Pressure-Induced Non-Fermi-Liquid Behavior of $PrNiO_3$

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Comprehensive temperature scans of the resistivity of a high-quality sample of $PrNiO_3$ were made under different pressures up to 30 kbar; they have revealed that the insulator phase is suppressed completely at $P \approx 13$ kbar, transforming to a non-Fermi-liquid phase in which the resistivity varies as $\Delta\rho = \rho(T) - \rho_0 \sim T^n$ with $n = 1.33$ and 1.60 over a broad pressure range.

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The σ-bonding electrons of the $RNiO_3$ perovskites undergo evolution from an enhanced Pauli paramagnetism in metallic $LaNiO_3$ to an antiferromagnetic insulator with Curie-Weiss paramagnetism in $SmNiO_3$ as the ionic radius of the R^{3+} ion decreases [1,2]. As the R^{3+}-ion radius increases from Lu to Sm, the Néel temperature T_N increases and a first-order insulator-metal (IM) transition temperature T_{IM} decreases until $T_N = T_{IM}$ in $NdNiO_3$ and $PrNiO_3$. There is no anomaly in the temperature dependence of the paramagnetic susceptibility on crossing $T_{IM} > T_N$ [2]; the Ni-O bond length on the insulator side of T_{IM} is intermediate between the metallic and the ionic equilibrium Ni-O bond lengths [3]. These characteristics of the IM transition are in sharp contrast to those found at the Mott transition in V_2O_3, for example [4]. In the phase diagram of the $RNiO_3$ family, T_{IM} falls to zero at the composition $x \approx 0.5$ of $La_{1-x}Pr_xNiO_3$ [5]. Physical properties associated with an order-disorder transition, even if it is first order, may develop some quantum critical characteristics as the transition temperature is reduced to near zero by a variation of parameters such as chemical composition, pressure, or an applied magnetic field [6]. New ground states commonly develop on the disorder side of the transition in the vicinity of a quantum critical point (QCP). Critical spin fluctuations at the QCP of a magnetic transition can result in non-Fermi-liquid (NFL) behavior [7–9] or even mediate superconductive pairing [10]. Previous measurements of $\rho(T)$ on $PrNiO_3$ [11–13] show that the IM transition and hysteresis loop vanish at $P_c \approx 13$ kbar. However, poor data quality due to porous samples prevents an adequate characterization of the metallic phase at $P >$ 13 kbar. The high-quality sample used in this work and an enhanced capacity of our piston-cylinder device allowed us to explore much more accurately the metallic phase in the vicinity of P_c and, more importantly, how the metallic phase evolves as pressure increases far beyond P_c. We have also made a comparison between the metallic phase of $PrNiO_3$ at highest pressure with $LaNiO_3$. Almost all NFL phases reported are formed in the vicinity of critical parameters (P_c, x_c, and H_c) where a magnetic transition in a Fermi-liquid (FL) metallic phase is suppressed. We investigate whether the transition from the antiferromagnetic insulator to the metallic phase in $PrNiO_3$ ends, under pressure, in a NFL phase and whether the NFL phase is the same or different from those previously reported for other NFL systems.

High-quality $RNiO_3$ samples have been fabricated by the cold-press technique described in a previous paper [14]. Cu wires were pressed onto the sample on small pieces of In foil. The contact resistance is less than 1 Ω. The high-pressure experiments were performed with a self-clamped Cu-Be cell to 3 GPa [13]. For comparison with results obtained by others on porous samples, we highlight a few remarkable aspects of the $\rho(T)$ curves of Fig. 1 on these high-quality samples: (1) The magnitude of $\rho(300\ \mathrm{K})$ of the $RNiO_3$ samples $R =$ La, Pr, Nd decreases with increasing Ni-O-Ni bond angle θ [3]. (2) The $\rho(300\ \mathrm{K})$ is significantly lower than has been reported in the literature, including that of a single-crystal $NdNiO_3$ film [15] shown in Fig. 1. (3) The residual resistivity $\rho_0 \approx 6\ \mu\Omega$ cm of the curves is smaller by at least a factor of 4 than the lowest ρ_0 previously reported for $LaNiO_3$ or $PrNiO_3$ under $P > 14$ kbar; moreover, a pressure-independent ρ_0 shows there is no grain-boundary effect

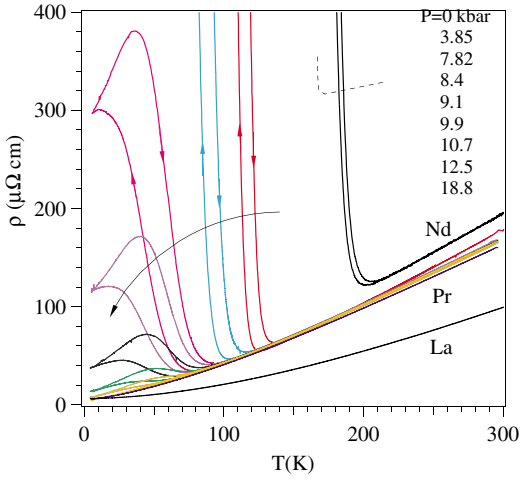

FIG. 1 (color online). Temperature dependence of the resistivity ρ for $RNiO_3$ and $PrNiO_3$ under pressure. The dashed line represents $\rho(T)$ for a $NdNiO_3$ film from Ref. [16].

in these high-quality samples. (4) High ratios $\rho(300\ \mathrm{K})/\rho_0 = 17$ for $LaNiO_3$ and 27 for $PrNiO_3$ under $P > P_c$ compares with reported ratios 4 to 9.5 for $LaNiO_3$ and 20 for $PrNiO_3$ under pressure, respectively. The analysis of the $\rho(T)$ leads us to the conclusion that a NFL phase is achieved in $PrNiO_3$ as the insulator phase is suppressed under pressure.

Figure 1 includes a succession of $\rho(T)$ curves for $PrNiO_3$ under increasing pressure; $T_{\mathrm{IM}} \approx 130$ K at ambient pressure moves progressively to lower temperature under pressure and the thermal hysteresis loop broadens. Unlike $BaVS_3$ [16] where the IM transition remains sharp as T_c is lowered under pressure, the IM transition in $PrNiO_3$ becomes much broader and more difficult to define as T_{IM} falls below 70 K. The hysteresis loop persists to around 50–60 K, but at pressures $P > 10.7$ kbar there is no low-temperature upturn of the resistivity to mark T_{IM}. With the best resolution in our measurement, a homogeneous metallic phase without a hysteresis loop is achieved under $P \geq 13$ kbar. In order to analyze the metallic phase stabilized under pressure, we have applied the power-law fitting to $\rho(T)$ of $PrNiO_3$ under pressure along with that of metallic $LaNiO_3$. Whereas a Fermi-liquid phase, namely, $\Delta\rho(T) = \rho(T) - \rho_0 \sim T^n$ with $n = 2$, is observed in $LaNiO_3$ under ambient pressure, an $n < 2$ has been obtained for the metallic phase of $PrNiO_3$ under pressure. As demonstrated in Fig. 2, the power-law formula with $n = 4/3$ fits the experimental data extremely well for the pressure range 8–20 kbar. The exponent n jumps to about 1.60 for the pressure range 20–30 kbar. The pressure dependence of the exponent n is plotted in Fig. 3 together with the evolution with pressure of the transition temperature T_{IM}. Curve fitting in the metallic phase is made within $5 \leq T \leq 20$ K, but the fitting curve based on this temperature range remains matched to $\rho(T)$ up to $T_{\mathrm{u}} = 100$ K for pressures around 20 kbar; T_u falls gradually to about 50–60 K on further increase of pressure. Significantly, this low-temperature metallic phase does not show a clear trend to a FL phase under the highest pressure of this work. Moreover, the extension to higher pressure of a formula fitting to the resistivity at 285 K versus P (inset of Fig. 3) appears not to meet the $\rho(285\ \mathrm{K})$ of $LaNiO_3$ at any pressures below 60 kbar, whereas a recent structural study [17] under high pressure places the orthorhombic-rhombohedral transition in $PrNiO_3$ at $P_c \approx 50$ kbar. These observations point out that the transition at low temperature from the orthorhombic NFL phase to the rhombohedral FL phase is going to be first order, which means that a FL phase may be realized only in the rhombohedral phase having a broader bandwidth than the orthorhombic phase. However, it is not yet clear at this point whether a first-order transition is required for the electronic state change on crossing the phase boundary between the NFL phase and a FL phase since the orthorhombic to rhombohedral structural transition is always first order in perovskite oxides [18].

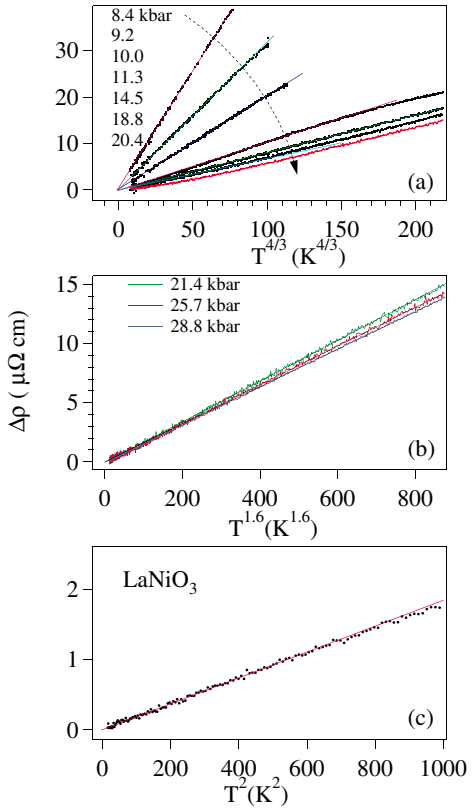

FIG. 2 (color online). (a) ρ versus $T^{4/3}$ for $PrNiO_3$ for $8 < P < 20$ kbar; (b) $\rho(T)$ versus $T^{1.6}$ for $P > 20$ kbar; (c) ρ versus T^2 for $LaNiO_3$ under ambient pressure.

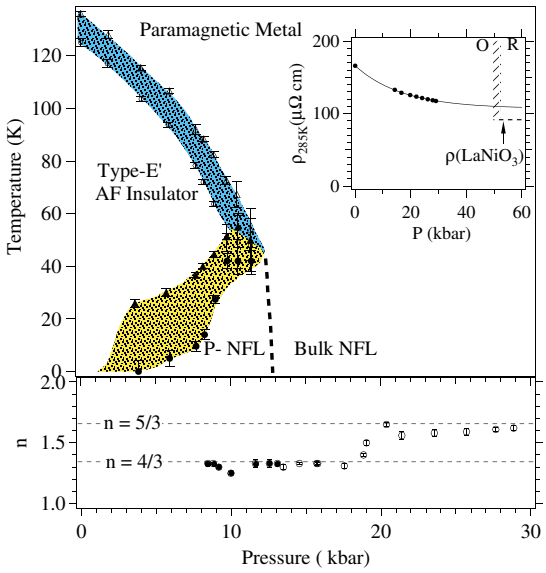

FIG. 3 (color online). The pressure-temperature phase diagram of $PrNiO_3$ and pressure dependence of the exponent n in a $\Delta\rho \sim T^n$. The shadowed area represents the thermal hysteresis in $\rho(T)$. P-NFL stands for the percolated non-Fermi-liquid phase.

As shown in Figs. 1 and 2, the reentrant metallic phase on warming up from 5 K under $8 < P < 10$ kbar can also be fit to a power law with $n \approx 4/3$, marked as solid circles in Fig. 3. It could be argued that the NFL behavior is related to phase inhomogeneity since a huge hysteresis loop is found in $\rho(T)$ under these pressures. However, we emphasize here that the NFL phase is stabilized over a broad range of pressure $13 \leq P < 30$ kbar where no thermal hysteresis of $\rho(T)$ has been observed. Therefore, the NFL behavior has nothing to do with a phase inhomogeneity. Instead, the transport property in the mixed-phase region should be dominated by the more conductive phase, i.e., a percolated connection of the NFL phase. In Fig. 3, the region of a percolated NFL phase is separated by a dashed line near 13 kbar from the bulk NFL at higher pressures.

A $\rho(T)$ following a power law with $n < 2$ at low temperatures has been generally argued to be caused by quantum critical fluctuations, and this behavior has been found in a metal where a spin ordering is suppressed under magnetic field or pressure [7–10]. Millis *et al.* [19] have predicted that quantum critical fluctuations lead to a power law with $n = 4/3$ in a two-dimensional system near the end point of a first-order transition. This prediction fits very well to the lower step $n \approx 4/3$ in Fig. 3 even though $PrNiO_3$ has a 3D perovskite structure. On the other hand, the pressure-temperature phase diagram of Fig. 3 is surprisingly similar to that of a Mott insulator $\kappa - (BEDT - TTF)_2Cu[N(CN)_2]Cl$ under pressure [20]. In this case quantum critical fluctuations near the end point of a Mott transition gives rise to superconductive pairing. On the left-hand side of the critical point in that phase diagram, a percolated superconducting phase has been detected within the insulator phase whereas a percolated NFL phase has been identified below 13 kbar in $PrNiO_3$. Moreover, it is clear from Fig. 3 that the NFL phase found in $PrNiO_3$ is not confined to the vicinity of P_c, but extends to much higher pressures as has been reported for MnSi [21,22] in which the ferromagnetic transition is suppressed at $P_c = 14$ kbar and the NFL remains stable under pressure up to 30 kbar. However, two distinct exponents n that are close to the fractional numbers $4/3$ and $5/3$ distinguish the NFL phase of $PrNiO_3$ from that in MnSi. This interesting feature calls for an experiment at even higher pressure (~ 50 kbar) in order to check whether n approaches $n = 2$ by steps of a fractional number.

As to the nature of the IM transition in $PrNiO_3$, which can be important for understanding the NFL behavior in which it terminates, we have made precise temperature scans of $\rho(T)$ and the thermal conductivity $\kappa(T)$ around the transition and compared them with some well-known transitions in manganites and magnetite. As shown in Fig. 1, the reentrant metallic phase, which is more clearly shown on the warming up loop, develops at a $T < T_{IM}$ with increasing pressure. The merging of two first-order transitions leaves a giant thermal hysteresis loop of $\rho(T)$ and an unusual circling of $\rho(T)$ for the reentrant metallic phase in comparison with that for the metal-insulator transition at T_c in $(La_{0.25}Nd_{0.75})_{0.7}Ca_{0.3}MnO_3$ under 7.5 kbar [23]. Typical $\rho(T)$ curves taken in the pressure range 8–10 kbar are shown in Fig. 4. On warming up from 5 K, $\rho(T)$ is precisely reproducible to that on cooling down—see Fig. 4(c)—as long as the upper bound temperature is below a T_{max} where resistivity shows a maximum on warming. It is within this temperature range that we carried out a curve fitting to the power law.

As a widely used explanation of the IM transition in $RNiO_3$ [24], charge disproportionation or bond-type ordering can be expected to enhance the thermal conductivity $\kappa(T)$ as found at the Verwey transition of Fe_3O_4 in Fig. 5(b) and orbital or charge ordering in $La_{1.875}Sr_{0.125}MnO_3$ [25]. In contrast to an enhancement below a critical temperature, the thermal conductivity of $PrNiO_3$ shown in Fig. 5(a), collapses below T_{IM} even after taking into account the change in the electronic contribution. This observation suggests that even the lattice undergoes critical fluctuations in the insulator phase below a $T_{Im} \leq 130$ K. Alternatively, as indicated in Fig. 3, a collapse of κ in the insulator phase below T_{IM} can be attributed to a two-phase mixture of the magnetic-insulator phase and the NFL phase at ambient pressure. In either case, lattice (charge) and spin should exhibit critical fluctuations near the end point of T_{IM}. These

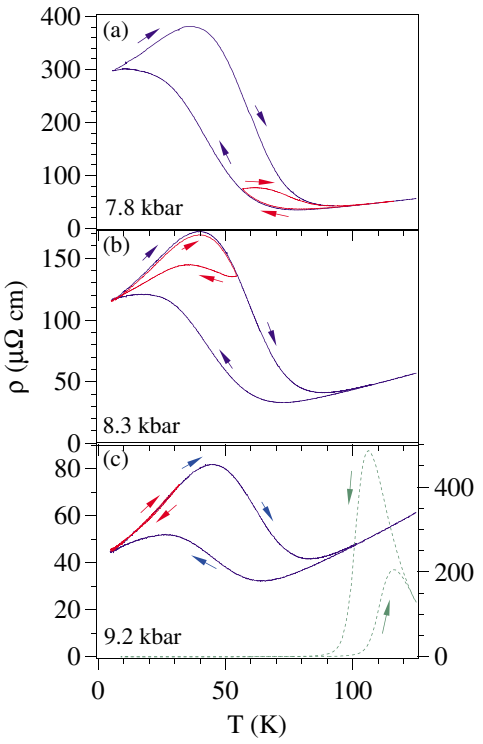

FIG. 4 (color online). $\rho(T)$ of $PrNiO_3$ under different pressures for various temperature-scan loops. Plotted in (c) as a dashed line is the $\rho(T)$ in $(La_{0.25}Nd_{0.75})_{0.7}Ca_{0.3}MnO_3$ under $p = 7.5$ kbar for comparison.

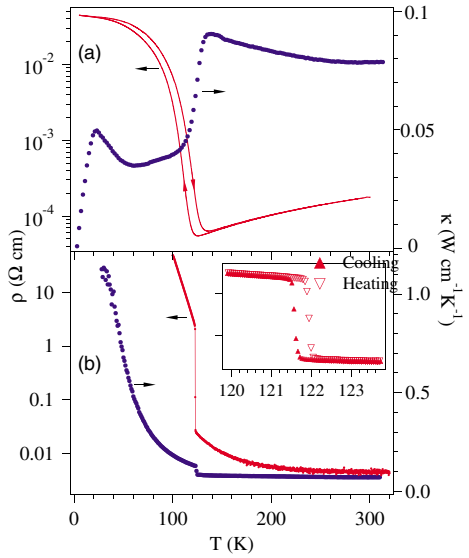

FIG. 5 (color online). Temperature dependence of the resistivity ρ and thermal conductivity κ for (a) $PrNiO_3$ at ambient pressure, and (b) an Fe_3O_4 crystal.

fluctuations are likely to have quantum effects below a Fermi degeneracy temperature as proposed by Imada [26]. Spin and charge fluctuations near an end point of T_{IM} distinguish the NFL phase in $PrNiO_3$ from that found at end points of a magnetic transition in a metal and a Mott transition. On the other hand, separated only by a tiny margin of bandwidth [3], $LaNiO_3$ shows a well-classified Fermi-liquid behavior up to 28 K. The coefficient $A = 1.8 \times 10^{-3}\ \mu\Omega\,\mathrm{cm\,K^{-2}}$ of $\rho(T) = \rho_0 + AT^2$ for $LaNiO_3$ is comparable to that found in other strongly correlated metallic oxides such as $La_{1.7}Sr_{0.3}CuO_4$ and Sr_2RuO_4 [27].

In conclusion, the metallic phase in $PrNiO_3$ at $P >$ 13 kbar where the metal-insulator transition temperature is terminated, has been found not to follow the description of Fermi-liquid theory. A power-law fitting to $\rho(T)$ of this metallic phase gives two distinct exponents, an $n \approx 1.33$ over the pressure range 8–20 kbar and an $n \approx 1.60$ in the range 20–30 kbar. Although a percolated NFL metallic phase may exist in the pressure range $8 < P < 13$ kbar, a bulk NFL phase exists at higher pressures, and the step in the exponent n at 20 kbar remains as a novel feature needing theoretical attention. This non-Fermi-liquid phase appears to be caused by both lattice (charge) and spin fluctuations where the transition temperature $T_{IM} = T_N$ is terminated under pressure. The results also suggest that a Fermi-liquid metallic phase can be achieved in the perovskites $RNiO_3$ family only with rhombohedral symmetry.

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[1] J. B. Torrance, P. Lacorre, A. I. Nazzal, E. J. Ansaldo, and Ch. Nidermayer, Phys. Rev. B **45**, 8209 (1992).
[2] J.-S. Zhou, J. B. Goodenough, B. Dabrowski, P. W. Klamut, and Z. Bukowski, Phys. Rev. Lett. **84**, 526 (2000).
[3] J.-S. Zhou and J. B. Goodenough, Phys. Rev. B **69**, 153105 (2004).
[4] D. B. McWhan, A. Menth, J. P. Remeika, W. F. Brinkman, and T. M. Rice, Phys. Rev. B **7**, 1920 (1973).
[5] M. Medarde, J. Phys. Condens. Matter **9**, 1679 (1997).
[6] S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, 1999).
[7] N. Doiron-Leyraud, I. R. Walker, L. Taillefer, M. J. Steiner, S. R. Julian, and G. G. Lonzarich, Nature (London) **425**, 595 (2003).
[8] J. Custers, P. Gegenwart, H. Wilhelm, K. Neumaler, Y. Toklwa, O. Trovarelli, G. Geibel, F. Steglich, C. Pepin, and P. Coleman, Nature (London) **424**, 524 (2003).
[9] K. H. Kim, N. Harrison, M. Jaime, G. S. Boebinger, and J. A. Mydosh, Phys. Rev. Lett. **91**, 256401 (2003).
[10] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. W. Haselwimmer, and G. G. Lonzarich, Nature (London) **394**, 39 (1998).
[11] X. Obradors, L. M. Paulius, M. B. Maple, J. B. Torrance, A. I. Nazzal, J. Fontcuberta, and X. Granados, Phys. Rev. B **47**, 12 353 (1993).
[12] P. C. Canfield, J. D. Thompson, S.-W. Cheong, and L. W. Rupp, Phys. Rev. B **47**, 12 357 (1993).
[13] J.-S. Zhou, J. B. Goodenough, B. Dabrowski, P. W. Klamut, and Z. Bukowski, Phys. Rev. B **61**, 4401 (2000).
[14] J.-S. Zhou, J. B. Goodenough, and B. Dabrowski, Phys. Rev. B **67**, 020404 (2003).
[15] U. Staub, G. I. Meijer, F. Fauth, R. Allenspach, J. G. Bednorz, J. Karpinski, S. M. Kazakov, L. Paolasini, and F. d'Acapito, Phys. Rev. Lett. **88**, 126402 (2002).
[16] L. Forro, R. Gaal, H. Berger, P. Fazekas, K. Penc, I. Kezsmarki, and G. Mihaly, Phys. Rev. Lett. **85**, 1938 (2000).
[17] J.-S. Zhou, J. B. Goodenough, and B. Dabrowski, Phys. Rev. B **70**, 081102 (2004).
[18] J.-S. Zhou and J. B. Goodenough, Phys. Rev. Lett. **94**, 065501 (2005).
[19] A. J. Millis, A. J. Schofield, G. G. Lonzarich, and S. A. Grigera, Phys. Rev. Lett. **88**, 217204 (2002).
[20] F. Kagawa, T. Itou, K. Miyagawa, and K. Kanoda, Phys. Rev. B **69**, 064511 (2004).
[21] C. Pfleiderer, D. Reznik, L. Pintschovius, H. v. Lohneysen, M. Garst, and A. Rosch, Nature (London) **427**, 227 (2004).
[22] C. Pfleiderer, S. R. Julian, and G. G. Lonzarich, Nature (London) **414**, 427 (2001).
[23] J.-S. Zhou, W. Archibald, and J. B. Goodenough, Nature (London) **381**, 770 (1996).
[24] J. A. Alonso, J. L. Garcia-Munoz, M. T. Fernandez-Diaz, M. A. G. Aranda, M. J. Martinez-Lope, and M. T. Casais, Phys. Rev. Lett. **82**, 3871 (1999).
[25] J.-S. Zhou and J. B. Goodenough, Phys. Rev. B **64**, 024421 (2001).
[26] M. Imada, J. Phys. Soc. Jpn. **73**, 1851 (2004).
[27] S. Nakamae, K. Behnia, N. Mangkorntong, M. Nohara, H. Takagi, S. J. C. Yates, and N. E. Hussey, Phys. Rev. B **68**, 100502(R) (2003).