Non-Fermi liquid behavior and putative magnetic quantum criticality in \((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\)

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\((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\) undergoes a bulk insulator-to-metal transition (IMT) at \(x \approx 0.04\). Through careful analysis of previously published data \((x = 0.053, 0.061, 0.076)\), we find that the resistivity deviates from ideal Fermi liquid \((\rho \sim T^2)\) behavior between 100 and 140 K, eventually becoming linear between 200 and 250 K. Meanwhile resonant (in)elastic x-ray scattering data \((x = 0.065)\) suggest a possible crossover between quantum paramagnetic and quantum critical phenomenology between 100 and 200 K. We put this into context with other results, and propose a possible phase diagram as a function of doping.

The properties of a system proximate to a quantum critical point (QCP) at \(g = g_c\) generally fit the following scenario. At low temperatures, and for \(g < g_c\), the ground state exhibits some form of long-range order (LRO). In antiferromagnets for instance, this corresponds to Néel order. This order is destroyed by classical thermal fluctuations, which dictate the scaling of thermodynamic properties in the vicinity of some critical temperature \(T_c\). Above the transition, quasiparticles may still be well-defined on intermediate length scales, even though LRO has disappeared. This corresponds to a so-called thermal disordered regime. At sufficiently high temperatures \((T \sim |g - g_c|^\nu)\), these quasiparticles are replaced by a critical continuum of excitations. This continuum is thermally excited: which leads to a characteristic \(\omega/T\) scaling of the spin fluctuations in the vicinity of the critical wavevector, and unconventional power-law temperature dependences of physical observables. If instead \(g > g_c\), then the ground state is disordered and characterized by well-defined quasiparticle excitations. The properties of the system are dictated primarily by the magnitude of a singlet-triplet gap \(\Delta \sim (g - g_c)^\nu\), which exists at all wavevectors. We refer to this as a quantum paramagnetic state, although it is also known as quantum disordered behavior in the literature. A crossover to quantum critical behavior typically occurs around \(T \sim \Delta\) (Fig. 2d). Detailed reviews of quantum phase transitions (QPTs) are given in Refs. 1 and 2.

A number of the cuprates have been proposed – albeit with some controversy – to undergo QPTs as a function of doping. These include the (hole-doped) high-temperature superconductor \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\) (LSCO), in which a magnetic QCP may lie underneath the superconducting dome.\(^3-5\) Some similarities can be drawn between LSCO and the electron-doped perovskite iridate \((\text{Sr}_{1-x}\text{La}_x)_2\text{IrO}_4\). This material, like LSCO, is an insulator and easy-plane antiferromagnet below the Néel temperature at low carrier doping. It undergoes an insulator-to-metal transition (IMT) at \(x \sim 0.04\),\(^6,7\) with evidence of a pseudogap and hole-like Fermi surface in the metallic phase \(x \geq 0.05,8,9\) along with possible spin density wave (SDW) order.\(^10\) In contrast with LSCO, however, nanoscale electronic phase separation can be observed well into the metallic regime.\(^7\) Moreover, experimental evidence of a QCP in this system is still outstanding.

Meanwhile the bilayer compound \((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\) has no direct analogues with any of the cuprates. Resistivity measurements have shown that \((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\) undergoes an IMT at \(x = 0.04,11-13\) similar to the single-layer material. Neutron scattering and second harmonic generation (SHG) measurements determined that the IMT is first order, with a structural phase transition occurring in the metallic phase at \(T_s \approx 200\) K.\(^12,14\) Yet no pseudogap could be observed by angle-resolved photo-electron spectroscopy (ARPES), with small electron-like Fermi pockets present in the metallic phase.\(^15-17\) Furthermore electronic phase separation does occur in the vicinity of the IMT, but disappears for \(x > 0.04.12\) Magnetization and neutron scattering measurements show that Néel LRO disappears above the IMT.\(^12\) Resonant elastic x-ray scattering (REXS) data demonstrates, however, that short range magnetic order persists up to \(300\) K,\(^18\) at least for \(x = 0.065.19\) Resonant inelastic x-ray scattering (RIXS) measurements on the undoped compound reveal strongly gapped spin wave excitations,\(^20\) with an additional longitudinal mode interpreted as evidence of possible quantum dimer character.\(^21\) Upon electron doping, the spin excitations become progressively more damped.\(^18,22\) Whilst there are discrepancies between the two studies, Lu et al. find that the spin gap collapses dramatically for \(x = 0.065\), proposing that the magnetic fluctuations become two-dimensional deep in the metallic phase.

We examine some of the previously published experimental data for \((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\) in more detail. What we find are three distinct electronic regimes for \(x > 0.05\), which can be discriminated through clear gradient changes in the resistivity as a function of temperature. These correlate well with phase boundaries determined by other techniques. Meanwhile REXS data suggests a
Therefore, unless otherwise stated, the following discussion applies to an effective exchange interaction \( J \) which includes the effects of the other terms in the Hamiltonian.

The QNLσM exhibits LRO at \( T = 0 \), provided that the coupling \( g = \hbar c \sqrt{2\pi} / (k_B \rho_0 a) < g_c \), where \( c = 2 \sqrt{\pi} Z_c S J a \) is the spin wave velocity, \( a \) is the lattice constant, \( \rho_0 \) is the spin stiffness, and \( Z_c = 1 + \eta \) is a renormalization factor which describes the effect of quantum fluctuations. From now on we take \( Z_c = 1 \), in order to better compare with the experimental results. Real materials typically order at non-zero temperatures as a consequence of weak anisotropies or further-neighbour interactions. Given the intrinsically broad magnetic Bragg peak at 30 K, and that bulk susceptibility data shows paramagnetic behavior at all temperatures, then this implies that the material lies on the putative QPT for \( x = 0.065 \).

In this regime, both \( \xi \) and \( S_0 \) are expected to scale

\[
\rho(T) \sim T^{3/2},
\]

for \( n = 1 \), or

\[
\rho(T) \sim T^n,
\]

for \( 1 < n < 2 \). This crossover regime is similar to that observed in overdoped cuprates.\(^{23}\) We also note that \( \rho \sim T^{3/2} \) is expected for dirty nearly antiferromagnetic metals (3D), which are in the vicinity of a QCP.\(^{2,24}\)

If \((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\) does indeed lie in the proximity of a QCP, then signatures of this should also be seen in the magnetic behavior. As mentioned previously, REXS measurements by Lu et al.\(^{18}\) reveal short-ranged magnetic order which persists above the IMT for \( x > 0.04 \). The magnetic \((0.5, 0.5, 28)\) Bragg peak appears to weaken and broaden with increasing temperature, however significant correlations are still observable at 300 K. Noting that two-dimensional layered materials frequently exhibit such behavior, we proceeded to examine the data in more detail (Fig. 2a). We fitted the data at each temperature to a power law, with the Gaussian component fixed to the width of a typical structural Bragg reflection, in order to approximate the instrumental resolution function. Varying this width within sensible bounds does not change our results significantly. Additionally, the background was fixed at all temperatures to that obtained from fitting at 20 K. What can be seen is that the correlation length \( \xi \) and equal-time structure factor \( S_0 \) are approximately constant up to 100 K (Fig. 2b,c), with these parameters decreasing at higher temperatures. There thus appear to be two distinct temperature regimes within the data.

Our findings shall initially be discussed in terms of the \( O(N) \) quantum non-linear sigma model (QNLσM); probably the simplest model to undergo a continuous quantum phase transition (QPT) in \( 2+1 \) dimensions. Specifically, we use key results given within Ref. 25, which have been obtained through exact solution in the \( N = \infty \) limit. Corrections to order \( 1/N \) are non-trivial to calculate for \( g > g_c \), which is why they have been neglected in this initial study. We note at this point that further-neighbour interactions (and anisotropies) are important for \((\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7\), which clearly manifest in the observed spin wave dispersion.\(^{18,20-22}\) Therefore, unless otherwise stated, the following discussion applies to an effective exchange interaction \( J \) which includes the effects of the other terms in the Hamiltonian.

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In this regime, both \( \xi \) and \( S_0 \) are expected to scale
Furthermore, Orange solid (pink dashed): Hertz-Millis-Moriya model (HMM) for 3D nearly antiferromagnetic metals ($d = 3, z = 2$). In the HMM picture, spin fluctuations become soft at the QCP, and are damped by a background of itinerant electrons. This is somewhat related to the paramagnon (SCR) theory by Moriya.\cite{Moriya1960} At low temperature ($T < T^*$), the inverse correlation length $\xi^{-1}$ exhibits the $T^2$ dependence characteristic of a Fermi liquid. Above $T^*$, the system exhibits quantum criticality. It has been determined that $\xi^{-1} \propto |g - g_c| + AT^{2/3}$, with the first term dominating in region I, and the second in region II (Fig. 2d). Meanwhile the equal-time structure factor $S_0$ is given by:

$$S_0(k \to 0) = \frac{1}{\pi} \int_{-\infty}^{\infty} |\omega| + 1 \left| \frac{A\xi^2}{1 - i\omega/\omega_{SF}} \right| \omega, \quad (3)$$

where $\omega_{SF} = 20 \text{ meV}$, which corresponds to the experimental spin wave energy at 30 K, and is fixed as a function of temperature. Again, we observe that the data below 100 K are well described by a Fermi liquid model, with the higher temperature data more representative of quantum critical phenomenology (mostly region II).

There is further evidence that the magnetic fluctuations at high temperature may be indicative of quantum critical behavior. In the vicinity of a QCP, spin fluctuations at the antiferromagnetic (AFM) wavevector $Q_{AF}$ are expected to exhibit $\omega/T^o$ scaling, where $\alpha$ is an independent scaling exponent. Different models for the criticality predict different results. For instance, in the HMM model, spin fluctuations are dominated by the

This theoretical model is compared with the experimental data in Figs. 2b and 2c. We find that the quantum paramagnetic model describes the experimental data below 100 K quite well. At higher temperatures however, it appears to underestimate the correlation length and structure factor. Meanwhile the quantum critical model agrees with the data above 200 K, but diverges at lower temperatures. This suggests that there may be a crossover between the two regimes in the temperature range 100–200 K. Unfortunately, there is currently no experimental data available which corresponds to this region.

The agreement between the experimental data and theory is also quantitative. Note that the value of $\Delta$ we obtain ($\Delta = 14 \text{ meV}$) is comparable with the magnon gap observed in RIXS for $x = 0.065$.\cite{et al 2018} Furthermore, $J = 62 \text{ meV}$ is in excellent agreement with the effective nearest-neighbor coupling derived from the RIXS data: $J = \sum_i J_i z_i = 64 \text{ meV}$, where $J_i$ are the individual coupling parameters (including anisotropies), and $z_i$ the number of neighbors. Whilst there are some question marks regarding the quantitative mapping of the QNLrM to $S = 1/2$ Heisenberg spin systems (for example, see the discussion in Ref. 27), the correlation is, nevertheless, remarkable.

We also plot the expected temperature dependence of the correlation length and structure factor within the Hertz-Millis-Moriya model (HMM) for 3D nearly antiferromagnetic metals ($d = 3, z = 2$). The HMM picture, spin fluctuations become soft at the QCP, and are damped by a background of itinerant electrons. This is somewhat related to the paramagnon (SCR) theory by Moriya.\cite{Moriya1960} At low temperature ($T < T^*$), the inverse correlation length $\xi^{-1}$ exhibits the $T^2$ dependence characteristic of a Fermi liquid. Above $T^*$, the system exhibits quantum criticality. It has been determined that $\xi^{-1} \propto |g - g_c| + AT^{2/3}$, with the first term dominating in region I, and the second in region II (Fig. 2d). Meanwhile the equal-time structure factor $S_0$ is given by:

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where $n(\omega) = (e^{\hbar\omega/k_BT} - 1)^{-1}$, and $\omega_{SF}$ is the characteristic energy for spin fluctuations. We assume that $\omega_{SF} = 20 \text{ meV}$, which corresponds to the experimental spin wave energy at 30 K, and is fixed as a function of temperature. Again, we observe that the data below 100 K are well described by a Fermi liquid model, with the higher temperature data more representative of quantum critical phenomenology (mostly region II).

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with $x_2 = k_B T/\Delta$, where $\Delta$ is the singlet-triplet gap:

$$\xi^{-1} = \frac{k_B T}{\hbar c} X_2(x_2)$$

$$S(k \to 0) \propto \frac{(\hbar c)^2}{k_B T} \frac{x_2 \coth(X_2/2)}{2X_2(x_2)}. \quad (1)$$

The parameter $X_2$ in the preceding expressions is a scaling function, which has the following asymptotic limits in the quantum paramagnetic ($x_2 \ll 1$) and quantum critical ($x_2 \gg 1$) regions:

$$X_2(x_2) = x_2^{-1} + 2e^{-1/x_2}, \quad x_2 \ll 1$$

$$= 2 \ln \left( \frac{\sqrt{5} + 1}{2} \right) + \frac{1}{\sqrt{5x_2}}, \quad x_2 \gg 1 \quad (2)$$

Hence this implies a correlation length which is on the order of $\Delta^{-1}$ (for $x_2 \ll 1$) or $T^{-1}$ (for $x_2 \gg 1$).
AFM order parameter. Consequently for a 3D nearly AFM metal, it predicts $E/T^{3/2}$ scaling of the dynamic spin susceptibility: $\chi^{-1}(Q_{AF},E,T) = a^{-1}(T^{3/2} - ibE)$. The parameter $b$ is related to the characteristic energy of spin fluctuations $\omega_{SF}$ defined earlier. Meanwhile anomalous exponents have been observed in some heavy fermion systems, which are believed to correspond to “local criticality”. Schröder and Poudel have proposed a modified Curie-Weiss law to describe the inverse dynamic susceptibility in such systems: $\chi^{-1}(q,E,T) = c^{-1}[\theta^{\alpha} + (T - iE)^{\alpha}]$, where $\theta(Q - Q_{AF})$ captures the wavevector dependence of the magnetic fluctuations similar to the Curie-Weiss temperature. Note that in the latter picture, the fluctuations become critical in the time domain everywhere in $q$, rather than just at $Q_{AF}$.

In Fig. 3a, we plot resonant inelastic x-ray scattering (RIXS) data at $(\pi, \pi)$ for $x = 0.065$, which was previously published in Ref. 18. The spin excitations appear highly damped, and extend to 0.4 eV energy loss at both 30 K and 295 K. Moreover, when we plot the data on double logarithmic axes (inset of Fig. 3a), it appears to scale approximately linearly for $E > 0.05$ eV. This implies that the dynamic critical exponent $z = 2/\alpha \sim 2$. Such a value is expected both for three-dimensional nearly AFM metals, and quantum dimer models on a square lattice. One complication is that the ideal QNLσM assumes Lorenz invariance, and hence $z = 1$. However the presence of disorder (caused by doping) can break this invariance and give rise to $z \neq 1.25$.

Scaling plots in Fig. 3b and 3c compare the experimental data to the theoretical predictions for the mean field ($\alpha = 1$) and HMM models respectively. Broadly speaking, both models provide an adequate description of the data. Some discrepancies at low $E/T$ can be overcome by subtracting off the elastic line and low-energy phonon contributions (compare filled and open symbols). We find that the best fit for the HMM model is obtained with $b = 0.14$, significantly smaller from the expected value of unity. Note that the simple model given here is defined precisely at the QCP and AFM wavevector, and assumes zero anisotropy. Yet the REXS data presented earlier suggests that the putative magnetic QCP lies at $x < 0.065$. Furthermore, the finite momentum resolution of the RIXS spectrometer means that we sample a number of momentum transfers close to $(\pi, \pi)$. Finally, the 20 meV spin gap present at 30 K may persist to some degree at high temperatures. Such a deviation, is therefore, not entirely unexpected.

We conclude by putting the analysis presented here in context with other experimental results. Specifically, we extend the temperature-doping phase diagram to include recent ARPES data, and our resistivity and REXS re-
sults (Fig. 4). A striking correlation is apparent between the onset of Fermi-liquid behavior at $T^*$, and the emergence of coherent spectral weight below $T_{coh}$ observed in ARPES.\textsuperscript{17} The emergence of $\rho \sim T$ behavior also appears to coincide with the structural phase transition observed by neutron scattering,\textsuperscript{12} and the loss of putative charge density wave (CDW) ordering from pump-probe optical reflectivity data.\textsuperscript{13} This suggests there are three separate electronic regimes present in $(\text{Sr}_{1-x}\text{La}_x)_2\text{Ir}_2\text{O}_7$ above the insulator-to-metal transition. We note the apparent similarity in the transport behavior as observed for overdoped cuprates.\textsuperscript{20} In the cuprates, however, the quasiparticle peak (from ARPES) persists into the crossover regime that we define here.

The REXS results also show a possible crossover between quantum paramagnetic and quantum critical phenomena somewhere between 100 and 200 K. This is consistent with our value of $T^*$, which implies that the Hertz-Millis-Moriiya picture (Fig. 2d) may be relevant for $(\text{Sr}_{1-x}\text{La}_x)_2\text{Ir}_2\text{O}_7$. Yet the (limited) RIXS data is less clear cut. There appears to be $E/T^*$ scaling of the dynamic spin susceptibility, as would be expected in the vicinity of a QCP. At present, it is not possible to conclusively distinguish between the HMM or simple mean-field pictures. Even so, it demonstrates that quantum criticality seems to extend to the spin dynamics.

Clearly there is a significant difference in the electronic and magnetic behavior of $(\text{Sr}_{1-x}\text{La}_x)_2\text{Ir}_2\text{O}_7$ compared to its single layer counterpart $(\text{Sr}_{1-x}\text{La}_x)_2\text{IrO}_4$. What the data presented here show is that structural, electronic, and magnetic degrees of freedom are directly coupled in $(\text{Sr}_{1-x}\text{La}_x)_2\text{Ir}_2\text{O}_7$, giving rise to a rich phase diagram, and potentially containing a hidden magnetic QCP. A number of outstanding questions remain. The first is the nature of the apparent crossover between the quantum paramagnetic and quantum critical regimes. This can only be definitively answered through collection of more data — both in the elastic and inelastic channels — between 100 K and 200 K. This in turn leads to whether the $E/T^*$ scaling is universal as a function of temperature and wavevector. If so, then this would imply a departure from the HMM picture, which is only expected to be relevant in the vicinity of $Q_{\text{AF}}$. It is also curious that the nominally three-dimensional $(\text{Sr}_{1-x}\text{La}_x)_2\text{Ir}_2\text{O}_7$ appears to map onto the 2+1 dimensional QNLσM, yet the single-layer $(\text{Sr}_{1-x}\text{La}_x)_2\text{IrO}_4$ does not (for low doping at any rate). This may be related to the electronic phase separation prevalent in the single layer compound above the MIT. Nevertheless, it may be worth revisiting $(\text{Sr}_{1-x}\text{La}_x)_2\text{IrO}_4$ to look for evidence of quantum criticality in the metallic phase.

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