Anomalous Spin Scattering Effects in the Badly Metallic Itinerant Ferromagnet $SrRuO_3$

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$SrRuO_3$ is an itinerant ferromagnet with $T_c \sim 150$ K. While the magnetization shows critical behavior that is well fit with universal critical exponents, the temperature derivative of the resistivity shows an unusually strong divergence as $T \to T_c^+$ with critical exponents higher than 0.9 and very weak divergence as $T \to T_c^-$. At low temperatures, the resistivity rapidly increases with temperature, and an unusual correlation with magnetization is found. We argue that the two phenomena stem from the fact that $SrRuO_3$ is an inherently bad metal. [S0031-9007(96)01229-X]

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The transport properties of good metallic ferromagnets, such as the $3d$ elemental ferromagnets, are by now fairly well understood. Experimentally, the temperature derivative of the resistivity dR/dT is found to exhibit the same critical behavior as the specific heat [1,2], and at low temperatures usually $R - R_0 \sim aT^2$ (where R_0 is the residual resistivity) with $a \sim 10^{-11}\Omega - \mathrm{cm/K^2}$ [3]. In these systems, $k_F l \gg 1$ well above the Curie temperature T_c (here l is the mean free path of the quasiparticle and k_F is the Fermi wave vector) and Boltzmann transport equations apply in the theoretical formulation of this behavior [3–7]. On the other hand, the behavior of bad metallic itinerant ferromagnets, for which $k_F l = O(1)$, is neither understood theoretically nor is there enough published data to form a coherent picture of their properties. In this Letter we report on magnetic and transport measurements of $SrRuO_3$ which strongly suggest that this itinerant ferromagnet, previously shown by Allen *et al.* [8] to be in the limit of $k_F l = O(1)$, exhibits anomalous behavior which is likely to be related to its bad metallicity, thus implying that the theory of itinerant ferromagnetism should be revisited when $k_F l = O(1)$.

In our measurements of $SrRuO_3$ we find the following: (1) an unusually strong divergence of dR/dT as $T \to T_c^+$ with an exponent of about 0.9 (compared to the expected specific heat exponent of $\sim$0.1); (2) a very weak divergence when $T \to T_c^-$; and (3) an increase in resistivity in the low-temperature range ($5 < T < 30$ K) that is 3 orders of magnitude larger than those observed in the elemental ferromagnets. To establish that the abnormal critical transport behavior is not due to magnetic peculiarities, we show that the magnetization data are well fit with conventional universal critical exponents. To investigate the rapid increase of R at low temperatures, we study its correlation with magnetization.

Poorly conducting metals [9] have been of interest for many decades, especially as they relate to the metal insulator transition and the superconducting phase transition [10,11] near $k_F l = O(1)$. In the past, such metals were realized by creating disorder in pure systems, either by alloying or by making them granular. In recent years, however, it has been shown that a variety of intriguing materials are in the $k_F l = O(1)$ limit in their highest quality form (e.g., well ordered crystals) and are, in some sense, intrinsically poor metals. Notable examples of such materials are high-temperature superconductors (HTS), fullerenes, and organic conductors. Emery and Kivelson [11] have recently examined the effects of bad metallicity in conjunction with HTS, but they noted that $R(T)$ in all of these "intrinsic" bad metals exhibit anomalous linear temperature dependence in a range which extends from $k_F l \leq 2\pi$ to $k_F l \geq 2\pi$. They further argued that the lack of change in the functional form of R as it passes through the Ioffe-Regel limit of $k_F l \approx 2\pi$ may indicate that Boltzmann transport equations are inapplicable in these materials over the *entire* temperature range (including $k_F l \geq 2\pi$). From this point of view, our results are evidence not only for a different interplay between transport and magnetism in the limit $k_F l = O(1)$ but also for this general assertion of Emery and Kivelson.

$SrRuO_3$ is a pseudocubic perovskite and an itinerant $4d$ ferromagnet with a $T_c \sim 165$ K for bulk materials and $\sim$150 K for films [12] with magnetic moments per ruthenium in bulk (films) above and below T_c of $1.6\mu_B$ ($1.4\mu_B$) and $2\mu_B$, respectively. Strong spin-orbit coupling of the ruthenium atoms yields large magneto-optic effects [13] and a magnetocrystalline anisotropy field on the order of several teslas which varies among the slightly different phases of $SrRuO_3$ exhibited by its bulk and thin-film forms. $SrRuO_3$ is metallic with carrier density inferred from Hall-effect measurements of $2 \times 10^{22}/\mathrm{cm^3}$ [14], which is an intermediate value between that of good metals (including the $3d$ elemental ferromagnets) and that of HTS, for example.

We have examined high-quality, single-domain thin films grown on miscut $SrRuO_3$ substrates by reactive electron beam coevaporation [15]. The films are orthorhombic with $a \sim 5.53$ Å, $b \sim 5.57$ Å, and $c \sim 7.85$ Å; and x-ray diffraction measurements demonstrate (110) growth, with the [001] ([$\bar{1}$10]) axis lying perpendicular (along) to the miscut direction. The full width at half maximum of the rocking curve taken around the (220) reflection

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is ~0.03 deg, which, along with the high resistivity ratio and the sharp kink at T_c, are indicative of the high quality of these films. The films display uniaxial magnetic anisotropy below T_c, and the magnetic moment lies in the (001) plane at a temperature dependent angle that continuously varies from close to 45° (with respect to the film plane) near T_c to about 60° at 5 K.

In Fig. 1 we show the resistivity curves of a 1000 Å thick film which was patterned for simultaneous resistivity measurements along the two crystalline axes. Above $T_c = 150$ K, the nearly linear temperature dependence (shown up to 300 K) persists up to 1000 K [8], despite the fact that by 500 K the mean free path is already less than 10 Å and Boltzmann transport theory should not be applicable. Similar behavior in HTS and in other materials such as VO_2 [16] was previously pointed out by Allen and others as indicative of non-Fermi-liquid behavior and was chosen by Emery and Kivelson [11] as the defining behavior of "bad metals."

The thermodynamic critical behavior is rather normal, in contrast to the anamalous critical transport behavior which we describe below. In Fig. 2 we show magnetization data for the film on which transport data were taken. These are consistent with our single crystal data. Figure 2(a) shows a good fit to the above-T_c zero-field-limit susceptibility, $\chi = \chi_0 t^{-\gamma}$, with Heisenberg exponent $\gamma = 1.38$ (for data not too close to T_c in order to avoid the crossover to Ising behavior) and a good fit to the spontaneous magnetization curve below T_c, $M = M_0 t^{\beta}$, with Ising exponent $\beta = 0.325$. Such crossover is expected due to the uniaxial anisotropy. Figure 2(b) shows the scaling of the entire magnetization data with the scaling function $H/t^{\beta+\gamma} = f(m/t^{\beta})$ which should have the asymptotic behavior of $f(x)_{x\to\infty} \to x^{\delta}$ with $\delta = 4.8$ and $f(x)_{x\to 0} \to x$. Ising exponents were used to scale the data from below T_c up to 1 K above T_c, and Heisenberg exponents to scale the rest of the data above T_c. The magnetization data clearly indicate a conventional transition with critical regime of at least ±20 K around T_c.

The sharp kink in the resistivity coincides with the magentic phase transition to within our experimental accuracy of ±0.1 K, and it is natural to compare this behavior to that of other ferromagnetic metals. The effect of critical spin fluctuations on resistivity in ferromagnetic metals has been of interest for many years. Fisher and Langer [5] found that the short range spin correlations made the important contribution to resistivity, leading to an energylike term in R_m (here R_m is the magnetic part of the resistivity) and critical behavior of dR_m/dT above T_c which is identical to that of the specific heat. Subsequent work [7] concluded that the same critical behavior is also expected below T_c. Experimental tests of these relationships with elemental ferromagnets [1,2] and some ferromagnetic intermetallic compounds [17] seem to confirm both the symmetry and the correspondence of dR_m/dT with the specific heat, which would imply a

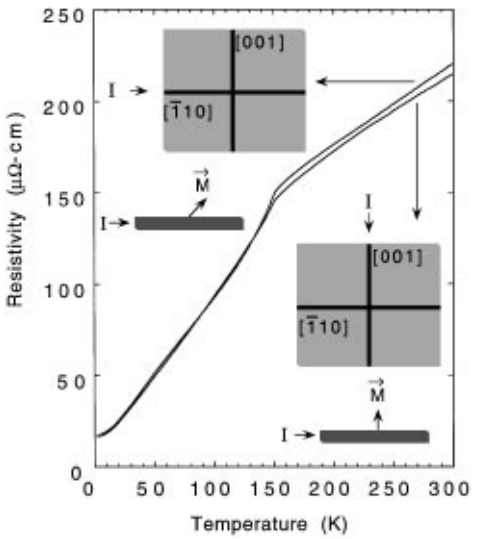

FIG. 1. Resistivity vs temperature in the [001] and [$\bar{1}$10] directions, and the relative orientation of the magnetic moments in the ordered state.

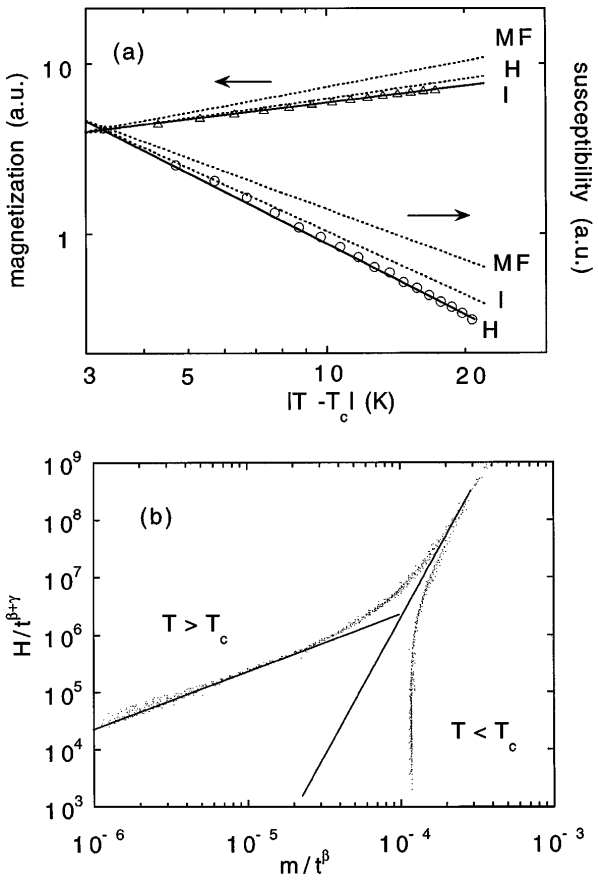

FIG. 2. (a) The spontaneous magnetization and the zero-field susceptibility with three different fits: mean field (MF), Heisenberg (H), Ising (I). (b) Scaling of magnetization vs temperature data taken at 15 different fields (200, 1250, 1500, 1750, 2000, 2250, 2500, 3000, 4000, 5000, 6000, 7000, 8000, 9000, and 10 000 Oe). Data from below T_c up to 1 K above T_c are scaled with Ising exponents, and the rest of the data above T_c are scaled with Heisenberg exponents. The two solid lines are the expected asymptotic behavior.

weak logarithmic divergence for Heisenberg ferromagnets and a weak power law with an exponent of about 0.1 for Ising ferromagnets. The critical transport properties of $SrRuO_3$, however, are much more strongly divergent than this, indicating that critical spin fluctuations affect the transport more strongly in this material than in conventional ferromagnetic metals.

Figure 3(a) shows dR/dT data of $SrRuO_3$ for currents in two different directions along with dR/dT data of iron [2] which we shifted up by 0.5 $\mu\Omega$ cm/K for presentation. The striking difference between the two systems cannot be a result of different R_m's; the estimated R_m for iron is $\sim$80 $\mu\Omega$ cm [18], and, while an estimate for R_m for $SrRuO_3$ is somewhat problematic [19], it is certainly lower than the total resistivity of $SrRuO_3$ near T_c which is not more than twice the R_m of iron. The inset shows dR/dT data of $SrRuO_3$ in a wider temperature range which allows us to estimate the nonmagnetic background of dR/dT to be in the range of 0.45–0.5 $\mu\Omega$ cm/K. We expect $dR_m/dT = (1 - t)^{-\alpha}$; thus we show in Fig. 3(b) the data of Fig. 3(a) (after subtracting from dR/dT a nonmagnetic part of 0.5 $\mu\Omega$ cm/K) on a log-log plot. There are several unusual properties of the critical behavior. The critical exponent 0.9–1.0 above T_c is much larger than the expected 0.1, and clearly much larger than that of iron; there is a clear asymmetry of the data above and below T_c. The data below T_c cannot be fit with any power law, and it appears that for current along the two directions the critical behavior is different not only in amplitude (which might be expected in this asymmetric geometry) but also in the critical exponent. Changing the background value to the smallest reasonable value of 0.45 $\mu\Omega$ cm/K yields exponents of 0.8–0.9 and an inferior fit. The flattening of the curves very close to T_c is probably due to rounding effects due to either inhomogeneities or some cutoff. We can exclude the possibility of crossover between a Gaussian regime (with an expected divergence of 0.5, which we also show for comparison) to a critical regime very close to T_c, since the magnetization data display a critical regime of at least 20 K. Moreover, the divergence above T_c is even larger than that expected from Gaussian fluctuations.

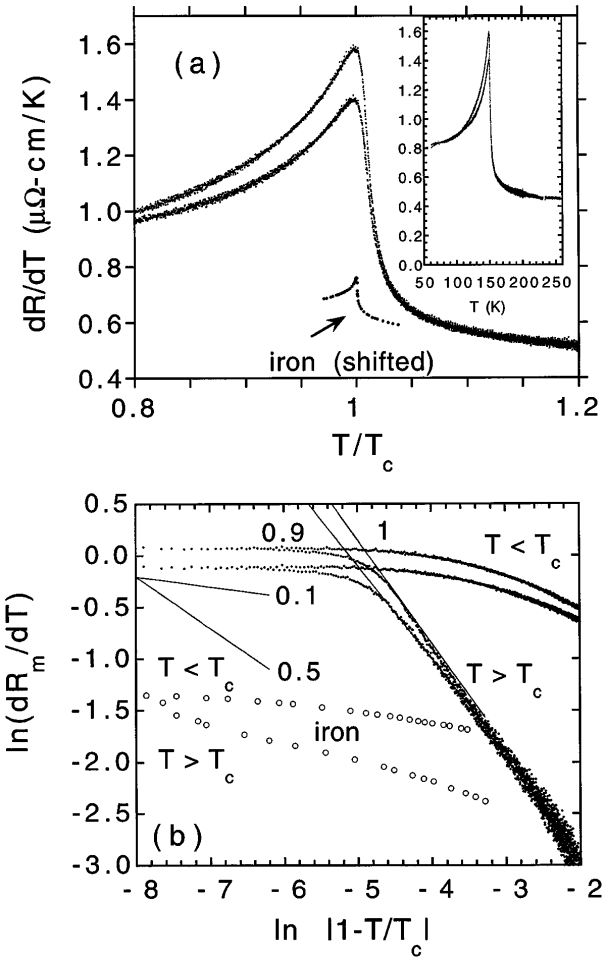

FIG. 3. (a) dR/dT with currents in the [001] and [$\bar{1}$10] directions—the stronger divergence is in the [$\bar{1}$10] direction. Iron data [2] are shown for comparison, shifted with a constant of 0.5 $\mu\Omega$ cm/K. The inset shows a larger temperature range that enables an estimate of the nonmagnetic part in dR/dT. (b) Log-log plot of dR_m/dT with slopes of 0.1 (which is expected), 0.5 (not expected Gaussian limit), and iron data [2] for comparison.

Figure 4 shows that also at low temperatures the resistivity of $SrRuO_3$ has unusual behavior. While elemental ferromagnets exhibit $R = R_0 + aT^2$ [3] with a on the order of 10^{-11} Ω cm/K^2, the resistivity of $SrRuO_3$ exhibits an increase three orders of magnitude larger than this, for

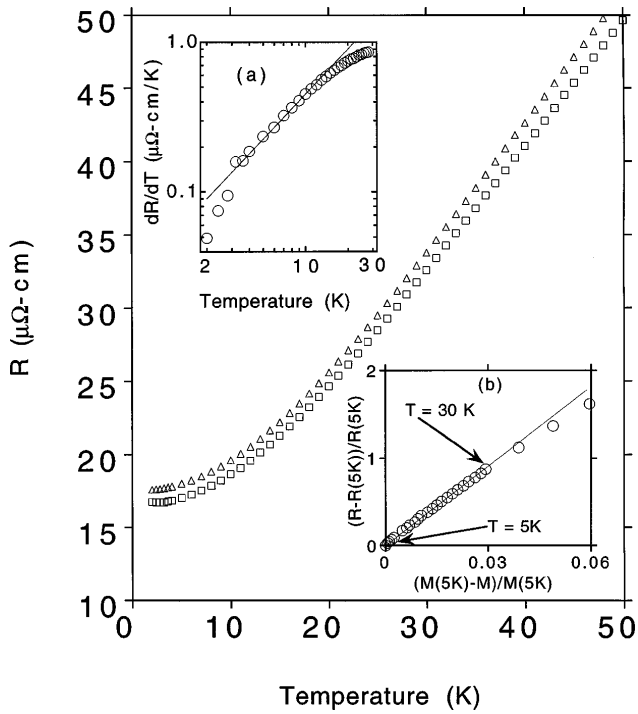

FIG. 4. Low-temperature resistivity vs temperature in the [001] and [$\bar{1}$10] directions. Inset (a) shows a log-log plot of dR/dT with a fit assuming $R = R_0 + aT^2$ in comparison. Inset (b) shows normalized resistivity change vs normalized magnetization change.

which the lower T_c can only partially account. Moreover, while inset (a) shows the limited success of a T^2 fit, inset (b) shows a surprising linear correlation between the magnetization and the resistivity which holds up to almost 30 K [20].

A main question that arises in light of these abnormal transport properties is whether they are likely to be related to the bad conductivity of $SrRuO_3$, and whether they reflect an underlying transport mechanism that is likely to be encountered in other bad metals. Clearly, an interesting issue in this context, which needs to be sorted out both experimentally and theoretically, is the relative importance of factors such as $k_F l$ and the carrier density in determining the unusual transport properties of $SrRuO_3$ and of bad metals in general. The strong divergence of dR/dT as $T \to T_c^+$ combined with the weak divergence as $T \to T_c^-$ is merely a manifestation of the resistivity nose dive around T_c, which means that transport is facilitated by the magnetic ordering more than in high-conductivity ferromagnets. Known cases of a dramatic effect of magnetic ordering on transport are the "colossal" magnetoresistance perovskites [21] or the Eu-rich EuO [22], where ferromagnetic ordering triggers an insulator to metal transition. It is not clear if any of the mechanisms proposed for these materials are relevant to $SrRuO_3$, but this similarity may suggest that a more local transport picture (as opposed to band transport) is a more adequate description for $SrRuO_3$ and perhaps for other bad metals as well. The behavior at low temperatures is somewhat reminiscent of that observed in dilute ferromagnetic alloys [23], where $R - R_0$ is larger than in elemental ferromagnets (although still much smaller than in $SrRuO_3$). In these materials, R_0 is on the order of several $\mu\Omega$ cm; and a $T^{3/2}$ term in $R(T)$ was observed, and attributed to the proportionality of the resistivity to the total number of magnons. This surprising similarity between high-quality single-crystal films and alloys may imply strong intrinsic sensitivity to disorder for $SrRuO_3$ and maybe of bad metals in general. A theoretical basis for this similarity may lie in the recent extension of the coherent potential approximation to interacting electron systems [24].

In this paper we have found that the low temperature and the critical transport properties of a poorly conducting itinerant ferromagnet deviates sharply from those observed in good metallic ferromagnets. We believe that this unusual behavior is related to its poor conductivity, and assert that a different theoretical treatment of the interplay between transport and magnetism may be required for poorly conducting ferromagnets; however, testing this hypothesis on a wider range of materials is clearly warranted. Our results may have further implications if the "bad-metal" classification [11] is proven to be related to an underlying common transport mechanism with different manifestations in different ground states, such as itinerant magnetism or superconductivity.

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