Magnetic Fluctuations and Specific Heat in Na_{x}CoO_{2} Near a Lifshitz Transition

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Magnetic fluctuations and specific heat in Na_xCoO_2 near a Lifshitz Fermi surface topological transition

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We analyze the temperature and doping dependence of the specific heat $C(T)$ in Na_xCoO_2. This material was conjectured to undergo a Lifshitz -type topological transition at $x = x_c = 0.62$, in which a new electron Fermi pocket emerges at the Γ point, in addition to the existing hole pocket with large k_F. The data show that near $x = x_c$, the temperature dependence of $C(T)/T$ at low T gets stronger as x approaches x_c from below and then reverses the trend and changes sign at $x \geq x_c$. We argue that this behavior can be quantitatively explained within the spin-fluctuation theory. We show that magnetic fluctuations are enhanced near x_c at momenta around k_F and their dynamics changes between $x \leq x_c$ and $x > x_c$, when the new pocket forms. We demonstrate that this explains the temperature dependence of $C(T)/T$. We show that at larger x ($x > 0.65$) the system enters a magnetic quantum critical regime where $C(T)/T$ roughly scales as $\log T$. This behavior extends to progressively lower T as x increases towards a magnetic instability at $x \approx 0.75$.

Introduction The layered cobaltates Na_xCoO_2 have been the subject of intense studies in recent years due to their very rich phase diagram and associated rich physics [1–7]. Their structure is similar to that of copper oxides and consists of alternatively stacked layers of CoO_2 separated by sodium ions. The Co atoms form a triangular lattice [8]. The hydrated compound Na_xCoO_2:yH_2O with $x \sim 0.3$ shows superconductivity [9], most likely of electronic origin. The anhydrated parent compound Na_xCoO_2 exhibits low resistivity and thermal conductivity and high thermopower [1, 2] for $0.5 < x < 0.9$ and magnetic order for $0.75 < x < 0.9$ (Refs.6, 7, 10, 11). In the paramagnetic phase Na_xCoO_2 shows a conventional metallic behavior at $x \leq 0.6$ and at larger x displays strong temperature dependence of both spin susceptibility and specific heat down to very low T . This change of behavior has been attributed [12] to a putative Lifshitz-type topological transition [13] (LTT) at $x_c \approx 0.62$, in which a small three-dimensional (3D) electron Fermi pocket appears around $k = 0$, in addition to the already existing quasi-2D hole pocket with large k_{F1} (Ref.14), see Fig. 1. Although the small pocket has not yet been observed directly, ARPES measurements at smaller x did find a local minimum in the quasiparticle dispersion at the Γ point [15]. Similar topological transitions have been either observed or proposed for several solid state [16–23] and cold atom systems [24], and the understanding of the role played by the interactions near the LTT transition is of rather general interest to condensed matter and cold atoms communities.

The subject of this paper is the analysis of the interaction contributions to the specific heat $C(T)$ in Na_xCoO_2 at around the critical x_c for LTT. The experimental data [12], show (see Figs. 3 and 4) that for doping near x_c, the temperature dependence of $C(T)/T$ is more complex than the $C(T)/T = $

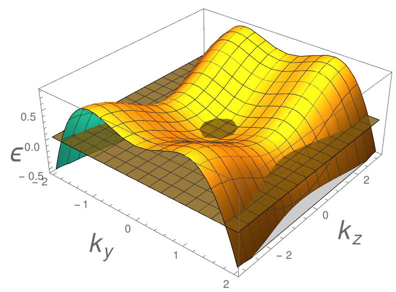

FIG. 1: The lattice fermionic dispersion $\epsilon(k)$ at $k_x = 0$ (in units of $t_1 \approx 0.1 eV$). See [25] for the values of the other hopping integrals. Note that the dispersion is approximately rotationally invariant in the $k_x - k_y$ plane and is quite shallow: the depth of the local minimum is around 20 meV.

$\gamma_1 + \gamma_3 T^2 + O(T^4)$ expected in an ordinary Fermi liquid (FL). The FL behavior itself is not broken in the sense that γ_1 remains finite. However the T dependence at $x = x_c$ is stronger than T^2, as evidenced by the fact that the fits of the data on $C(T)/T$ to $\gamma_1 + \gamma_3 T^2$ behavior [12] in finite intervals around different T yield larger γ_3 as T goes down (see Ref.36). This does not allow one to interpret γ_1 directly as a density of states, and the full computation is needed to compare the data with the theory. For doping levels $0.65 < x < 0.75$ the data show [3] that, to a good approximation, $C(T)/T \propto \log T$ in a wide range of temperatures $T \sim 1 - 10$ K, see Fig. 4a. This logarithmic temperature dependence progressively spans over larger temperature range as x approaches 0.75, where a magnetic order develops (Refs.[6, 7, 10, 11]).

Some qualitative features of the experimental data of $C(T)$ at $x \sim x_c$ are reproduced by the free-fermion formula for specific heat, with the quasiparticle dispersion taken from first-principle calculations (Fig. 2a). In particular, γ_1 increases and

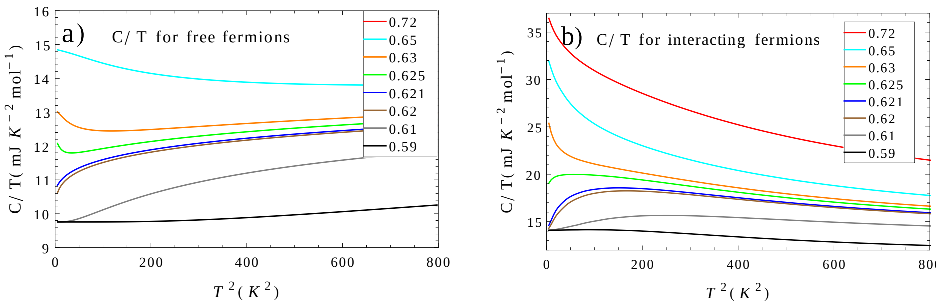

FIG. 2: Theoretical results for the specific heat $C(T)/T$ for for several Na dopings x for free fermions (a) and for fermions with magnetically-mediated interaction with $\xi = 7a_0$ (b). Both are obtained without expanding in T, using the dispersion from Fig.1.

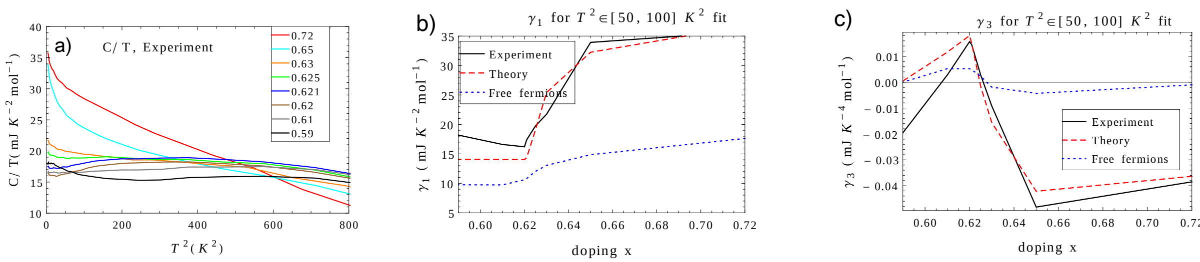

FIG. 3: (a) The data [12] for $C(T)/T$ for $x = 0.59$ to 0.72 with the doping-independent phonon contribution subtracted. (b,c) The fits of experimental and theoretical $C(T)/T$ to $C(T)/T = \gamma_1 + \gamma_3 T^2$ for T^2 between $50K^2$ and $100K^2$.

γ_3 passes through a maximum around $x = 0.62$, see Fig. 3b,c . However, the magnitudes of γ_1 and γ_3 are much smaller than in the data and the maximum in γ_3 is too shallow. A strong temperature dependence of $C(T)/T$ may potentially come from phonons, but γ_3 due to phonons is highly unlikely to become singular at $x = x_c$. This implies that the observed features of $C(T)$ are most likely caused by electron-electron interactions. Interactions with a small momentum transfer q give rise to linear in T dependence of $C(T)/T$ in 2D due to non-analyticity associated with the Landau damping [26]. That a linear in T term has not been observed in Na_xCoO_2 near x_c implies that small-q fluctuations are weak near this doping[27]. Interactions with a finite momentum transfer $q \approx k_{F1}$ are expected to be strong and sensitive to the opening of a new piece of electron FS as the static fermionic polarization operator $\Pi(k_{F1})$ gets enhanced as x approaches x_c. An enhancement of $\Pi(k_{F1})$ generally implies that spin fluctuations at k_{F1} get softer and mediate fermion-fermion interaction at low energies [27].

The spin-fluctuation contribution to γ_3 has been analyzed before for systems with a single 3D FS[31]. In this situation, the sign of γ_3 is negative. This negative sign can be traced back [31] to positive sign of the prefactor for the ω^2 term in the dynamical spin susceptibility $\chi(q,\omega)$. The latter behaves at small frequencies and at momenta $q < 2k_F$, which connects points on the FS, as $\chi^{-1}(q,\omega) \propto \xi^{-2} + b\omega^2 - i\gamma\omega$ with $b \propto 1/q^2 > 0$. We show that in our case relevant momenta are around k_{F_1} and situation with $b > 0$ holds for $x > x_c$, when a small 3D pocket emerges and k_{F_1} connects fermions at the two FSs. For $x < x_c$, when only the 2D FS is present, we found that the sign of b is negative. This gives rise to positive γ_3 at $x \leq x_c$ and negative γ_3 at $x > x_c$, consistent with the data in Na_xCoO_2 (see Fig. 3b,c). We further show that b is singular at small μ and this gives rise to non-monotonic behavior of γ_3 around x_c – it increases upon approaching x_c from below, passes through a maximum and then rapidly decreases and changes sign at $x \geq x_c$ (Fig. 3c). We argue that this behavior is fully consistent with the data.

When the temperature exceeds $1/(\xi^2\gamma)$, the system enters into a quantum-critical regime. We found that in this regime, the specific heat can be well fitted by $C(T)/T \propto \log T$ (see Fig. 4). The lower boundary of quantum-critical behavior extends to lower T as x increases towards the onset of a magnetic transition at $x \approx 0.75$. This is again consistent with the experiment [3] which observed $C(T)/T \propto \log T$ down to 0.1 K at $x = 0.747$.

The model. We follow earlier works[14, 32] and consider fermions with the tight-binding dispersion $\epsilon(k)$ on a triangular lattice with hopping up to second neighbors in xy plane and to nearest neighbors along z-direction [25]. The dispersion, shown in Fig. 1, has a hole-like behavior at large momentum $(\partial\epsilon(k)/\partial k < 0)$ and a local minimum at the Γ point $\mathbf{k} = 0$. At $\mu < 0$, $(x < x_c = 0.62)$ the Fermi surface consists of a single quasi-2D hole pocket with large $k_F = k_{F1}$. As μ crosses zero and becomes positive,

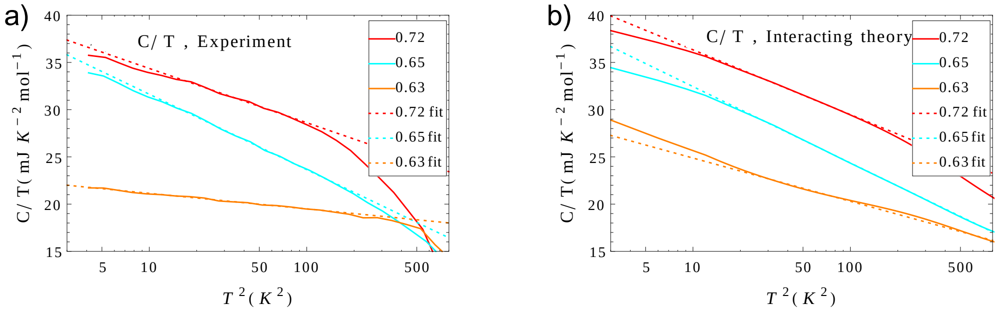

FIG. 4: Experimental data for doping $x = 0.63, 0.65, 0.72$ from Ref.12 (a) and theoretical (spin-fluctuation) result (b) for $C(T)/T$ in semi-logarithmic temperature scale. The dashed lines correspond to $C(T)/T \propto \log T$ fit. The prefactor of the $\log T$ depends on magnetic correlation length ξ

a new 3D Fermi pocket appears, centered at the Γ point (see Fig. 1). For the specific heat analysis at small $|\mu|$ we can approximate the dispersion near $k = 0$ by $\epsilon(k) = k^2/(2m) + k_z^2/(2m_z)$ and approximate the large Fermi surface by an effectively 2D dispersion $\epsilon(k) \approx v_{F1}(k - k_{F1})$, where $k = \sqrt{k_x^2 + k_y^2}$. In our analysis, we do not consider Na charge ordering. Such an ordering does indeed develop at intermediate dopings [33, 34]. However, in the measurements in Ref. 12, which we compare with our theory, the samples were quenched from high temperature to room temperature without showing any signs of Na-ordering during characterization and were argued to be in a quasi-equilibrium state [35].

$C(T)$ for free fermions. To set the stage for the analysis of interaction effects we first compute the specific heat for free fermions with non-monotonic dispersion $\epsilon(k)$. The grand canonical potential is given by

$$\Omega(T, \mu, V) = -T \int \rho(\epsilon) \ln(1 + e^{-(\epsilon-\mu)/T}) d\epsilon, \quad (1)$$

Evaluating the entropy $S(T, \mu, V)$, extracting $\mu = \mu(T, V)$ from the condition on the number of particles and expanding $C(T) = C_V(T) = T\left(\frac{\partial S}{\partial T}\right)_V$ in temperature, we obtain at the lowest T

$$C(T)/T = \gamma_1 + \gamma_3 T^2 + O\left(T^4\right)$$
$$\gamma_1 = \frac{\pi^2 \rho}{3}, \quad \gamma_3 = \frac{\pi^4}{30} \frac{\left(7\rho\rho'' - 5\left(\rho'\right)^2\right)}{\rho} \quad (2)$$

where $\rho(\mu)$ and its derivatives over μ are computed at $T = 0$. The low-T expansion in (2) is valid for $T < |\mu|$. Analyzing (2), we find that for $\mu < 0$, when there is no electron pocket, the T dependence comes from a large hole pocket and is non-singular. For $\mu > 0$, the electron pocket appears with $\rho(\mu) \propto \sqrt{\mu}\theta(\mu)$. This gives rise to negative γ_3, which diverges at small μ as $1/\mu^{3/2}$. At $\mu = 0$ the analytic expansion in powers of T^2 doesn't work even at the lowest T. We found[36] that in this case

$$\frac{C(T)}{T} = \gamma_1 + 2.88 \frac{m\sqrt{2m_z}}{\pi^2} \sqrt{T} + \mathcal{O}(T) \quad (3)$$

The same behavior holds at a finite μ, when $T > |\mu|$. Observe that the prefactor for $\sqrt{T}$ term is positive, opposite to that of $T^2/\mu^{3/2}$ term. This implies that the temperature dependence of $C(T)/T$ changes sign at some positive μ. The actual T dependence of $C(T)/T$, obtained without expanding in T, is presented in Fig. 2a, and γ_1 and γ_3 extracted from fitting this $C(T)/T$ by $\gamma_1 + \gamma_3 T^2$ in different windows of T are shown in Fig.3b,c and in Ref.36. We see that both γ_1 and γ_3 depend on where the T window is set, and γ_3 as a function of doping changes sign at some $x > x_c$, i.e., at some positive μ, as expected.

Interaction contribution to $C(T)$. At a qualitative level, the free-fermion formula for $C(T)$ is consistent with the data. At the quantitative level, it strongly differs from the measured $C(T)$, even if we would use a renormalized dispersion with larger effective density of states. To see the inconsistency, we compare in Fig.3b,c the theoretical and experimental doping dependence of $C(T)$ and particularly the values of γ_1 and γ_3 fitted over various temperature ranges. We see that the magnitude of $C(T)/T$ for free fermions and the strength of doping variation of γ_3, extracted from it, is much smaller than in the data. These discrepancies call for the analysis of interaction contributions to $C(T)$.

A fully renormalized fermion-fermion interaction can be decomposed into effective interactions in the charge and in the spin channel. For systems with screened Coulomb repulsion, the effective interaction in the spin channel get enhanced and, if the system is reasonably close to a Stoner instability, can be viewed as mediated by spin fluctuations. Na_xCoO_2 does develop a magnetic order at $x > 0.75$ [6, 7, 10, 11], and it seems reasonable to expect that magnetic fluctuations develop already at $x \approx x_c$.

The spin-fluctuation contribution to the thermodynamic potential is given by [31, 37, 38]

$$\Omega = \Omega_0 + \int \frac{d\omega}{\pi} n_B(\omega) \int \frac{d^3q}{(2\pi)^3} \mathrm{Im} \ln \chi^{-1}(q,\omega) \quad (4)$$

where Ω_0 is the free-fermion part, n_B is the Bose function, and $\chi(q,\omega)$ is fully renormalized dynamical spin susceptibility.

To obtain $\chi(q,\omega)$ we use the same strategy as in earlier works [39, 40]: compute first the static spin susceptibility $\chi_0(q,\omega=0)$ of free fermions, then collect RPA-type renormalization and convert $\chi_0(q,\omega=0)$ into full static $\chi(q,\omega=0)$, and then compute the bosonic self-energy coming from the interaction with low-energy fermions and obtain the full dynamical $\chi(q,\omega)$ at low frequencies. The result is[36]

$$\chi^{-1}(q,\omega) = \frac{\overline{\chi}}{\xi^{-2} + (q-k_{F1})^2 + b\omega^2 - i\gamma\omega} \quad (5)$$

where ξ is a magnetic correlation length and the last term is the Landau damping. The sign of γ_3 term in $C(T)$ depends on the sign of b – the prefactor for the ω^2 term (see Eq. (9) below). To obtain b in our case we first evaluated the susceptibility of free fermions $\chi_0(q,\omega)$ and then obtained $\chi(q,\omega)$ using Random Phase Approximation (RPA). For most relevant $q \approx k_{F_1}$ we obtained (see [36] for details)

$$\chi_0(q,\omega) = \frac{\sqrt{mm_z}}{4\pi^2 v_{F1}} \left[(\omega-\tilde{\mu})\log(|\omega-\tilde{\mu}|) - (\omega+\tilde{\mu})\log(|\omega+\tilde{\mu}|)\right] + ... \quad (6)$$

where $\tilde{\mu} = \mu - (q-k_{F_1})^2/(2m)$ and dots stand for regular terms. Expanding in ω and substituting into the RPA formula, we obtain

$$b = \frac{\sqrt{mm_z}}{4\pi^2 m_z a_0 v_{F1}} \frac{1}{\tilde{\mu}} \quad (7)$$

$$\gamma = \frac{\sqrt{mm_z}}{4\pi m_z a_0 v_{F1}} \theta(\tilde{\mu}) + \frac{1}{\sqrt{3}\pi v_{F1}^2 m_z a_0 a_z}, \quad (8)$$

where a_0 is of order of lattice spacing in xy plane, a_z is inter-layer spacing. Note that near $\mu = 0$ the quadratic coefficient b is singular and its dependence on q becomes important. The $1/\tilde{\mu}$ dependence of b originates from the singularity in the derivative of density of states at the Lifshitz transition. The T^3 term in $C(T)$ at $x < x_c$ and small T ($T < |\mu|$ and $T < 1/(\xi^2\gamma)$) comes from expanding $\mathrm{Im}\ln\chi^{-1}$ in (4) to order ω^3 and integrating over q near $q = k_{F1}$. When $|\mu| > \xi^{-2}/m$ the q-dependence of b and γ may be neglected and we obtain

$$\gamma_3 = \gamma k_{F1} \xi^3 \frac{\pi^3}{10} \left(-4b - (\gamma\xi)^2\right) \quad (9)$$

Eq.(7) for b suggests a singular behavior of γ_3 near $\mu = 0$. For small $|\mu| < \xi^{-2}/m$ the singularity is smoothed by q-dependence of γ and b and eq.(9) needs to be replaced by the result of numerical integration. The results, in particular a sharp maximum in γ_3 near x_c, are in good agreement with experiment, see Fig. 3b,c.

At higher temperatures, when $T > 1/(\xi^2\gamma)$ the system enters into a quantum-critical regime where it shows the same behavior as at $\xi^{-1} = 0$. The form of $C(T)/T$ at such temperatures in principle depends on the effective dimensionality of spin fluctuations around $q = q_0$ (see Ref. 36). We find, however, that such dimension-specific behavior holds only at high T, while in the intermediate regime $T \gtrsim 1/(\xi^2\gamma)$, $C(T)/T$ can be well fitted by $\log T$ even for effectively 1D spin fluctuations. This agrees with the data which show a $\log T$ behavior even at doping $x = 0.65$, see Fig. 4. As ξ and γ increase at larger x, the lower boundary of $\log T$ behavior of $C(T)/T$ stretches to progressively smaller T and a prefactor of $\log T$ grows, in agreement with the experiments at higher doping (Ref. (3, 12)).

For quantitative comparison with the data, we compute the dynamical part of particle-hole bubble without expanding in frequency and use (4) to obtain the thermodynamic potential and the specific heat. To estimate ξ we use the experimental data for $\chi(0,0)/\gamma_1$ at $x \approx x_c$ and our numerical RPA result for the prefactor for $(q-q_0)^2$ term in $\chi^{-1}(q,\omega)$. Extracting ξ from these data we obtain $\xi \approx 7a_0$ near $x = 0.62$ and it grows with the doping. For better comparison we subtract from the data the contribution from phonons $C_{ph} \approx T^3 \cdot 0.07 mJK^{-4}mol^{-1}$, which only weakly depends on doping [41]. The results are shown in Fig. 2b and Fig. 3b,c. We see that theoretical and experimental $C(T)$ agree quite well over a wide range of temperatures, and the agreement between γ_1 and γ_3, extracted from the data and from spin-fluctuation theory, is also very good. We emphasize that the doping variation of γ_3 is not affected by the phonon contribution and thus measures solely the contribution to $C(T)$ from spin fluctuations. From this perspective, a good agreement with the data is an indication that magnetic fluctuations with large $q = k_{F1}$ are strong in Na_xCoO_2 near the LTT. The $\log T$ behavior of $C(T)/T$, which we found at $T \sim 3 - 10K$ for $x \approx 0.7$ is also consistent with the data, see Fig.4. Finally, we note that the experimental data on γ_1, fitted at $T \sim 10$K, show a small discontinuity as a function of doping, Figs.3b,c, which is expected if the LTT is first order [42]. The jump in μ is estimated to be 5 to 10 meV. When we take this into account, we obtain a sharper doping dependence of γ_3, leading to an even better agreement with the data.

Conclusions. In this work we have analyzed the specific heat in the layered cobaltate Na_xCoO_2. Near $x = 0.62$ the system exhibits a non-analytic temperature dependence and strong doping variation of the specific heat coefficient $C(T)/T$. We explained the data based on the idea that at $x_c = 0.62$ the system undergoes a LTT in which a new electron pocket appears. We demonstrated that the non-analytic temperature dependence of $C(T)/T$ at $x = x_c$ and its strong doping variation is quantitatively reproduced if the interaction is mediated by spin fluctuations peaked at the wave-vector which connects the original and the emerging Fermi surfaces. We argued that the observed $\log T$ behavior of $C(T)/T$ at larger doping $0.65 \lesssim x < 0.75$ is an indication that the system enters into the magnetic critical regime.

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