

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 physics¹⁻⁷. 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 lattice⁸. The hydrated compound $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ with $x \sim 0.3$ shows superconductivity⁹, most likely of electronic origin. The anhydrous 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 attributed¹² to a putative Lifshitz-type topological transition¹³ (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 point¹⁵. 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 interaction contributions to the specific heat $C(T)$ in Na_xCoO_2 at around the critical x_c for LTT. The experimental data¹², show (see Figs. 3 and 4) that for doping near x_c , the temperature dependence of $C(T)/T$ is more complex than the $C(T)/T = \gamma_1 + \gamma_3 T^2 + O(T^4)$ expected in an ordinary Fermi liquid (FL). The FL behavior itself is not broken in

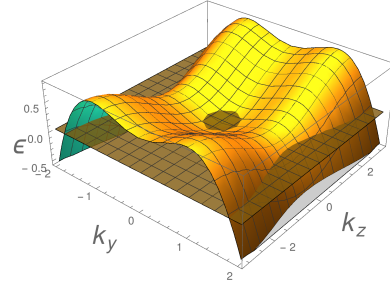


FIG. 1: The lattice fermionic dispersion $\epsilon(k)$ at $k_x = 0$ (in units of $t_1 \approx 0.1\text{eV}$). See²⁵ for the values of the other hopping integrals. Note that the dispersion is approximately rotationally invariant in the $k_x - k_y$ plane and is quite shallow: the depth of the local minimum is around 20 meV.

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 $\gamma_1 + \gamma_3 T^2$ behavior¹² 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 show³ 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 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 maxi-

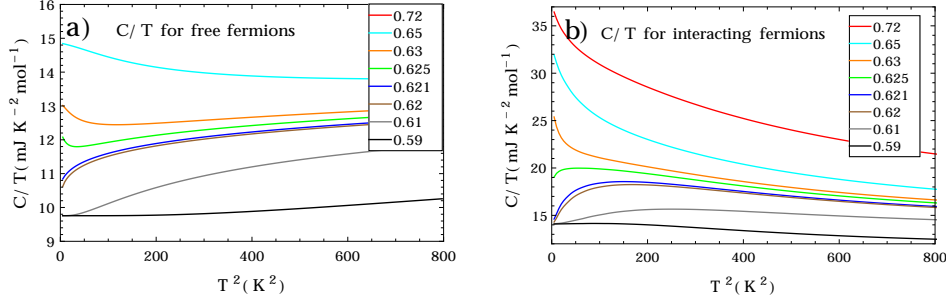


FIG. 2: Theoretical results for the specific heat $C(T)/T$ for several Na dopings x for free fermions (a) and for fermions with magnetically-mediated interaction with $\xi = 7a_0$ (b). Both are obtained without expanding in T , using the dispersion from Fig.1.

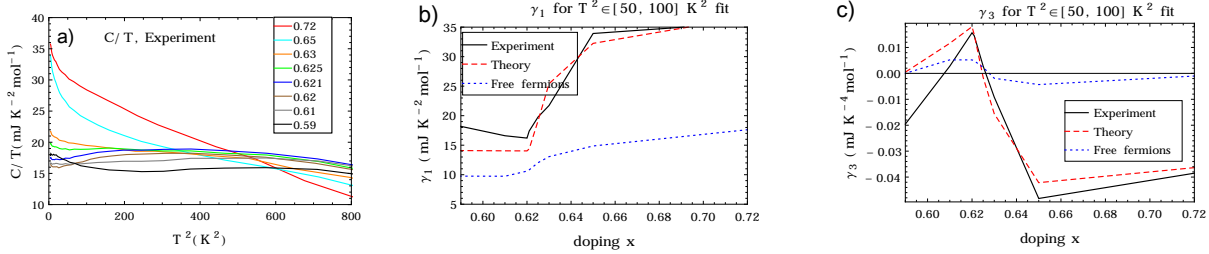


FIG. 3: (a) The data¹² for $C(T)/T$ for $x = 0.59$ to 0.72 with the doping-independent phonon contribution subtracted. (b,c) The fits of experimental and theoretical $C(T)/T$ to $C(T)/T = \gamma_1 + \gamma_3 T^2$ for T^2 between 50K^2 and 100K^2 .

imum 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 damping²⁶. 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 doping²⁷. 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 energies²⁷.

The spin-fluctuation contribution to γ_3 has been analyzed before for systems with a single 3D FS³¹. In this situation, the sign of γ_3 is negative. This negative sign can be traced back³¹ 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_{F1} and situation with $b > 0$ holds for $x > x_c$, when a small 3D pocket emerges and k_{F1} connects fermions at the two FSs. For $x < x_c$, when only 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 experiment³ 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 -direction²⁵. 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, 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

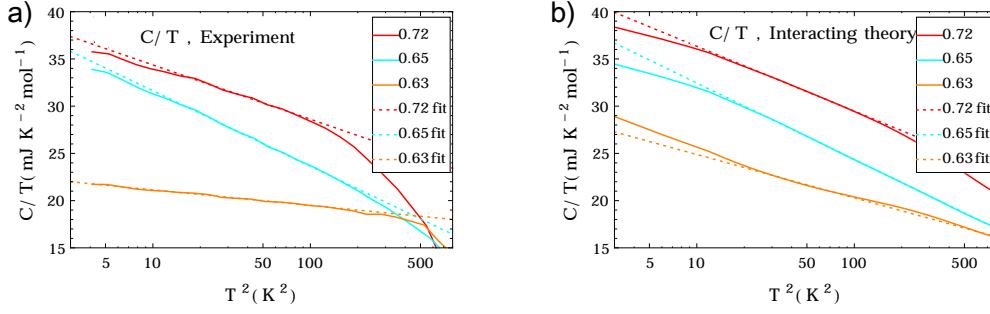


FIG. 4: Experimental data for doping $x = 0.63, 0.65, 0.72$ from Ref.12 (a) and theoretical (spin-fluctuation) result (b) for $C(T)/T$ in semi-logarithmic temperature scale. The dashed lines correspond to $C(T)/T \propto \log T$ fit. The prefactor of the $\log T$ depends on magnetic correlation length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

Na charge ordering. Such an ordering does indeed develop at intermediate dopings^{33,34}. However, the experimentalists, who performed the measurements on quenched samples of Na_xCoO_2 at $x \geq 0.6$, did not observe time-dependent phenomenon and argued³⁵ that their quenched samples are in quasi-equilibrium state.

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 $C(T) = C_V(T) = T (\frac{\partial S}{\partial T})_V$ in temperature, we obtain at the lowest T

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

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 found³⁶ that in this case

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

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}

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

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 is³⁶

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

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 RPA. For most relevant $q \approx k_{F1}$ we obtained (see [36] for details)

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

where $\tilde{\mu} = \mu - (q - k_{F1})^2/(2m)$ and dots stand for regular terms. Expanding in ω and substituting into 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 $\text{Im} \ln \chi^{-1}$ 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

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

The 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 $T > 1/(\xi^2\gamma)$ we find that the system enters into a quantum-critical regime where the system behavior is the same 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 compute 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 doping⁴¹. 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 Figs.4. Finally, we note that the experimental data on γ_1 , fitted at $T \sim 10K$, show a small discontinuity as a function of doping, Figs.3b,c, which is expected if the LTT is first order⁴². 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 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 dopings $0.65 \lesssim x < 0.75$ is an indication that the system enters into the magnetic critical regime.

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Supplementary

A. Magnetic susceptibility.

We follow earlier works³⁹ assuming that the static magnetic susceptibility and regular part of its frequency dependence are governed by high-energy processes. Then, the Landau damping term and the singular part of the remaining frequency dependence come from fermions with low energies and can be obtained within the low-energy spin-fermion model. As a result, the contributions from high-energy fermions can be incorporated into the static susceptibility through a tunable “magnetic correlation length” parameter, and we then focus on the particle-hole contributions coming from low fermion energies.

The free-fermion susceptibility χ_1 , coming solely from the large cylindrical Fermi-surface (FS) is a 2D Lindhard function:

$$\text{Im}\chi_1(\omega) = \omega \frac{2\chi_0}{\sqrt{3}k_{F1}v_{F1}a_z} = \omega \frac{1}{\pi\sqrt{3}v_{F1}^2a_z} \quad (10)$$

$$\text{Re}\chi_1(\omega) \approx \frac{k_{F1}}{2\pi v_{F1}a_z} \quad (11)$$

where a_z is inter-layer lattice spacing. This χ_1 does not carry any interesting frequency or chemical potential dependence.

On the contrary, the susceptibility $\chi_{12}(q, \omega)$, coming from the particle-hole processes with total momentum $q \approx k_{F1}$, depends non-trivially on ω and μ . We take the momenta q to be near the distance between Fermi momentum for the hole FS and Γ point where electron FS emerges for $x > x_c$, i.e., consider $q = k_{F1} + \tilde{q}$ and assume \tilde{q} to be small. Because k_{F2} for the electron pocket is either zero ($x < x_c$) or very small ($x > x_c$), we deal with a special case when the frequency may exceed the Fermi energy of the small pocket. This gives rise to a non-linear frequency dependence of the imaginary part of the susceptibility at $q \approx k_{F1}$. To simplify the discussion, we approximate the hole dispersion as purely 2D and the dispersion near Γ as a 3D parabola. Evaluating the imaginary part of the particle-hole bubble involving hole-like and electron-like excitations, we obtain

$$\text{Im}\chi_{12}(q, \omega) = \frac{1}{16\pi^2 v_{F1}} S \quad (12)$$

Here S is the area in the k_y, k_z plane, where $\mu - \omega < \tilde{q}^2/(2m) + k_y^2/(2m) + k_z^2/(2m_z) < \mu + \omega$. This area is a ring for $|\omega| < \mu - \tilde{q}^2/(2m) \equiv \tilde{\mu}$ and an ellipse otherwise; the ellipse shrinks to an empty set if $\tilde{\mu} + |\omega| < 0$. In explicit form:

$$S = 2\pi\sqrt{mm_z}((\tilde{\mu} + \omega)\theta(\tilde{\mu} + \omega) - (\tilde{\mu} - \omega)\theta(\tilde{\mu} - \omega)) \quad (13)$$

where $\tilde{\mu} = \mu - \tilde{q}^2/(2m)$. We also need an extra factor of 2, if we add the contribution from the opposite patch of the large pocket.

Analyzing $S(\omega, \tilde{q} = 0)$, we find that it has a linear frequency dependence at the lowest frequencies at $\mu > 0$, when the small pocket is present, then there is a cusp at $\omega = \mu$, and then another linear dependence, with twice smaller slope. For $\mu < 0$, when there is no pocket but the dispersion has a local minimum at Γ , the slope is zero at $\omega < -\mu$ and becomes finite only after the cusp at $-\mu$, see Fig.5. At a nonzero \tilde{q} the results are the same as at $\tilde{q} = 0$ if one replaces μ by $\tilde{\mu}$.

The frequency-dependent part of $\text{Re}\chi_{12}(q, \omega)$ can be computed elegantly from Kramers-Kronig transformation. As the second frequency derivative of the imaginary part is a delta-function at the cusp, it is easy to compute the second frequency derivative of the real part:

$$\partial_\omega^2 \text{Re}\chi_{12}(\omega) = \frac{\sqrt{mm_z}}{4\pi^2 v_{F1}} \left(\frac{1}{\omega - \tilde{\mu}} - \frac{1}{\omega + \tilde{\mu}} \right) \quad (14)$$

This expression is singular at frequencies $\omega = \pm\tilde{\mu}$, see Fig. 5. It is essential for our analysis that $\partial_\omega^2 \text{Re}\chi_N(\omega) > 0$ for $\tilde{\mu} < 0$ and that it diverges when $\tilde{\mu} \rightarrow 0$.

Integrating Eq. (14) over ω we obtain the full analytic expression for the frequency dependence of susceptibility:

$$\chi_{12}(q, \omega) = \chi_{12}(q, 0) + \frac{\sqrt{mm_z}}{4\pi^2 v_{F1}} [(\omega - \tilde{\mu}) \log(\omega - \tilde{\mu}) + \tilde{\mu} \log(\tilde{\mu}^2) - (\omega + \tilde{\mu}) \log(-\omega - \tilde{\mu})] \quad (15)$$

The result is presented in Fig. 5. At $\tilde{q} = \pm k_{F2}$, $\tilde{\mu} = 0$, and the singularity in $\chi_{12}(\omega)$ is located at zero frequency. The static part

$$\chi_{12}(q, 0) \approx a_0 m_z (q - q_0)^2 + \text{const} \quad (16)$$

has non-universal high energy contributions which have to be computed numerically and are effectively included in our calculations through the value of the correlation length. The parameter a_0 is of the order of the lattice spacing in xy plane.

B. Direct derivation of the prefactor of the ω^2 term in spin susceptibility at $q = k_{F1}$.

In this section, we offer an alternative derivation of the prefactor b . Consider the case $\mu < 0$ and $q = k_{F1}$

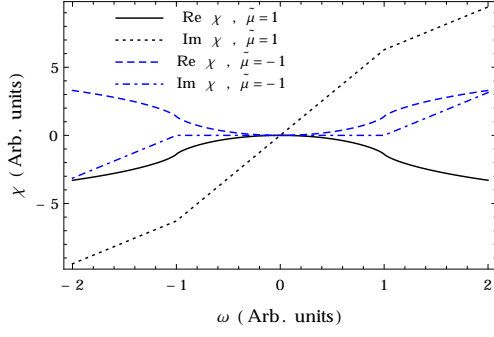


FIG. 5: The frequency dependence of the susceptibility $\chi_{12}(\omega)$

along the x -axis. Working in Matsubara formalism we obtain for the free-theory susceptibility at $T = 0$:

$$\chi_0(k_{F1}, i\Omega) = - \int \frac{d^3k d\omega}{(2\pi)^4} G(k, i\omega) G(k+k_{F1}, i\omega+i\Omega) \quad (17)$$

The Green's functions are

$$G(k+k_{F1}, i\omega+i\Omega) = \frac{1}{i(\omega+\Omega) - \left(\frac{k_x^2+k_y^2}{2m} + \frac{k_z^2}{2m_z} + |\mu| \right)} \quad (18)$$

$$G(k, i\omega) = \frac{1}{i\omega + v_{F1}k_x} \quad (19)$$

One Green's function is taken near the $k = 0$ (where the small pocket is due to appear) and another is near the large hole pocket. The contribution is doubled to account for the reverse situation. Expanding in Ω we get the quadratic term:

$$\chi_0^{(2)}(k_{F1}, i\Omega) = +2\Omega^2 \int \frac{dk_z dk_x dk_y d\omega}{(2\pi)^4} \frac{1}{\left(i\omega - \left(\frac{k_x^2+k_y^2}{2m} + \frac{k_z^2}{2m_z} + |\mu| \right) \right)^3 (i\omega + v_{F1}k_x)} \quad (20)$$

For $k_x > 0$ the frequency integration contour passes between the poles and we get

$$\chi_0^{(2)}(k_{F1}, i\Omega) = -2\Omega^2 \int \frac{dk_z dk_y}{(2\pi)^3} \int_0^\infty dk_x \frac{1}{\left(v_{F1}k_x + \frac{k_x^2+k_y^2}{2m} + \frac{k_z^2}{2m_z} + |\mu| \right)^3} \quad (21)$$

for small $|\mu|$ the integration is peaked at small k and we can neglect $k_x^2/(2m)$ term compared to $v_{F1}k_x$. Then defining $x = k_x v_{F1}/|\mu|$, $y = k_y/\sqrt{2|\mu|m}$ and $z = k_z/\sqrt{2|\mu|m_z}$ we get:

$$\begin{aligned} \chi_0^{(2)}(k_{F1}, i\Omega) &= -2\Omega^2 \frac{2\sqrt{mm_z}}{v_{F1}|\mu|} \int_{-\infty}^\infty \frac{dz dy}{(2\pi)^3} \int_0^\infty dx \cdot \\ &\quad \frac{1}{(x+y^2+z^2+1)^3} \\ &= -\Omega^2 \frac{\sqrt{mm_z}}{4\pi^2 v_{F1}|\mu|} \end{aligned} \quad (22)$$

The continuation to real frequencies provides a positive coefficient of Ω^2 in χ_0 , which behaves as $1/|\mu|$ for $\mu < 0$. This coincides with the more general eq.(14), if we set $\omega = 0$ and $q = k_{F1}$ in eq.(14). This unconventional result appears because the momentum q connects a Fermi-surface with a region where all states are above the Fermi level. When the small pocket appears and fermions are connected by the vector q are simultaneously at the Fermi-surface, an opposite sign is obtained.

C. Temperature expansion of the specific heat.

At low temperatures, when $|\omega| < |\mu|$ and $T < \mu$, we can expand the full free-particle susceptibility $\chi_0(q, \omega) = \chi_1(q, \omega) + \chi_{12}(q, \omega)$ in frequency as

$$\chi_0(q, \omega) = \chi_0 - m_z a_0 (q - q_0)^2 + b_0 \omega^2 + i\gamma_0 \omega \quad (23)$$

From eqs.(10,15) we extract:

$$\gamma_0 = \frac{\sqrt{mm_z}}{2\pi v_{F1}} \theta(\tilde{\mu}) + \frac{1}{\sqrt{3}\pi v_{F1}^2 a_z} \quad (24)$$

$$b_0 = -\frac{\sqrt{mm_z}}{4\pi^2 v_{F1} \tilde{\mu}} \quad (25)$$

The full RPA magnetic susceptibility $\chi(q, \omega) = \chi_0(q, \omega)/(1 - U\chi_0(q, \omega))$ is then expressed as

$$\chi(q, \omega) = \frac{\bar{\chi}}{\xi^{-2} + (q - q_0)^2 + b\omega^2 - i\gamma\omega} \quad (26)$$

where $\bar{\chi} \approx \chi_0/(Um_z a_0)$, $\xi^{-2} = (1/U - \chi_0)/(m_z a_0)$, and

$$b = -\frac{\chi_0 b_0 + \gamma_0^2}{\chi_0 a_0} \approx -\frac{b_0}{m_z a_0} = \frac{\sqrt{mm_z}}{4\pi^2 m_z a_0 v_{F1}} \frac{1}{\tilde{\mu}} \quad (27)$$

$$\gamma = \frac{\gamma_0}{m_z a_0} = \frac{\sqrt{mm_z}}{2\pi m_z a_0 v_{F1}} \theta(\tilde{\mu}) + \frac{1}{\sqrt{3}\pi v_{F1}^2 m_z a_0} \quad (28)$$

The spin-fluctuation contribution to the grand thermodynamic potential is³¹:

$$\Omega_{int} = \int \frac{d\omega}{\pi} n_B(\omega, T) \int \frac{d^3q}{(2\pi)^3} \text{Im} \ln \chi^{-1} \quad (29)$$

$n_B(\omega, T)$, however the form of temperature dependence of Ω_{int} depends on the frequency dependence of $\chi(q, \omega)$. Expanding the integrand in frequency and differentiating Ω_{int} over T we obtain the temperature expansion of the interaction contribution to the entropy $S = -\partial\Omega_{int}/\partial T$.

$$\begin{aligned} S &= \frac{T}{3(2\pi)^{D-1}} \int \frac{\gamma d^D q}{\xi^{-2} + (q - q_0)^2} - \\ &- \frac{T^3}{15(2\pi)^{D-3}} \left(\int \frac{\gamma b d^D q}{(\xi^{-2} + (q - q_0)^2)^2} + \frac{1}{3} \int \frac{d^D q \gamma^3}{(\xi^{-2} + (q - q_0)^2)^3} \right) \end{aligned} \quad (30)$$

The momentum integral is peaked at $q = q_0 \approx k_{F1}$ and we assume that it is cylindrically symmetric (the actual dispersion suggests that $q_z = \pi$ may be more important than other values of q_z , but this only changes the overall prefactor). Extracting $C(T)$ from the entropy we obtain $C(T) = T\gamma_1 + T^3\gamma_3$, where, if we neglect q -dependence of γ and b ,

$$\gamma_3 = \gamma k_{F1} \xi^3 \frac{\pi^3}{10} (-4b - (\gamma\xi)^2) \quad (31)$$

These are the expressions used in the main text. If we are close to $\mu = 0$, then b is singular, so we cannot neglect its q dependence. The integration has been done numerically, while the qualitative analytic picture is given below. At small enough μ , $1/\tilde{\mu}$ has to be replaced by $-(2m)/(q - k_{F1})^2$. Typical $q - k_{F1}$ is of order ξ^{-1} , hence b saturates at the value of order $\xi^2(m^3m_z)^{1/2}/(m_z a_0 v_{F1})$. Note that the ratio $4|b|/(\gamma\xi)^2$ does not depend on ξ . This ratio exceeds one, for the dispersion we consider, hence, according to eq.(31), $\gamma_3 > 0$. A positive γ_3 , which increases as x approaches x_c from below, is precisely what the data show (see Figs.6 c,d).

At $\mu > 0$ ($x \geq x_c$), when the new pocket appears, γ changes by a finite amount and b evolves from a negative constant at $\mu = 0$ to a positive $b \propto 1/\mu$ given by (27) for $\mu > \xi^{-2}/(2m)$. The positive b is consistent with earlier result³¹ for a single 3D FS as in both cases $q \approx k_{F1}$ connects points on the FS and our small pocket is three-dimensional. As a result, γ_3 rapidly decreases as x increases above x_c , changes sign and becomes negative. This is again consistent with the data.

D. Specific heat of free fermions for $|\mu| \ll T$.

Let us fix $x = x_c$, so that $\mu(T = 0) = 0$. The 3D pocket produces a singularity in the density of states:

$$\rho = B + A\sqrt{\mu}\theta(\mu) \quad (32)$$

where in our case $B = \frac{k_{F1}}{\pi v_{F1}}$ and $A = \frac{m\sqrt{2m_z}}{\pi^2}$. The grand canonical potential is $\Omega = -\int \tilde{\rho}(e)n_F((e - \mu)/T)de$, where $\tilde{\rho}(e) = \int^\epsilon \rho(e)de$. Then the entropy is $S = -T^{-2} \int \tilde{\rho}(e)n'_F((e - \mu(T))/T)(e - \mu)de = -\int \tilde{\rho}(eT)n'_F(e - \mu/T)(e - \mu/T)de$. The specific heat is obtained by substituting $\mu(T)$ and evaluating $C/T = (\frac{\partial S}{\partial T})_N$.

The condition on the chemical potential $\mu(T)$ is

$$\int (n_F\left(\frac{e - \mu(T)}{T}\right) - \theta(-e))(B + Ae^{1/2}\theta(e))de = 0 \quad (33)$$

this equation can be expanded in series in $\mu(T)/T$:

$$\mu(T) = -\frac{0.678 AT^{3/2}}{0.536 A \sqrt{T} + B} + \mathcal{O}((\mu/T)^2) \quad (34)$$

This expansion is valid for moderate temperatures, where $A\sqrt{T} \ll B$, then, indeed $\mu/T \ll 1$, the actual expansion parameter is $A\sqrt{T}/B$.

For the specific heat we obtain

$$C/T = \frac{\pi^2}{3}B + 2.88A\sqrt{T} - \frac{0.88A^2T}{B} + \mathcal{O}\left(\frac{A\sqrt{T}}{B}\right)^3 \quad (35)$$

E. Quantum criticality near the transition to ordered phase.

If the small pocket and the large pocket have parts with matching curvatures, the susceptibility will be strongly peaked at the momentum vector connecting them, which may result in spin density wave magnetic order. This is indeed what happens when the small pocket has grown sufficiently large at $x > 0.75$ ^{14,32}. For smaller doping, the nesting is not good enough for magnetic order, but magnetic correlation length ξ is nevertheless large. When $T \ll \epsilon_\xi$ and $T \ll \mu$, a regular Fermi-liquid expansion of $C(T)/T$ in powers of T^2 works. At slightly higher temperatures, when ξ^{-2} cannot be assumed large (see eq.(38)) this expansion does not hold. This temperature regime is relevant to the description of the behavior of $C(T)/T$ at intermediate T at x near $x = x_c = 0.62$ and down to quite low $T \sim 0.1K$ at $x = 0.747$ (Ref. 3), which is very close to $x = 0.75$ at which $\xi = \infty$. The fact that critical behavior extends to such low temperatures is remarkable.

For analytic estimate, we set ω to be of order T and using earlier results obtain

$$C(T) \sim -\int d^3\vec{q} \text{Im} \left(\log \left(\xi^{-2} + \left(\sqrt{q_x^2 + q_y^2} - k_{F1} \right)^2 - i\gamma\omega \right) \right) \Big|_{\omega \approx T} \quad (36)$$

where γ is given in eq.(28). Here we integrate over cylindrical shell of momenta near the vectors that connect Γ -point with large pocket. Here we assumed the large pocket to be cylindrical, but this assumption can be easily relaxed not changing the result. There is a whole 2D surface of important momenta that contribute to near-critical spin fluctuations (kinetic part $(\sqrt{q_x^2 + q_y^2} - k_{F1})^2$ is degenerate in q_z and angle). When $T \gtrsim \xi^{-2}/\gamma$, the system enters the critical region. Deep in this regime, the specific heat behaves as

$$\frac{C(T)}{T} \sim \frac{1}{T} \text{Im} \sqrt{\xi^{-2} + iT\gamma} \rightarrow \frac{\gamma}{\sqrt{T}} \quad (37)$$

This behavior can be traced back to the fact that the dispersion of spin fluctuations near $q = q_0$ is effectively one-dimensional since it is independent on the direction of \mathbf{q} in the xy plane and on q_z . For effectively 2D dispersion, we would obtain $C(T)/T \propto$

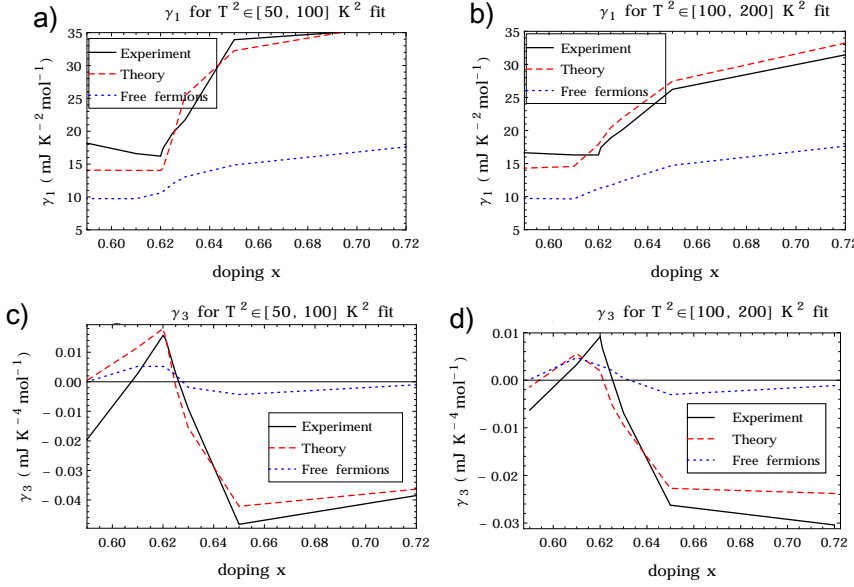


FIG. 6: The fits of the experimental data for the specific heat $C(T)/T$ for various x and theoretical formulas for free fermions and for fermions with magnetically-mediated interaction with $\xi = 7$ to $C(T)/T$ to $C(T)/T = \gamma_1 + \gamma_3 T^2$ in two temperature ranges centered at different T .

$\log T$, and for 3D dispersion $C(T)/T$ would remain finite.

We have found, however, that in our, effectively one-dimensional, case the $1/\sqrt{T}$ dependence of $C(T)/T$ holds only at high T , while in a wide range of temperatures the function $\text{Im}(\sqrt{a + iT})/T$ can be well approximated numerically by $(0.44 - 0.095 \log \frac{T}{a})/\sqrt{a}$. This behavior holds at $a \lesssim T \lesssim 40a$. In our case $a = \xi^{-2}/\gamma$ and we expect a $\log T$ behavior at $T \gtrsim \xi^{-2}/\gamma$. Assuming that the largest contribution to γ comes from the approximate nesting of small and large Fermi-surfaces, we get $\gamma^{-1} \sim a_0 v_{F1}$, so

$$T \gtrsim \frac{a_0 v_{F1}}{\xi^2} = \frac{v_{F1}}{a_0} \left(\frac{a_0}{\xi} \right)^2 \quad (38)$$

the factor v_{F1}/a_0 can be estimated to be of the order of effective hopping $t_1 \approx 1000\text{K}$. Hence, the correlation length of order of 10 lattice spacings can bring the $\log T$ behavior to the range of several Kelvins.

The calculation can be easily repeated if one assumes that lattice effects produce a significant lifting of degeneracy of the kinetic term for spin fluctuations. E.g. let us replace $(\sqrt{q_x^2 + q_y^2} - k_{F1})^2 \rightarrow ((q_x - k_{F1})^2 + q_y^2)$, then the integration in eq.(36) gives logarithmic behavior of C/T in the same range $T > \xi^{-2}/\gamma$. In the main text we used the approximate theoretical formula $C(T)/T \propto \log T$ to fit the experimental data.

F. Fits of $C(T)/T$ to $\gamma_1 + \gamma_3 T^2$ in temperature intervals centered at different T .

As emphasized in the main text, the T -dependence of the experimental $C(T)/T$ around $x = 0.62$ 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 in finite intervals around different T yield larger γ_3 with decreasing T . We show the fits in two temperature intervals centered at different T in Fig. 6.

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