Possible non-Fermi-liquid behavior of CaRuO$_3$

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We measured the resistivity (ρ) and magnetoresistance ($\Delta\rho/\rho$) of the metallic pseudocubic perovskite CaRuO$_3$. At high temperatures, $\rho = a + bT^{0.5}$ and $\Delta\rho/\rho$ is negative. At low temperatures, $\rho = a + bT^{1.5}$ and $\Delta\rho/\rho$ changes sign as a function of the applied magnetic field. Both at high temperatures and at low temperatures, ρ deviates strongly from the resistivity of Fermi-liquid metals; we therefore discuss the possibility that CaRuO$_3$ is a non-Fermi-liquid metal. [S0163-1829(99)04808-0]

I. INTRODUCTION

One of the central challenges in recent years has been the understanding of transport properties of metals with electronic correlations strong enough to invalidate the applicability of the Fermi-liquid theory.[1] Among these metals, metallic perovskites such as cuprates and more recently ruthenates have attracted particular interest. Here we present transport measurements of CaRuO$_3$ that suggest that this metal might also be a non-Fermi-liquid metal.

CaRuO$_3$ is a pseudocubic metallic perovskite (effective lattice parameter ~ 3.84 Å) with orthorhombic structure[2] ($a = 5.541$ Å, $b = 5.362$ Å, and $c = 7.686$ Å). While CaRuO$_3$ was first studied in the late 1950s,[3,4] an intensive investigation of this compound has started only following the discovery of the high-T_c cuprates. This interest was initially motivated by the potential use of thin films of CaRuO$_3$ in heterostructures with high-T_c films.[5,6] However, more recently the main focus shifted to studying the *physical properties*[7–9] of CaRuO$_3$ as part of a very active investigation of other ruthenates [such as SrRuO$_3$,[10,11] Sr$_2$RuO$_4$,[12] and Ca$_3$Ru$_2$O$_7$ (Ref. 13)] whose intriguing properties are probably driven by strong electron correlations.

Here we report resistivity (ρ) and magnetoresistance measurements of thin films of CaRuO$_3$. We find that at high temperatures $\rho = a + bT^{0.5}$ and that at low temperatures $\rho = a + bT^{1.5}$ (see Fig. 1). This resistivity (in both limits) is very different from the resistivity expected for Fermi-liquid metals. Therefore, we address the possibility that CaRuO$_3$ is a non-Fermi-liquid metal.

For this study we have used thin films of CaRuO$_3$ prepared by pulsed laser deposition on substrates of LaAlO$_3$ and SrTiO$_3$. The room-temperature resistivity of our films is typically ~ 500 $\mu\Omega$ cm and their resistivity ratio (ρ_{273}/ρ_0) is typically ~ 6.

II. MEASUREMENTS

We start by examining the zero-field resistivity of CaRuO$_3$. Figure 1 shows the rescaled resistivity ($\widetilde{\rho}$) of two different films deposited on LaAlO$_3$, $\widetilde{\rho} = (\rho - \rho_0)/(\rho_{273} - \rho_0)$. Here ρ_{273} is the resistivity at $T = 273$ K and ρ_0 is the extrapolated zero-temperature resistivity. It is important to note the reproducibility of the temperature dependence of the resistivity; this is a clear indication of the intrinsic nature of the resistivity, which we analyze in the following.

The resistivity (ρ) is clearly sublinear at high temperatures, turning to stronger than linear at low temperatures—qualitative features generally observed either in thin films[5] or in bulk samples.[8] For a quantitative analysis we want to fit the data with a particular function. Lacking any *a priori* reason to choose a specific fit, we assume the simplest possible form, $\rho = a + bT^{\alpha}$, and look for the best α.

For fitting the high-temperature resistivity we used data points from the temperature range 50 K$<T<$200 K. We randomly chose 50 data points and calculated the error of the least-squares fit for a range of values $0.4 \leq \alpha \leq 0.6$ with intervals of 0.001. The α with the smallest error was taken as the best fit for the specific set of points chosen. We repeated the process 500 times and the distribution of the values of α is shown in the inset of Fig. 1(b). We see that the distribution of α is extremely narrow: $0.493 \leq \alpha \leq 0.506$. Since $\alpha = 0.5$ is within this narrow range we show in Fig. 1(b) the resistivity as a function of $T^{0.5}$ to demonstrate the quality of the fit at high temperatures.

We repeated the same procedure with the low-temperature data. In the inset of Fig. 1(c) we show the distribution of α resulting from a process where 15 data points were selected 500 times from a temperature range of 2 K $<T<$10 K. We find that $1.41 \leq \alpha \leq 1.61$. Removing the upper and lower 5% of the values of α already cuts the interval to $1.45 \leq \alpha \leq 1.53$. In Fig. 1(c) we show the resistivity as a function of $T^{1.5}$. (We would like to comment here that in some films the zero-field resistivity becomes qualitatively different below $T<3.5$ K in a way indicative of a first-order phase transition. We will discuss this behavior elsewhere.)

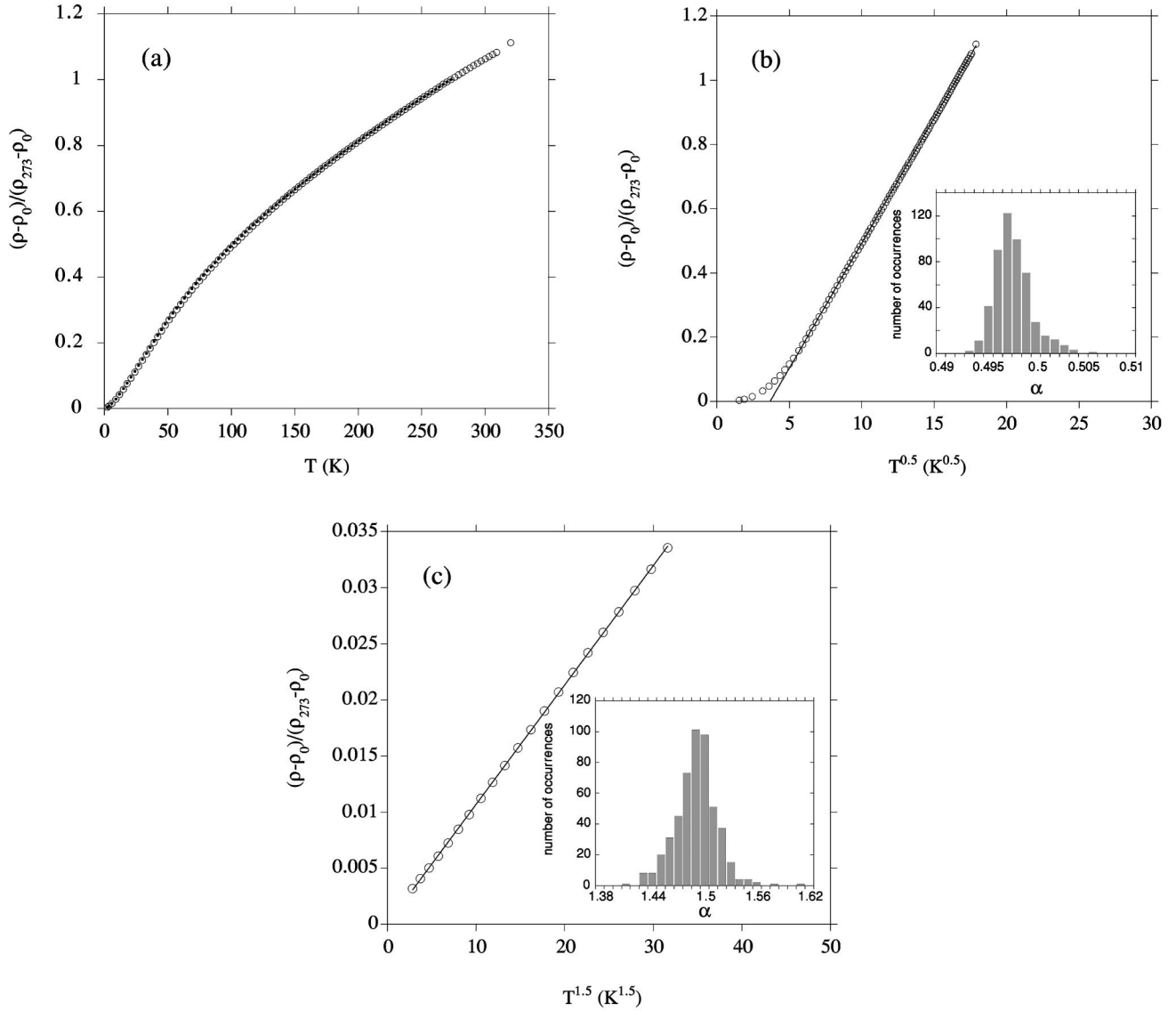The crossover between the high-temperature and the low-temperature resistivity is very gradual: the high-temperature

FIG. 1. (a) The rescaled zero-field resistivity of two different $CaRuO_3$ films (~ 1000 Å thick) on $LaAlO_3$ substrates. (b) The rescaled resistivity vs $T^{0.5}$. Inset: The distribution of the power α which yields the best fit in the form $\rho = a + bT^{\alpha}$, to a randomly selected subset (50 points) of the resistivity data in the temperature interval 50 K$<T<$200 K. The total number of iterations is 500. (c) The rescaled resistivity vs $T^{1.5}$. Inset: The distribution of the power α, which yields the best fit in the form $\rho = a + bT^{\alpha}$, to a randomly selected subset (15 points) of the resistivity data in the temperature interval 2 K$<T<$10 K. The total number of iterations is 500.

fit breaks down below $T \sim 40$ K, while the low-temperature fit breaks down above $T \sim 15$ K. More insight into the nature of this crossover is obtained with magnetoresistance (MR) measurements that are discussed next and which show that changes in the temperature dependence of the resistivity are accompanied by qualitative changes in the MR.

Figure 2(a) shows the MR ($[\rho(H) - \rho(0)]/\rho(0)$) with field parallel to the current at three different temperatures: $T = 5$ K, $T = 20$ K, and $T = 50$ K. Figure 2(b) shows the MR at fields (parallel to the current) $H = 2,4,6$ T and temperatures 5 K$\leqslant T \leqslant$100 K where the data have been divided by H^2. While the MR is overall very small we can clearly observe quantitative changes in its properties. At high temperatures the MR is negative and it increases in magnitude as the temperature decreases, reaching its peak at $T \sim 15$ K. At lower temperatures the MR decreases in magni-

tude becoming positive above a threshold field that decreases as temperature decreases. Thus, for instance, we can see that at $T = 5$ K the differential MR becomes positive above 4 T. The crossover from negative to positive MR affects the functional form of the negative MR well before it actually turns positive. At high temperatures the negative MR is proportional to H^2 as is evident from the coincidence of the MR points at $T \geqslant 40$ K [see Fig. 2(b)]. On the other hand, the splitting of the points at lower temperatures (so that the MR points of the high fields are lower) indicates the MR is weaker than H^2. We see that the deviation from H^2 dependence occurs well above $T = 5$ K where we observe the relative positive MR. We note that the region in which $\rho = a + bT^{0.5}$ overlaps the region in which $\Delta\rho/\rho \propto H^2$. In addition, the region in which $\rho = a + bT^{1.5}$ overlaps the region where $\Delta\rho/\rho$ crosses over from negative to positive.
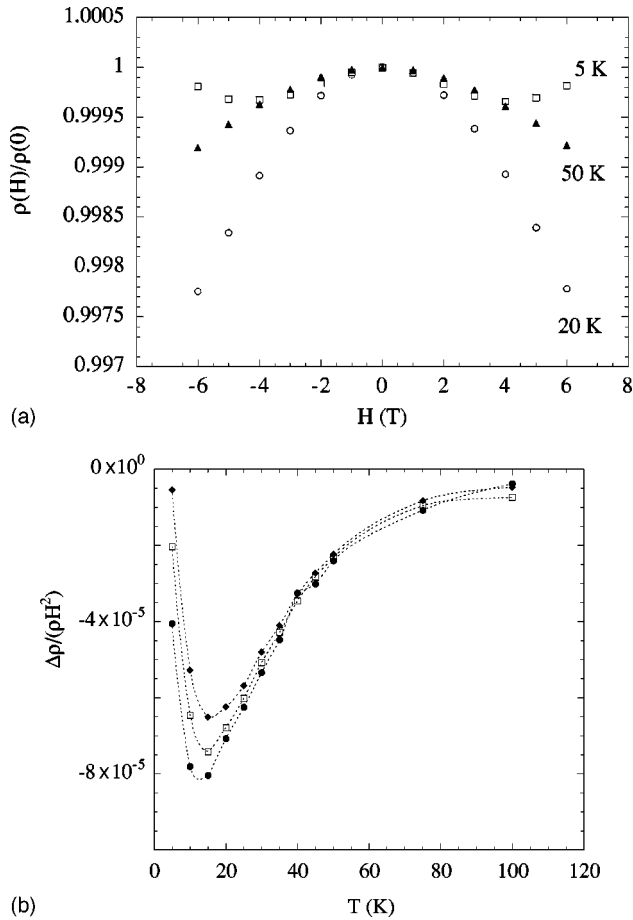
FIG. 2. (a) MR of $CaRuO_3$ at $T = 5$ K, 20 K, and 50 K. Note the change in the differential MR at $T = 5$ K and $H \sim 4$ T. (b) The temperature dependence of the MR of $CaRuO_3$ at $H = 2,4,6$ T divided by H^2 (in teslas).

III. DISCUSSION

The resistivity and MR measurements identify two different regions: (a) Region A: a high-temperature region ($T \geqslant 40$ K) characterized by resistivity whose temperature dependence is $\rho = a + bT^{0.5}$ and MR that is negative and proportional to H^2. (b) Region B: a low-temperature region ($T \leqslant 15$ K) characterized by low-field resistivity whose temperature dependence is $\rho = a + bT^{1.5}$. The MR in region B is complicated: turning from negative at high temperatures to positive at low temperatures. Therefore, it is possible to mark a crossover line within region B that will separate the region where $d\rho/dH > 0$ from the region where $d\rho/dH < 0$.

Region A

The temperature dependence of the resistivity in this region is quite puzzling. Clearly, the resistivity $\rho = a + bT^{0.5}$ deviates from the Bloch-Grüneisen resistivity expected to be linear at high temperatures. On the other hand, the resistivity of $CaRuO_3$ may not be so peculiar if it results from ''saturation.''[14] ''Saturation'' is an effect observed in different compounds, such as A15: when the mean-free path approaches the Ioffe-Regel limit $k_F l = \mathcal{O}(1)$, there is deviation from the (high-temperature) linear resistivity and the resistivity becomes sublinear as if it is limited by a saturation

value that roughly corresponds to the mean-free path given by the Ioffe-Regel limit. Here l is the mean-free path and k_F is the Fermi wave vector. This possibility is not excluded; however, since there are no expectations for a particular form of saturation, it is doubtful that the narrow distribution of α, which includes $\alpha = 0.5$, is purely accidental. An alternative approach is to assume that the power $\alpha = 0.5$ is not accidental and examine the applicability of models that specifically predict $\rho = a + bT^{0.5}$. Such a model is the two-channel Kondo model, which is believed to describe correctly the non-Fermi-liquid resistivity in the form of $\rho = a + bT^{0.5}$ observed in uranium-based and cerium based compounds.[15] Currently, we cannot confirm or exclude the relevance of this model to the unusual resistivity of $CaRuO_3$. Nevertheless, we believe that it is more likely that the high-temperature resistivity of $CaRuO_3$ reflects a non-Fermi-liquid behavior rather than ''saturation.''

The other interesting behavior in region A is the sign and temperature dependence of the MR. Negative MR is commonly found in magnetic metals—the applied field suppresses the spin fluctuations that contribute to the electrical resistivity, thus yielding negative MR. $CaRuO_3$ is a paramagnetic metal (at least in region A); therefore, one may expect spin contribution to resistivity and hence negative MR. The qualitative temperature dependence of the MR is not surprising; the susceptibility increases as temperature decreases; therefore, both the induced magnetic ordering and the MR (in its absolute value) increase. The unexpected behavior of the MR occurs in the intermediate range between region A and region B; the MR is no longer proportional to H^2, and the MR reaches a *maximum* at $T \sim 15$ K. We believe that the deviation from the H^2 dependence and, even more strongly, the maximum in MR (whereas the susceptibility keeps on increasing at lower temperatures) are clear indications that the crossover from region A to region B involves (or is due to) changes in magnetic properties.

Region B

While the low-temperature resistivity of Fermi liquid metals follows $\rho = a + bT^2$, the low-temperature resistivity in this region follows $\rho = a + bT^{1.5}$. This form of non-Fermi-liquid resistivity is theoretically expected when there is a zero-temperature paramagnetic-to-antiferromagnetic quantum phase transition; consequently, it has been suggested as the source of such resistivity observed in, e.g., $CeNi_2Ge_2$.[16] If this is the reason for the unusual resistivity in $CaRuO_3$, it would be theoretically surprising, since band calculations[17,18] indicate the proximity of $CaRuO_3$ to a *ferromagnetic* instability expected to yield a different temperature dependence of ρ. On the other hand, we note that $CaRuO_3$ is the end member of the series consisting of Ca_2RuO_4 and $Ca_3Ru_2O_7$ both of which are antiferromagnetic.[13,19]

The MR in this region is quite complicated. At $T = 5$ K, for instance, it starts negative and then turns positive at higher fields (~ 4 T), while at $T = 10$ K it is negative at least up to $H = 6$ T. This may indicate that region B should be split into two subregions according to the differential MR with a crossover region in between. It is interesting to note again the similarity with $CeNi_2Ge_2$, which also exhibits such a transition between $d\rho/dH < 0$ at high temperatures to

$d\rho/dH < 0$ at low temperatures. In the case of $CeNi_2Ge_2$ the negative MR was correlated with non-Fermi resistivity ($\rho = a + bT^{1.5}$) while positive MR was correlated with Fermi-liquid behavior ($\rho = a + bT^2$). In our case, however, the effective zero-field power (1.5) increases only slightly with the field reaching ~ 1.55 at $H = 6$ T.

The apparent non-Fermi-liquid behavior of $CaRuO_3$ is not very surprising. An important indication for such a possibility is the specific-heat measurements reported by Cao $et\ al.$[8] who measured the specific heat of $Sr_xCa_{1-x}RuO_3$ and found that for $CaRuO_3$ $\gamma = 73$ mJ/mol K^2. Such large γ reflects a large mass enhancement indicating electron correlations that are likely to be even stronger than in the related compound $SrRuO_3$ for which transport[10] and infrared measurements[11]

have demonstrated non-Fermi-liquid behavior. Since non-Fermi-liquid behavior is driven by strong electron correlation we could have expected that $CaRuO_3$ would also be a non-Fermi-liquid metal. Our results strongly suggest that this is the case and should encourage further study of $CaRuO_3$ together with other ruthenates as part of the ongoing effort to understand the intriguing class of non-Fermi-liquid metals.

[1] See, e.g., P. W. Anderson, *The Theory of Superconductivity in the High-T_c Cuprates* (Princeton University Press, Princeton, NJ, 1997), Chap. 3; V. J. Emery and S. A. Kivelson, Phys. Rev. Lett. **74**, 3253 (1995).

[2] M. Shepard, S. McCall, G. Cao, and J. E. Crow, J. Appl. Phys. **81**, 4978 (1997).

[3] J. J. Randall and R. Ward, J. Am. Chem. Soc. **81**, 262 (1959); A. Callaghan, C. W. Moeller, and R. Ward, Inorg. Chem. **5**, 1572 (1966).

[4] R. J. Bouchard and J. L. Gillson, Mater. Res. Bull. **7**, 873 (1972).

[5] K. Char, M. S. Colclough, T. H. Geballe, and K. E. Myers, Appl. Phys. Lett. **62**, 196 (1993).

[6] L. Antognazza, K. Char, and T. H. Geballe, Appl. Phys. Lett. **68**, 1009 (1996).

[7] R. A. Rao, Q. Gan, C. B. Eom, R. J. Cava, Y. Suzuki, J. J. Krajewski, S. C. Gausepohl, and M. Lee, Appl. Phys. Lett. **70**, 3035 (1997).

[8] G. Cao, S. McCall, M. Shepard, and J. E. Crow, Phys. Rev. B **56**, 321 (1997).

[9] G. Cao, S. McCall, J. Bolivar, M. Shepard, F. Freibert, P. Henning, and J. E. Crow, Phys. Rev. B **54**, 15 144 (1996); M. Shepard, G. Cao, S. McCall, F. Freibert, and J. E. Crow, J. Appl. Phys. **79**, 4821 (1996); G. Cao, F. Freibert, and J. E. Crow, *ibid.* **81**, 3884 (1997); S. C. Gausepohl and M. Lee, Phys. Rev. B **54**, 8996 (1996).

[10] L. Klein, J. S. Dodge, C. H. Ahn, G. J. Snyder, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, Phys. Rev. Lett. **77**, 2774 (1996); L. Klein, J. S. Dodge, C. H. Ahn, J. W. Reiner, L. Mieville, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, J. Phys.: Condens. Matter **8**, 10 111 (1996).

[11] P. Kostic, Y. Okada, N. C. Collins, Z. Schlesinger, J. W. Reiner, L. Klein, A. Kapitulnik, T. H. Geballe, and M. R. Beasley, Phys. Rev. Lett. **81**, 2498 (1998).

[12] Y. Maeno *et al.*, Nature (London) **372**, 532 (1994).

[13] G. Cao, S. McCall, J. E. Crow, and R. P. Guertin, Phys. Rev. Lett. **78**, 1751 (1996).

[14] Z. Fisk and G. W. Webb, Phys. Rev. Lett. **36**, 1084 (1976).

[15] D. L. Cox and M. Jarrel, J. Phys.: Condens. Matter **8**, 9825 (1996).

[16] F. Steglich, B. Buschinger, P. Gegenwart, M. Lohmann, R. Helfrich, C. Langhammer, P. Hellmann, L. Donnevert, S. Thomas, A. Link, C. Geibel, M. Lang, G. Sparn, and W. Assmus, J. Phys.: Condens. Matter **8**, 9909 (1996).

[17] I. I. Mazin and D. J. Singh, Phys. Rev. B **56**, 2556 (1997).

[18] G. Santi and T. Jarlborg, J. Phys.: Condens. Matter **9**, 9563 (1997).

[19] G. Cao, S. McCall, M. Shepard, J. E. Crow, and R. P. Guertin, Phys. Rev. B **56**, R2916 (1997).