on a triangular lattice,[1] it is of particular interest to study the low-energy physics of correlated electrons as they acquire some itinerancy on the triangular lattice. Na<sub>x</sub>CoO<sub>2</sub> is metallic, crystallizes in a layered structure with edge-sharing CoO<sub>2</sub> octahedra, and Co occupies a triangular lattice. Originally known for its unusually large thermoelectric effect[2] this compound exhibits a rich phase diagram as a function of temperature and Na content. Particularly notable are the superconductivity in water-intercalated material ( $x$  near 0.3 to 0.4),[3] a CDW-like state near  $x = 0.5$ ,[4] and a SDW-like state near  $x = 0.8$ . [5,6]

While changing the electron count in the CoO<sub>2</sub> layer by variation of the Na content appears both simple and effective, it is also to be expected that the Coulomb interaction with the Na ions will influence the charge dy-

namics of the itinerant electrons. This additional complexity becomes even more relevant because of the partial mobility of the Na ions even at ambient and modestly elevated temperature, and the resulting possibility for the Na ions to order. The CDW-like electronic state is a clear example as it has been associated with a stripe-like ordering of the Na ions,[7] and several indications for Na-ordering at other values of  $x$  have been observed (e.g. Ref. [8]). Thus it will be desirable to identify the influence of the Fermi level shift and that of the interactions with the Na ions, even as there is reason to expect some interplay between them.[9,10] In this report we focus on the composition range where Na ordering appears not to dominate the electronic states, at least not on the level as it does at  $x = 0.5$ .

The specific heat and electrical resistivity were measured in a Quantum Design PPMS setup with a He-3 insert and a 14 T magnet. Polycrystalline samples were synthesized in a traditional way and the Na content was confirmed by the unit cell parameters, in particular by the  $c/a$  ratio.  $\mu^+$ SR studies were performed at the Paul Scherrer Institute and were particularly helpful to check the magnetic homogeneity of the samples.

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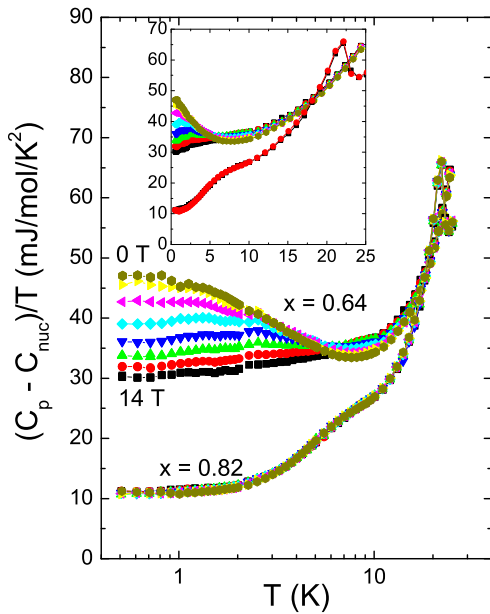


Fig. 1. Low-temperature specific heat  $C_p/T$  of two  $\text{Na}_x\text{CoO}_2$  samples representing the regions of contrasting electronic low- $T$  states. The nuclear contribution has been subtracted for clarity. The  $x = 0.64$  sample is from the broad composition region where  $C_p/T$  increases below about 10 K, indicative of strong low energy spin fluctuations, which are suppressed by a magnetic field. The  $x = 0.82$  sample undergoes a SDW-like transition at 22 K. The low- $T$  value of  $C_p/T$  is much lower than in the spin-fluctuation region, and is not influenced by a magnetic field. The inset shows the same data on a linear  $T$  scale.

The essential features of the low- $T$  specific heat for two representative compositions are shown in the figure. A well defined nuclear contribution stemming predominantly from Co has been subtracted for clarity. The sample with  $x = 0.64$  is representative of samples with a sodium content in the range from  $x \approx 0.6$  to  $x \approx 0.75$ . Zero field  $\mu^+$ SR spectra down to 1.7 K in a  $x = 0.7$  sample indicate the absence of magnetic order. For this composition range, the specific heat reveals a high density of low energy excitations, giving rise to an increase of  $C_p/T$  below about 10 K to reach values as large as  $47 \text{ mJ}/(\text{mol K}^2)$ , which has been observed in various studies (e.g. [11,12]). Additional insights come from measurements in an external magnetic field. In a magnetic field, these excitations are significantly suppressed, shown in the figure as the set of data for 0 T to 14 T with 2 T intervals. The suppression tends to saturate in high fields and from an extrapolation a total reduction by about  $20 \text{ mJ}/(\text{mol K}^2)$ , corresponding to as much as 43 %, can be estimated. Apparently, the low-energy excitations are of magnetic origin. The residual value of  $C_p/T$  is  $26$  to  $28 \text{ mJ}/(\text{mol K}^2)$  and it is still enhanced by about a factor of two compared to

the measured electronic  $C_p/T$  for the superconducting compound at lower  $x$ , [13] or the  $x = 0.82$  sample also shown in the figure. For comparison, an early LDA calculation for  $\text{Na}_{0.5}\text{CoO}_2$  gave a  $C_p/T$  of roughly  $11 \text{ mJ}/(\text{mol K}^2)$ . [14] The measured  $C_p/T$  thus suggests two contributions to the low-energy excitation spectrum. One with a typical energy scale of about 1 meV, which is suppressed in a magnetic field, and another that extends to a significantly higher energy.

The situation is different at higher Na composition where a SDW develops, marked by a distinct anomaly at 22 K. [6,12] In the present study we have focused on the low- $T$  region and the dependence on a magnetic field. The  $\mu^+$ SR results of such a sample reveal the excellent quality in terms of the magnetic volume fraction. A fit of the  $\mu^+$ SR data using three exponential relaxation functions and a Kubo Toyabe function indicates that over 80 % of the muons experience a static magnetic field. Below about 2 K  $C_p/T$  saturates and at 0.5 K amounts to  $11 \text{ mJ}/(\text{mol K}^2)$ . Interestingly, within the experimental uncertainty of a few percent, this value remains constant in fields up to 14 T. The same also holds true for the entire temperature range covered: It is worth noting that neither the transition temperature at 22 K nor the broad hump near 5 to 10 K, which has been associated with another magnetic transition, [12] are modified in a field up to 14 T.

The low-temperature electrical resistivity  $\rho$  in the spin fluctuation region ( $x = 0.7$ ) significantly deviates from a  $T^2$  behavior. At low  $T$ , it follows a temperature dependence  $\rho = \rho_0 + \tilde{A}T^a$ , with  $a$  smaller than 2. The exponent  $a$  starts at about 1.3 in zero field and increases rapidly with increasing field reaching about 1.8 at 14 T.

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