

## Giant Electron-Electron Scattering in the Fermi-Liquid State of $\text{Na}_{0.7}\text{CoO}_2$

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The in-plane resistivity  $\rho$  and thermal conductivity  $\kappa$  of single crystal  $\text{Na}_{0.7}\text{CoO}_2$  were measured down to 40 mK. Verification of the Wiedemann-Franz law,  $\kappa/T = L_0/\rho$  as  $T \rightarrow 0$ , and observation of a  $T^2$  dependence of  $\rho$  at low temperature establish the existence of a well-defined Fermi-liquid state. The measured value of coefficient  $A$  reveals enormous electron-electron scattering, characterized by the largest Kadowaki-Woods ratio  $A/\gamma^2$  encountered in any material. The rapid suppression of  $A$  with magnetic field suggests a possible proximity to a magnetic quantum critical point. We also speculate on the possible role of magnetic frustration and proximity to a Mott insulator.

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Electron behavior in the layered cobaltate  $\text{Na}_x\text{CoO}_2$  shows evidence of strong electron-electron correlations: the specific heat at low temperature points to a significant mass enhancement [1]; the thermopower is 10 times larger than that of typical metals at 300 K [2,3]; the material can be made superconducting by intercalation with water [4]. These and other observations have stimulated extensive interest in this material and a quest for possible new electronic phases [5].

A fundamental question is whether these strong electron correlations can be captured by the standard model of metals, namely, a Fermi-liquid (FL) description of the ground state and low-energy excitations. A FL description is found to be generally valid for heavy-fermion materials, for example, even though correlations in these systems lead to a huge renormalization of the electron effective mass. On the other hand, such a description is *not* generally valid for cuprates, except at the highest carrier concentrations. In this Letter, we report on two tests of FL theory applied to  $\text{Na}_x\text{CoO}_2$ . The first is a test of the Wiedemann-Franz (WF) law, which determines whether the delocalized fermionic excitations of the system carry charge  $e$  and are therefore the usual Landau quasiparticles. The second is a measurement of electrical resistivity at low temperature, which looks at the lifetime of these quasiparticles and determines whether the electron-electron scattering rate varies as  $T^2$ . We find that the WF law is satisfied and we observe a clear  $T^2$  regime in the resistivity:  $\Delta\rho = \rho - \rho_0 = AT^2$ . What is striking is the huge value of the coefficient  $A$ . When normalized by the quasiparticle effective mass, it is 2 orders of magnitude larger than in heavy-fermion materials, such that the Kadowaki-Woods ratio reaches an unprecedented value:  $A/\gamma^2 \simeq 600 \mu\Omega \text{ cm mol}^2 \text{ K}^2/\text{J}^2$ .

$\text{Na}_x\text{CoO}_2$  has a hexagonal layered structure consisting of stacked two-dimensional  $\text{CoO}_2$  planes separated by spacer layers of  $\text{Na}^+$  ions. The Co ions in each  $\text{CoO}_2$  plane are arranged on a triangular lattice. In the undoped

$\text{CoO}_2$  (without Na), each Co atom is in the  $\text{Co}^{4+}$  valence state with spin 1/2, and the material is speculated to be a Mott insulator. Because of the triangular geometry, those spins are frustrated. With Na doping, each dopant atom contributes one electron, changing  $\text{Co}^{4+}$  to a spinless  $\text{Co}^{3+}$  state. The effect of doping is to modify spin correlations and introduce mobile charge carriers, just as in cuprates, but also relax magnetic frustration, so that a rich interplay of spin and charge degrees of freedom is expected.

Single crystals of  $\text{Na}_x\text{CoO}_2$  were grown from NaCl flux according to a procedure described elsewhere [6]. The Na concentration was determined to be  $x = 0.7$  from a measurement of the  $c$ -axis lattice parameter (where  $c = 10.94 \text{ \AA}$ ), using the calibration in Ref. [7]. Two samples, A and B, were cut to rectangular shapes with  $\sim\text{mm}$  dimensions in the  $ab$  plane and  $20\text{--}50 \mu\text{m}$  along the  $c$  axis. Contacts were made with silver epoxy, diffused at  $500^\circ\text{C}$  for 1 h, and were used to measure both electrical resistivity  $\rho(T)$  and thermal conductivity  $\kappa(T)$  in a dilution refrigerator down to 40 mK. The contact resistance was typically  $\approx 0.1 \Omega$  at low temperature. The thermal conductivity was measured using a standard four-wire steady-state method with two  $\text{RuO}_2$  chip thermometers calibrated *in situ* against a reference Ge thermometer. Currents were made to flow in the  $ab$  plane and the magnetic field was applied parallel to the current direction.

In Fig. 1, we show the temperature dependence of the in-plane conductivity below 1.2 K, both electrical, plotted as  $L_0/\rho(T)$  using the Lorenz number  $L_0$  (see below), and thermal, plotted as  $\kappa(T)/T$ , in a magnetic field  $H = 0$  and 10.5 T. There are two contributions to thermal conduction, coming, respectively, from electrons and phonons. In the limit of electrons being scattered predominantly by defects (e.g., Na impurities), the former will be linear in  $T$ . In the limit of phonons being scattered predominantly by electrons, the usual case for metals, phonon conduction is

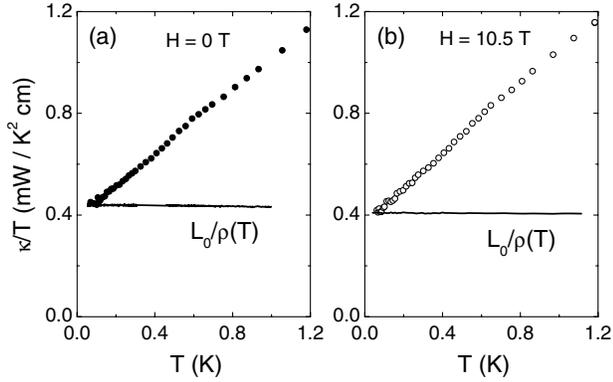


FIG. 1. Temperature dependence of the in-plane thermal conductivity, plotted as  $\kappa(T)/T$ , and the in-plane electrical conductivity, plotted as  $L_0/\rho(T)$ , for sample A at two values of a magnetic field applied parallel to the current: (a)  $H = 0$ , and (b) 10.5 T.  $L_0 = (\pi^2/3)(k_B/e)^2$ . The quantitative convergence of the two conductivities shows that the Wiedemann-Franz law is satisfied. Note that the roughly linear increase in  $\kappa(T)/T$  is due to phonon conduction.

quadratic in  $T$ . This is indeed what is observed in  $\text{Na}_{0.7}\text{CoO}_2$  below 1 K or so:  $\kappa = \alpha T + \beta T^2$ . Irrespective of the particular temperature dependence, by taking the limit of  $\kappa(T)/T$  as  $T \rightarrow 0$  one reliably obtains the purely electronic contribution at  $T = 0$ . In the most general terms, this residual linear term in the thermal conductivity,  $\kappa_0/T$ , is the entropy transport by the delocalized fermionic excitations of the system. In a Fermi liquid,  $\kappa_0/T$  is directly related to the residual electrical resistivity,  $\rho_0 = 55.8 \mu\Omega \text{ cm}$  for 0 T, via the WF law:  $\kappa_0/T = L_0/\rho_0$ , where  $L_0 \equiv (\pi^2/3)(k_B/e)^2 = 2.45 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$  is a universal constant. Fundamentally, the WF law says that Landau quasiparticles carry heat and charge with strictly identical abilities, when no energy is lost through collisions. One can see from Fig. 1 that the WF law is satisfied in  $\text{Na}_{0.7}\text{CoO}_2$ , both at  $H = 0$  and  $H = 10.5$  T.

In this particular sense,  $\text{Na}_x\text{CoO}_2$  is like all other metals but one. Indeed, the WF law has never been violated except in one instance: in the normal state of cuprate superconductor  $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$  at optimal doping ( $x = 0.15$ ) [8]. However, at higher doping (in the overdoped regime), cuprates are found to also obey the WF law [9,10]. This does not mean that the electron behavior in  $\text{Na}_x\text{CoO}_2$  necessarily conforms to FL theory. A basic property of FL theory is that the electron-electron (or rather quasiparticle-quasiparticle) scattering rate should grow as  $T^2$ . In the absence of any other inelastic scattering (such as electron-phonon), this implies that the resistivity should have the form  $\rho = \rho_0 + AT^2$ . In the overdoped cuprate Tl-2201 with a carrier concentration of 0.26 holes per Cu atom, the WF law is very accurately obeyed but there is a prominent non-Fermi-liquid-like linear term in the resistivity at low temperature:  $\rho = \rho_0 + aT + bT^2$  [9]. By going to the highest achievable concentration of

0.3 holes per Cu atom in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , Nakamae *et al.* [10] were able to show that cuprates do eventually show a purely quadratic dependence:  $\rho = \rho_0 + AT^2$ .

The in-plane resistivity  $\rho(T)$  of  $\text{Na}_{0.7}\text{CoO}_2$  at low temperature is shown in Fig. 2 for different values of the magnetic field, plotted as  $\rho - \rho_{\text{offset}}$  vs  $T^2$ . A  $T^2$  regime is clearly observed for all fields, below a crossover temperature  $T_0$  that grows with field. The field dependence of  $T_0$  and  $A$  is shown in Fig. 3;  $T_0$  goes roughly linearly from  $T_0 = 1$  K at  $H = 0$  to  $T_0 = 4$  K at  $H = 16$  T while  $A$  decreases from 0.96 to 0.22  $\mu\Omega \text{ cm K}^{-2}$ . (The low value of  $T_0$  explains why the  $T^2$  dependence had not been seen in previous studies [11,12].) The observed relation  $\Delta\rho = AT^2$  indicates that the behavior of electrons in this system is well described by FL theory. The remarkable aspect is that the magnitude of the electron-electron scattering is enormous: in zero field,  $A = 1.0 \mu\Omega \text{ cm K}^{-2}$ . This is as large as in heavy-fermion systems, where the strong quasiparticle-quasiparticle scattering is due to the enormous density of states at the Fermi energy, as measured by the residual linear term in the specific heat,  $\gamma = C/T$  as  $T \rightarrow 0$ , or equivalently the huge effective mass,  $m^*$ , as measured, for example, by the de Haas-van Alphen effect. In these materials,  $A$  is found to be roughly proportional to  $\gamma^2$ . In  $\text{UPt}_3$ , for example, the relation  $A \propto \gamma^2$  holds very well as a function of pressure [13]. In fact quite generally, the ratio

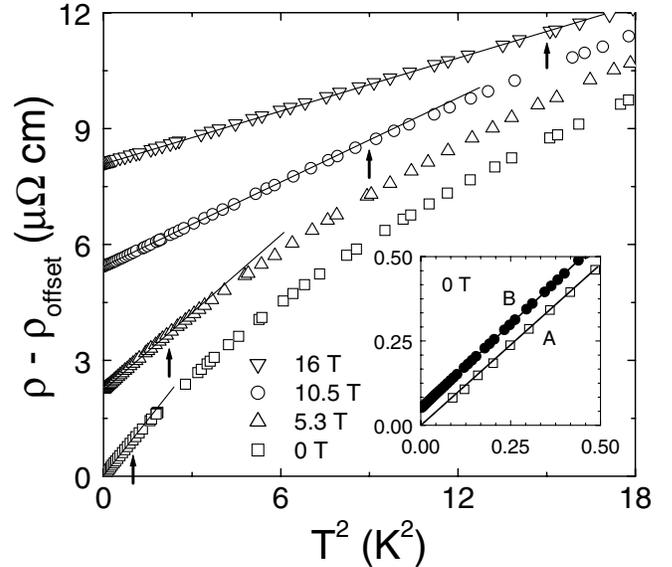


FIG. 2. Temperature dependence of the electrical resistivity at low temperature, plotted as  $\rho(T) - \rho_{\text{offset}}$  vs  $T^2$ , for sample A at several magnetic fields:  $H = 0, 5.3, 10.5$ , and 16 T.  $\rho_{\text{offset}}$  is a constant arbitrary offset chosen for clarity of display. The solid lines are linear fits to the data in a range below a temperature  $T_0$  indicated by arrows. The slope of these lines is the inelastic electron-electron scattering coefficient  $A$ , plotted vs field in Fig. 3. Inset: The zero-field data at the lowest temperatures. Measurements on a second sample (B) show the  $T^2$  behavior down to 50 mK.

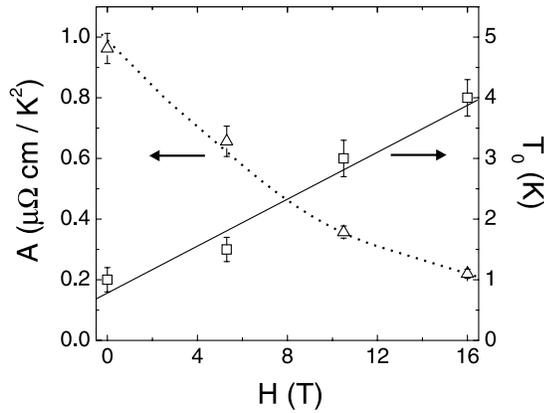


FIG. 3. Field dependence of the  $T^2$  coefficient  $A$  (in  $\Delta\rho = AT^2$ ), and the upper limit of the  $T^2$  range,  $T_0$  (arrows in Fig. 2), for sample A. Lines are guides for the eye.

$r_{\text{KW}} \equiv A/\gamma^2$ , known as the Kadowaki-Woods ratio, has been shown to have a nearly universal value of about  $10 \mu\Omega \text{ cm mol}^2 \text{ K}^2/\text{J}^2 \equiv a_0$  in heavy-fermion systems [14,15] (where  $\gamma$  is measured per mole of magnetic ion).

In Fig. 4, a log-log ("Kadowaki-Woods") plot of  $A$  vs  $\gamma$  is reproduced for a number of materials. In such a plot, it is important to take into account the effect of anisotropy, since  $A$  is in general dependent on the current direction

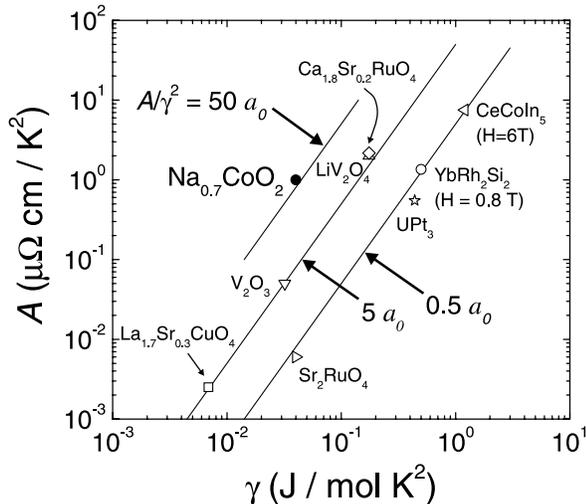


FIG. 4. Kadowaki-Woods plot of coefficient  $A$  (in  $\Delta\rho = AT^2$ ) vs  $\gamma$ , the residual linear term in the specific heat ( $\gamma = C/T$  as  $T \rightarrow 0$ ), for a number of metals. The three lines are lines of constant Kadowaki-Woods ratio  $r_{\text{KW}} = A/\gamma^2$ , for values of 0.5, 5, and  $50a_0$ , as indicated. The first line at  $0.5a_0$  is characteristic of heavy-fermion materials and also accounts for the quasi-2D Fermi-liquid  $\text{Sr}_2\text{RuO}_4$ ; the second line at  $5a_0$  corresponds to the highest values observed until now (typically in systems with magnetic frustration or close to a Mott insulator); the third line at  $50a_0$  shows the order-of-magnitude larger value found in  $\text{Na}_{0.7}\text{CoO}_2$ . (The data used in this plot are referenced in the text.) The data for  $\text{YbRh}_2\text{Si}_2$  are for a sample doped with 5% Ge, for which the QCP is pushed to very low fields (see text).

whereas  $\gamma$  is an average over all directions on the Fermi surface. In the 3D metal  $\text{UPt}_3$ , for example, that hexagonal crystal structure leads to a mass tensor anisotropy of 2.7 which is reflected in the conductivity of the FL regime (below  $T_0 \approx 1.5$  K), where one finds  $A = 0.55(1.55) \mu\Omega \text{ cm K}^{-2}$  for a current parallel (perpendicular) to the hexagonal  $c$  axis [13]. In Fig. 4, we use the lower value (direction of maximum conductivity) and  $\gamma = 0.44 \text{ J/mol K}^2$  [13], so that  $r_{\text{KW}} = 0.28a_0$ . In quasi-2D systems, transport anisotropy can be much more extreme. The best characterized example of a Fermi-liquid state with quasi-2D conductivity is the layered ruthenate  $\text{Sr}_2\text{RuO}_4$  [16]. Conduction perpendicular to the  $\text{RuO}_2$  planes is coherent only at low temperature, and the mass tensor anisotropy of the Fermi surface is 1000 or so. This gives rise to a FL regime below  $T_0 \approx 20$  K with  $A \approx 0.006(6) \mu\Omega \text{ cm K}^{-2}$ , for in-plane (out-of-plane) transport [16]. Given that the quasiparticle states essentially all have wave vectors in the plane, it only makes sense to compute a Kadowaki-Woods ratio for in-plane transport: with  $\gamma = 0.04 \text{ J/mol K}^2$  [16], we get  $r_{\text{KW}} \approx 0.4a_0$ .

The striking fact about the quasi-2D cobalt oxide  $\text{Na}_x\text{CoO}_2$  is that although it has the same  $\gamma$  as the quasi-2D ruthenium oxide, its in-plane  $A$  coefficient is 2 orders of magnitude larger, i.e., of the same magnitude as in heavy-fermion systems. Indeed in  $\text{Na}_{0.7}\text{CoO}_2$ ,  $\gamma = 0.04 \text{ J/mol K}^2$  (per mole of Co) [11], so that the Kadowaki-Woods ratio is more than 100 times larger than in  $\text{Sr}_2\text{RuO}_4$ :  $r_{\text{KW}} \approx 60a_0$ . This reveals that the strong electron correlations in  $\text{Na}_x\text{CoO}_2$  are responsible for enhancing not so much the effective mass of quasiparticles as their scattering rate. Enhanced values of  $r_{\text{KW}}$  beyond the typical value of  $10 \mu\Omega \text{ cm mol}^2 \text{ K}^2/\text{J}^2$  have been seen in a few cases. We now consider these examples to suggest possible mechanisms for the huge  $r_{\text{KW}}$  value in  $\text{Na}_x\text{CoO}_2$ .

The first possible mechanism is proximity to a quantum critical point (QCP). In the heavy-fermion material  $\text{YbRh}_2\text{Si}_2$ , a magnetic-field-induced QCP occurs when antiferromagnetic order is suppressed by applying a field greater than a critical field  $H_c$  [17]. This leads to a divergence of both  $A$  and  $\gamma$  as  $H \rightarrow H_c$ , where  $A \sim (H - H_c)^{-\alpha}$ , with power  $\alpha = 1.0$ . The value of  $H_c$  can be made very small (30 mT) by substituting 5% of Si for Ge. In  $\text{YbRh}_2(\text{Si}_{0.95}\text{Ge}_{0.05})_2$  [17],  $r_{\text{KW}}$  is roughly independent of field at large values of the field: it is constant at  $0.54a_0$  for  $(H - H_c)/H_c > 10$ . However, as the field is lowered towards  $H_c$ , a distinct rise in  $r_{\text{KW}}$  is observed, reaching a value of  $2a_0$  at  $(H - H_c)/H_c \approx 1$ . A similar field-tuned QCP is observed in the heavy-fermion material  $\text{CeCoIn}_5$  [18,19], with  $H_c = 5.1$  T and  $\alpha = 4/3$ , where one finds  $A = 7.5(1.0) \mu\Omega \text{ cm/K}^2$  [18] and  $\gamma = 1.2(0.64) \text{ J/mol K}^2$  [19], so that  $r_{\text{KW}} = 0.52(0.24)a_0$  at  $H = 6(9)$  T. So here again a field-induced enhancement of  $r_{\text{KW}}$  is observed as one approaches the QCP. It is interesting to note that a similar effect is observed in  $\text{Na}_{0.7}\text{CoO}_2$ . In Fig. 3, the coefficient  $A$  determined from

data in Fig. 2 is plotted as a function of the magnetic field. While there is no divergence *per se*, a fivefold increase is nevertheless observed as  $H$  goes from 16 to 0 T. Using the specific heat data of Brühwiler *et al.* [11], where  $\gamma = 0.04(0.025)$  J/mol K<sup>2</sup> in  $H = 0(14)$  T, we get  $r_{\text{KW}} = 60(40)a_0$ , at  $H = 0(14)$  T. This therefore suggests that one interpretation of the strong field dependence of  $A$  is a close-by QCP of magnetic nature. In support of this interpretation, there is evidence of a spin density wave transition in  $\text{Na}_x\text{CoO}_2$  at a slightly higher Na concentration, namely,  $x = 3/4$  [20–22], and  $\rho(T)$  below 20 K is indeed steepest at  $x = 0.75$  [7], while  $\gamma$  remains constant between  $x = 0.55$  [1] and 0.7 [11]. Quantum critical behavior has also been theoretically predicted in  $\text{Na}_x\text{CoO}_2$  based on discrepancies between ferromagnetism predicted by density-functional calculations and the measured paramagnetic ground state [23].

A second comparison, to the transition metal oxide  $\text{LiV}_2\text{O}_4$ , is highly suggestive. Both the cubic spinel structure of  $\text{LiV}_2\text{O}_4$ , in which V ions lie on a sublattice of corner-sharing tetrahedra, and the layered structure of  $\text{Na}_x\text{CoO}_2$ , in which Co ions lie on a two-dimensional triangular lattice, give rise to strong magnetic frustration. The extraordinarily heavy mass observed in the FL state of  $\text{LiV}_2\text{O}_4$ —characterized by  $\gamma \approx 0.175$  J/mol K<sup>2</sup> (per mole of V),  $A = 2.0 \mu\Omega \text{ cm/K}^2$  and  $T_0 \approx 1.5$  K [24]—has been attributed to geometric frustration [24,25]. Although this has not been emphasized, this material has also held the record for the largest Kadowaki-Woods ratio until now:  $r_{\text{KW}} = 6.5a_0$ . This is 1 order of magnitude larger than in typical heavy-fermion materials—like  $\text{YbRh}_2\text{Si}_2(0.54)$ ,  $\text{UPt}_3(0.28)$ , and  $\text{CeCoIn}_5(0.24)$ —but still 1 order of magnitude *smaller* than in  $\text{Na}_x\text{CoO}_2$ . The latter discrepancy might have to do with the different dimensionalities of the electron system in these two oxides: 3D in  $\text{LiV}_2\text{O}_4$ , 2D in  $\text{Na}_x\text{CoO}_2$ .

Finally, we mention a third instance of anomalously large Kadowaki-Woods ratios: proximity to a Mott insulator. The transition metal oxide  $\text{V}_2\text{O}_3$  is close to a metal-insulator transition and it has  $\gamma \approx 0.032$  J/mol K<sup>2</sup> (per mole of V) and  $A = 0.05 \mu\Omega \text{ cm/K}^2$ , so that  $r_{\text{KW}} \approx 5a_0$  [15]. Replacing Sr by Ca in  $\text{Sr}_2\text{RuO}_4$  produces a Mott insulator. By gradually replacing Sr, Nakatsuji *et al.* have shown how  $r_{\text{KW}}$  in  $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$  goes from  $0.4a_0$  at  $x = 2$ , to  $a_0$  at  $x = 0.5$ , to a value as large as  $7a_0$  at  $x = 0.2$  [26]. For  $x < 0.5$ , a structural transition appears to induce a Mott gap on part of the Fermi surface, with concomitant  $S = 1/2$  localized moments, as evidenced by a decrease of  $\gamma$  with decreasing  $x$ . The authors interpret the large enhancement of  $A$  near the Mott insulator as a combination of the narrowing of the remaining conduction band and the additional scattering of conduction electrons on these (antiferromagnetically coupled) localized moments [26]. In this respect, we note that cuprates, the most infamous doped Mott insulators, also exhibit anomalously large  $r_{\text{KW}}$ . Specifically, in overdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $x = 0.3$ , the FL state is characterized

by  $\gamma \approx 7$  mJ/mol K<sup>2</sup>,  $A \approx 2.5$  n $\Omega \text{ cm/K}^2$ , and  $T_0 \approx 1.5$  K [10], so that  $r_{\text{KW}} \approx 5a_0$ . Although this doping value ( $x = 0.3$ ) is not usually thought to be close to the Mott insulator ( $x = 0$ ),  $r_{\text{KW}}$  is still 10 times larger than in the isostructural material  $\text{Sr}_2\text{RuO}_4$ . (Note that in none of these cases is geometric frustration an issue.)

In conclusion,  $\text{Na}_x\text{CoO}_2$  with  $x = 0.7$  adopts a Fermi-liquid state at low temperature that is characterized by the largest Kadowaki-Woods ratio ever observed. Comparison with other materials suggests that the unprecedented magnitude of the electron-electron scattering is due either to magnetic frustration or to the proximity of a nearby magnetic quantum critical point or a Mott insulator. Theoretical exploration of the impact of magnetic frustration on electron scattering, including the role of magnetic field and reduced dimensionality, would be most useful.

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