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1996 J. Phys.: Condens. Matter 8 10111

(http://iopscience.iop.org/0953-8984/8/48/026)

J. Phys.: Condens. Matter **8** (1996) 10111–10126. Printed in the UK

Transport and magnetization in the badly metallic itinerant ferromagnet $SrRuO_3$

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Received 17 June 1996, in final form 13 September 1996

Abstract. $SrRuO_3$ is an itinerant ferromagnet with $T_c \sim 160$ K and a 'bad metal' in the limit of $k_F l = \mathcal{O}(1)$. While the magnetic properties of $SrRuO_3$ in the paramagnetic phase, near the ferromagnetic phase transition and at low temperatures are normal and consistent with its being a strong itinerant ferromagnet, the transport properties (resistivity and magnetoresistance) sharply deviate from that of good metallic ferromagnets. We conjecture that the distinct transport behaviour of $SrRuO_3$ is related to its being a 'bad metal' in the $k_F l = \mathcal{O}(1)$ limit, and discuss the possible relevance of our results to the unusual transport properties of other 'bad metals' such as high-temperature superconductors.

1. Introduction

The study of the interplay between magnetism and transport in ferromagnets has been mainly focused on two groups: on the one hand, good metals such as iron, nickel, and cobalt [1–7], and on the other hand, poor conductors such as the perovskite manganites [8] (the so-called 'colossal'-magnetoresistance materials) or Eu-rich EuO [9]. In the good metals $k_F l \gg 1$ even at temperatures T much larger than the Curie temperature, T_c (k_F is the Fermi wavevector and l is the charge-carrier mean free path), and Boltzmann equations can be used to calculate the magnetic resistivity, ρ_m, over the entire temperature range; this yields: $\rho_m \rightarrow$ constant for T well above T_c; $\mathrm{d}\rho_m/\mathrm{d}T \propto C$ near T_c (here C is the specific heat), and negligible contribution when $T \rightarrow 0$ [1–7]. On the other hand, in the bad conductors the effect of magnetization is much more dramatic yielding: polaronic-like behaviour (with $\mathrm{d}\rho/\mathrm{d}T < 0$) above T_c which is related to the magnetic disorder, a metal-to-insulator transition at T_c, and metallic behaviour ($\mathrm{d}\rho/\mathrm{d}T > 0$) with a strong correlation between resistivity and magnetization well below T_c [8, 9].

Here we report on $SrRuO_3$ which is an itinerant ferromagnet ($T_c \sim 160$ K) that may represent a class of badly metallic ferromagnets in the limit of $k_F l = \mathcal{O}(1)$—a class that, we believe, cannot be classified either with good metallic ferromagnets or with the manganites, for example. We find:

- at $T > T_c$ the resistivity ρ increases almost linearly with T up to more than 600 K although (as pointed out by Allen *et al* [10]) saturation is expected;
- as $T \rightarrow T_c^+$ the temperature derivative of the magnetic part of the resistivity, $\mathrm{d}\rho_m/\mathrm{d}T$, diverges with an exponent of the order of 1, an order of magnitude larger than the expected specific heat exponent of $\sim$0.1;

0953-8984/96/4810111+16$19.50

- while the critical behaviour of $d\rho_m/dT$ around T_c is usually found to be symmetric (although with different amplitudes), very weak divergence of $d\rho_m/dT$ is observed as $T \to T_c^-$;
- at low temperatures (below 30 K) ρ of the films with low residual resistivity rapidly increases with increasing temperature and exhibits a clear correlation with the magnetization;
- at $T < 4$ K for low-residual-resistivity films, and at temperatures as high as 40 K for high-residual-resistivity films, resistivity minima are observed;
- anisotropic magnetoresistance which for fields parallel (perpendicular) to the current is negative (positive) and as high as ∼20 % at low temperatures and a 6 T field;
- large anisotropic domain-wall resistivity.

The effect of magnetization on transport in $SrRuO_3$ is much stronger than in iron and nickel but less dramatic (at least around T_c) than its effect in the manganites. Usually, the distinction is made between ferromagnets with metallic conductivity and those with polaronic conductivity above T_c. Here we show that further classification of the metallic ferromagnets might be warranted, since the resistivity of $SrRuO_3$, which is a badly metallic ferromagnet in the limit of $k_F l = \mathcal{O}(1)$, qualitatively deviates from that of good metallic ones. This observation may carry implications that go beyond the study of itinerant ferromagnets in the $k_F l = \mathcal{O}(1)$ limit. Here we examine the specific character of bad metallicity in the presence of ferromagnetic interactions, but the results may be relevant to a large group of intriguing materials such as high-temperature superconductors (HTS), organic conductors and fullerenes which, as pointed out by Emery and Kivelson [11], are all 'bad metals' in the range of $k_F l = \mathcal{O}(1)$.

The paper is organized as follows. We start by presenting some general properties of $SrRuO_3$. We then present the magnetic properties, which are rather common, and the resistivity data, which are quite unusual. We end with a discussion of the results.

2. General properties of $SrRuO_3$

$SrRuO_3$ is a pseudocubic perovskite and an itinerant ferromagnet with a Curie temperature of ∼160 K. Most reports on bulk materials indicate that it is orthorhombic with lattice parameters of $a \cong 5.55$ Å, $b \cong 5.56$ Å and $c \cong 7.84$ Å [12, 13]; however, there are also reports of different phases, particularly, a tetragonal phase with lattice parameters of $a \cong b \cong 5.62$ Å and $c \cong 7.81$ Å [14].

Efforts to grow films of this compound were originally motivated by their compatibility with HTS films [15, 16]. Different substrates have been used. Films on $LaAlO_3$/[001] substrates are orthorhombic with the c-axis mainly lying in the plane of the film (along either of the two principal directions); however, there is also a varying component of c-perpendicular grains. The dense twinning of $LaAlO_3$ substrates is probably responsible for the relatively poor quality of these films: transmission electron microscopy measurements indicate a typical grain size of a few hundred ångströms, the magnetic phase transition is smeared over a few degrees, and the magnetization curves exhibit coercivities of several teslas [17].

Much better films can be grown on $SrTiO_3$ substrates. The films are orthorhombic with $a \cong 5.53$ Å, $b \cong 5.57$ Å and $c \cong 7.85$ Å, and although they mostly grow with the c-axis in the film plane (again along either of the two principal directions on the [001] substrate) [15], some component (even as high as 20%) of c-perpendicular grains is occasionally observed in transmission electron microscopy [18]. Even better films can be grown on miscut $SrTiO_3$ susbstrates for which the c-axis of the film lies perpendicular to the miscut direction. We

have used $\sim 2^\circ$ miscut substrates to grow high-quality twin-free films with more than 99% of the film with the *c*-axis in the same direction (however, our measurements do not exclude the possible existence of few per cent of *c*-perpendicular grains). The measured full width of the rocking curve of these films, taken around the (220) reflection at half-maximum, is $\sim 0.03^\circ$, and the resistivity ratio between room temperature and the zero-temperature limit is as high as ~ 34, greater than any reported value for single crystals [12]. The T_c of these high-quality films is 150 K (0.2), and the sharpness of the magnetic phase transition as observed in magnetization and transport measurements indicates that T_c is not reduced due to disorder or atomic deficiencies, but rather due to structural difference between these films and bulk samples with $T_c \sim 160$ K.

An important source for some of the intriguing properties of $SrRuO_3$ is the relative strength of the spin–orbit coupling of the ruthenium atom (900 cm^{-1} compared to 400 cm^{-1} in iron, for example [19]). This property is the origin of the strength of features such as magneto-optic effects [17], magnetocrystalline anisotropy, anisotropic magnetoresistance, and the anomalous Hall effect [20].

Other properties that are relevant to the transport properties of $SrRuO_3$ are the carrier density and the quasiparticle effective mass. High-field Hall measurements (in the regime of saturated anomalous Hall effect) indicate electron-like quasiparticles with carrier densities of $\sim 2 \times 10^{22}$ cm^{-3} [20], which is intermediate between that of good metals and that of optimally doped HTS, for example. The γ-value in the specific heat is 30 mJ mol^{-1} K^{-2}, which is intermediate between that of good metals and that of heavy fermions, but even more importantly γ is 3.7 times larger than that estimated by local spin-density approximation calculations [10], indicating strong electron correlations.

3. Magnetization

3.1. Magnetic properties

Magnetism in $SrRuO_3$ is itinerant and primarily due to electrons with Ru 4d character. The measured moment above T_c is 2 μ_B, which implies, in an oversimplified local description, a low-spin state of the four ruthenium electrons. The spontaneous magnetization in the zero-temperature limit in bulk (films) is 1.6 μ_B (1.4 μ_B), which is consistent with recent band calculations that also show strong 2p–4d hybridization [10, 21]. The low-temperature moments for both crystals and the films reported here are larger than those reported previously, most probably due to a combination of large magnetocrystalline anisotropy (discussed below) and the difficulty in making single-domain films and crystals. The ratio q between the high-temperature moment and the zero-temperature saturated magnetization of $SrRuO_3$ is ~ 1.3. The q-value is a common measure of the itinerant magnetism strength: $q \sim 1$–2 indicates strong (i.e. more local in real space) itinerant ferromagnetism, while higher values of q indicate weak and less localized magnetism [22]. Therefore, we expect the magnetic properties of $SrRuO_3$ to be similar to those of the elemental 3d ferromagnets nickel, cobalt and iron for which $q \sim 1$–2, and different, for example, from those of the weak itinerant ferromagnet $ZrZn_2$ for which $q \geqslant 5$. As discussed below, we find at low temperatures a $T^{3/2}$ spin-wave contribution to the magnetization, and near T_c we find a non-mean-field magnetic critical behaviour. Such behaviour, both at low temperatures and near T_c, is expected for strong itinerant ferromagnets and is found experimentally in the 3d elemental ferromagnets but not in $ZrZn_2$, which shows pure T^2-dependence at low temperatures and mean-field behaviour near T_c.

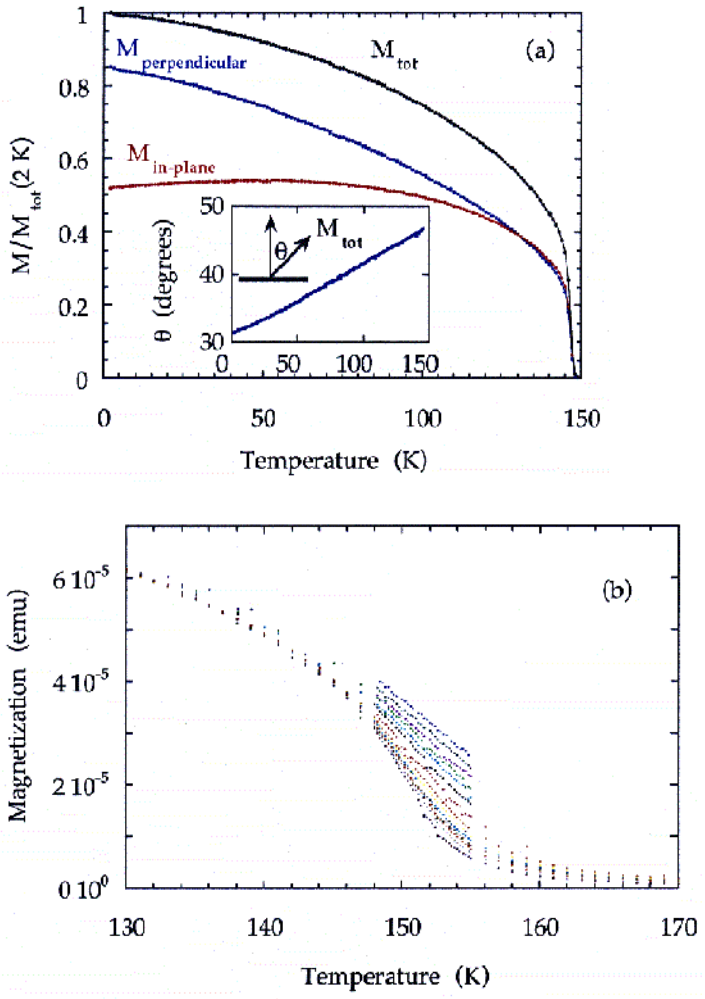

Figure 1. (a) Temperature dependences of the in-plane, out-of-plane, and total remanent magnetizations of film (a) from figure 2. The film was cooled in a saturating field down to 5 K and the magnetization was measured upon warming after removing the applied field. The temperature dependence of the angle between the magnetic moment and the normal to the film plane is also shown. Similar behaviour was also exhibited by the film (b), and the film (c) (for the latter only the perpendicular component could be compared due to twinning). (b) Magnetization data for 15 different fields (200 Oe, 1250 Oe, 1500 Oe, 1750 Oe, 2000 Oe, 2250 Oe, 2500 Oe, 3000 Oe, 4000 Oe, 5000 Oe, 6000 Oe, 7000 Oe, 8000 Oe, 9000 Oe, 10 000 Oe) of the film (b) from figure 2. (c) The spontaneous magnetization and the zero-field susceptibility with three different fits: mean field (MF), Heisenberg (H), Ising (I). (d) Scaling of the magnetization data shown in (a) from below T_c up to one degree above T_c with Ising exponents and scaling of the rest of the data above T_c with Heisenberg exponents. The two solid lines show the expected asymptotic behaviour.

3.2. Magnetocrystalline anisotropy

The reported magnetocrystalline anisotropy of bulk materials is somewhat confusing. Kanbayasi reports torque measurements of different phases of single crystals. In one case

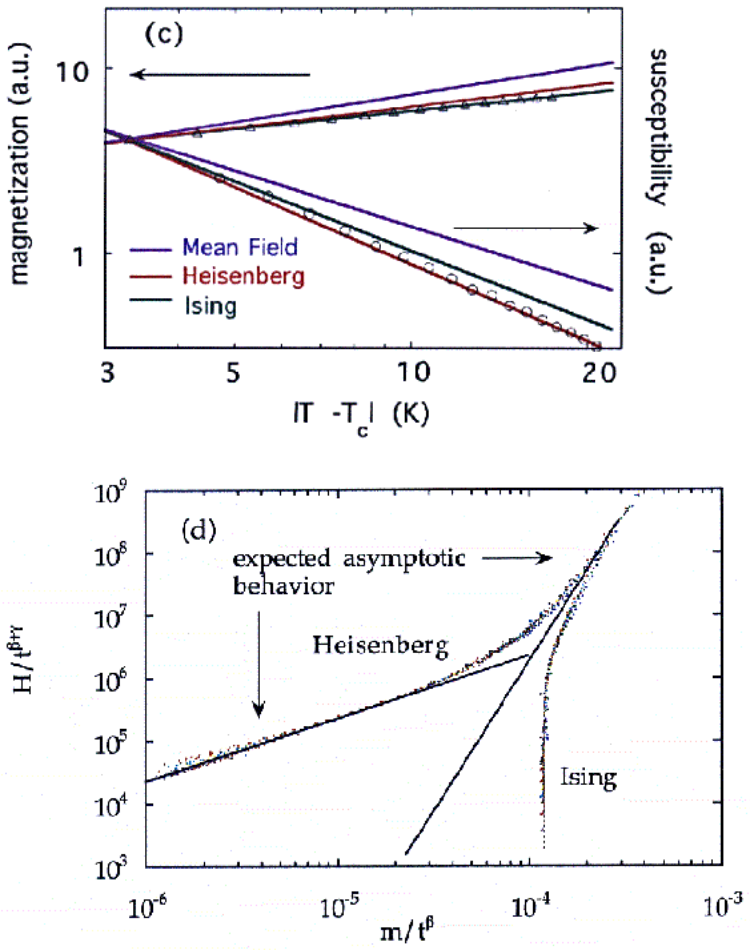

Figure 1. (Continued)

he reports pseudocubic anisotropy with the ⟨110⟩ directions (in the cubic frame) being the easy axes and an anisotropy field of ∼2 T [23]. However, he also reports on a tetragonal phase with easy axes only in the (001) plane [14] and an anisotropy field (inferred from the reported anisotropy energy) larger than 10 T. Magnetization measurements on single crystals from our group [24] indicate pseudocubic anisotropy but with fields larger than 2 T.

The magnetocrystalline anisotropy of films is best studied with twin-free films grown on miscut $SrTiO_3$ substrates. We found from magnetic measurements of both the longitudinal and transverse components of the magnetization of such films that there is a single easy axis which lies in the (001) plane. The cubic symmetry is probably broken by strain effects of the substrate (see the discussion of the orientational transition below). It is surprising, however, that this different magnetocrystalline anisotropy is observed in films 1000 Å thick without relaxation to the anisotropy of bulk, which may indicate multiple structural metastable states. Films grown on $LaAlO_3$ are twinned, so we can only indirectly deduce that these films have uniaxial anisotropy as well, but as we elaborate below, it is probably different from that of films on $SrTiO_3$. We do not have an accurate value for the anisotropy field, but it is clearly higher than the irreversibility field, which at low temperatures is ∼2 T.

3.3. Low-temperature excitations

Figure 1(a) shows the temperature dependence of the in-plane and out-of-plane remanent magnetization for one of our largely twin-free films. The pronounced temperature dependence of the magnetization at low temperatures is due to spin-wave and Stoner excitations, which yield $T^{3/2}$- and T^2-terms, respectively [17, 25]. Fitting the temperature dependence of the full magnetic moment assuming $M/M_0 = (1 - AT^{3/2} - BT^2)$ yields $A = 1 \times 10^{-4}$ K$^{-3/2}$ and $B = 2 \times 10^{-5}$ K^{-2}, which are close to the theoretical predictions. The contribution of the two kinds of excitation was predicted theoretically by Herring and Kittel [26] and was observed in other strong itinerant ferromagnets as well [27].

3.4. The orientational transition

On top of the temperature dependence of the magnetization there is also an orientational transition [28] in which the easy axis in the (001) plane continuously changes its angle θ with respect to the normal from $\sim 30^\circ$ at low temperatures to $\sim 45^\circ$ at T_c, at a basically constant rate of $\sim 0.1^\circ$ K^{-1} (see figure 1(a)). One trivial source of such an orientational transition can come from the competition between the magnetocrystalline anisotropy and the demagnetization field. Here, however, we would expect the easy axes to be at 45°, and competition with the demagnetization field would only increase this angle. Therefore, the orientational transition is likely to be related to an additional term in the anisotropy energy which yields an effective temperature-dependent uniaxial anisotropy. The in-plane magnetization is given by $M(T)\sin(\theta(T))$, where $\mathrm{d}M/\mathrm{d}T < 0$ and $\mathrm{d}\sin(\theta)/\mathrm{d}T > 0$. The competition between the two terms leads to a maximum of the in-plane magnetization at about 50 K, and as will be discussed elsewhere, this has some consequences on transport properties as well. Since no maximum was observed in the in-plane magnetization of films on $LaAlO_3$ it is likely that the temperature dependence of the reorientation transition of these films is at least weaker.

3.5. Critical behaviour

Figure 1(b) shows magnetization data for $SrRuO_3$ at around T_c for different applied fields. Figure 1(c) demonstrates that mean-field exponents cannot fit the data in a temperature interval of ± 20 K around T_c [29]. This implies that the itinerant magnetism of $SrRuO_3$ is different from that of the archetypal weak itinerant ferromagnet $ZrZn_2$ which exhibits mean-field behaviour down to $|t| = |(T - T_c)/T_c| = 0.01$ [30]. The data are much closer to that of the elemental 3d ferromagnets which are strong ferromagnets with 'local' magnetic features and exhibit critical behaviour with non-mean-field exponents up to at least $|t| = 0.1$ [31] (although still somewhat different from pure Heisenberg behaviour). The distinction between the quality of the fit with Ising exponents and that with Heisenberg exponents is much more subtle and requires (as usual) stringent determination of T_c. After determining that $T_c = 150 \pm 0.1$ K, based on Arrott plots and transport data, we found that a better fit to the above-T_c zero-field-limit susceptibility, $\chi = \chi_0 t^{-\gamma}$, is obtained with $\gamma = 1.38$ (Heisenberg). On the other hand, a better fit to the spontaneous magnetization below T_c, $M = M_0 t^{\beta}$, is obtained with $\beta = 0.325$ (Ising), which indicates a crossover, as might be expected in a ferromagnet with uniaxial anisotropy.

Figure 1(d) shows the scaling of the magnetization data (shown in figure 1(b)) with the scaling function $H/t^{\beta+\gamma} = f(m/t^{\beta})$ which should have the asymptotic behaviour of $f(x)_{x\to\infty} \to x^{\delta}$ with $\delta \approx 4.8$ and $f(x)_{x\to 0} \to x$ (the two lines of the expected asymptotic behaviour are also shown in the figure). We used Ising exponents to scale the data from

below T_c up to one degree above T_c and Heisenberg exponents to scale the rest of the data above T_c.

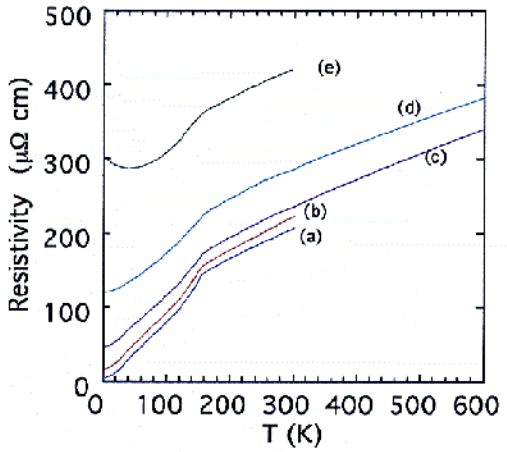

Figure 2. Resistivity curves of $SrRuO_3$ films 1000 Å thick: (a) and (b) are on miscut $SrTiO_3$ substrates and the current is perpendicular to the c-axis, (c) is on a regular $SrTiO_3$ substrate, (d) is on a $LaAlO_3$ substrate, and (e) is on an YSZ substrate.

The observation of normal critical behaviour of the magnetic phase transition as probed by the magnetic measurements is important to our claim below that the abnormal transport behaviour is not related to any magnetic anomalies but signals unusual transport properties.

4. Resistivity

4.1. Resistivity at $T > T_c$

Figure 2 shows resistivity as a function of temperature for different $SrRuO_3$ films. At $T > T_c$ the resistivity $\rho(T)$ increases almost linearly with T without saturation. Similar behaviour in HTS and in other materials such as VO_2 [32] was previously pointed out by Allen and others as indicative of non-Fermi-liquid behaviour and was chosen by Emery and Kivelson [11] as the defining behaviour of 'bad metals'. Examining resistivity curves of the films with the lower residual resistivity, one can see that the high-temperature resistivity does not extrapolate to the residual resistivity, but to $\sim$70 $\mu\Omega$ cm higher. Since the origin of the quasi-linear slope in this range is not understood, it is not clear whether it is appropriate to use the Bloch–Grüneisen fit together with Mathiessen's rule in order to decouple the contribution of phonons and spins to the resistivity. However, it is interesting to note that if we do apply the Bloch–Grüneisen equation, it implies that the paramagnetic spin-scattering resistivity, ρ_m, contributes $\sim$70 $\mu\Omega$ cm to the resistivity, consistent with the observation that in many good metallic ferromagnets $\rho_m \approx cS(S+1)$ [33] where $c = 30$ $\mu\Omega$ cm and S is the spin (1 in our case). Therefore, the estimated ρ_m in $SrRuO_3$ by itself does not provide evidence for abnormal behaviour.

The difficulty of unequivocally excluding the possibility that the high-temperature resistivity is simply the expected Bloch–Grüneisen behaviour is a common problem of 'bad metals'. One needs detailed knowledge of the electronic structure and its temperature dependence in order to make such conclusive statements. As we discuss below, the observed

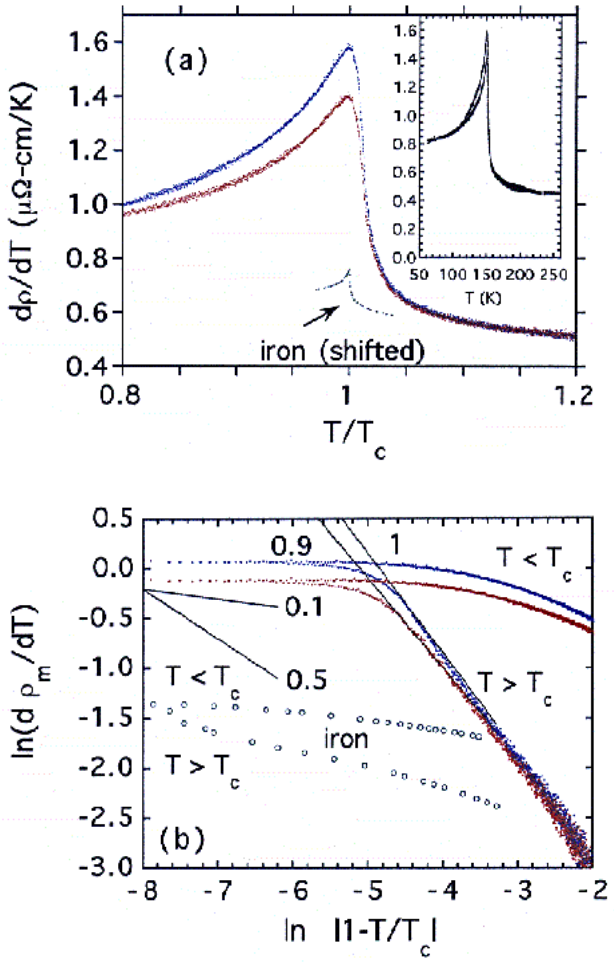

Figure 3. (a) $\mathrm{d}\rho/\mathrm{d}T$ with currents in the [001] (red) and [$\bar{1}$10] (blue) directions of film (b) from figure 2 with iron data (green) from reference [2], for comparison. (b) Log–log plots of $\mathrm{d}\rho_m/\mathrm{d}T$ with slopes of 0.1 (which is expected) and 0.5 (the Gaussian limit), and iron data (green) from reference [2], for comparison. The flattening of the curves very close to T_c is probably due to rounding effects, either due to inhomogeneities or due to some cut-off. We can exclude the possibility of crossover between a Gaussian regime with an expected divergence of 0.5 to a critical regime very close to T_c, since the magnetization data display a critical regime of at least $\pm$20 K. Moreover, the divergence above T_c is even larger than that expected from Gaussian fluctuations.

anomalous behaviour of the resistivity of $SrRuO_3$ near T_c is particularly interesting also because it suggests that the high-temperature resistivity is not the phononic Bloch–Grüneisen behaviour (with an offset). This conclusion may be relevant to other 'bad metals' as well.

4.2. Critical behaviour of $\mathrm{d}\rho_m/\mathrm{d}T$ *near* T_c

Figure 3 shows the critical behaviour of $\mathrm{d}\rho/\mathrm{d}T$ near T_c [29]. Since $\mathrm{d}\rho/\mathrm{d}T$ is fairly constant far enough above T_c, we subtract an offset of 0.5 $\mu\Omega$ cm in order to study the contribution to $\mathrm{d}\rho/\mathrm{d}T$ related to the magnetic fluctuations, $\mathrm{d}\rho_m/\mathrm{d}T$. We see power-law divergence with

an exponent of the order of 1 as $T \to T_c^+$ and very weak divergence as $T \to T_c^-$. Changing the background value to the smallest reasonable value of 0.45 $\mu\Omega$ cm yields for the data above T_c exponents of 0.8–0.9 and an inferior fit. This behaviour should be compared to the theoretical predictions and experimental observations in good metallic ferromagnets. The effect of critical spin fluctuations on the resistivity near T_c was first treated by de Gennes and Friedel [4] who stressed the importance of the diverging spin–spin correlation near T_c and anticipated a peak in the resistivity. That this peak was often not observed experimentally motivated Fisher and Langer to revisit the issue [5]. They found that it was the short-range spin correlations which governed the resistivity, leading to an energy-like term in ρ_m and critical behaviour of $\mathrm{d}\rho_m/\mathrm{d}T$ above T_c which is identical to that of the specific heat. Subsequent work [7] concluded that the same critical behaviour is also expected below T_c. Experimental tests of these relationships with good metallic ferromagnets [1, 2] seem to confirm both the symmetry and the correspondence of $\mathrm{d}\rho_m/\mathrm{d}T$ with the specific heat. This picture implies a weak logarithmic divergence for Heisenberg ferromagnets and a weak power law with an exponent of $\sim$0.1 for Ising ferromagnets. Although resistivity kinks at around the Curie temperature are observed in various ferromagnetic intermetallic compounds as well, their critical behaviour has been studied only in rare cases such as for TbZn that exhibits mean-field behaviour above T_c with an exponent of 0.5 that crosses over to the expected logarithmic behaviour close to T_c [34].

In figure 3, we compare our data with that for iron [2], the measured critical behaviour of which is close to that expected of Heisenberg ferromagnets [31]. It is apparent that the critical transport properties of $SrRuO_3$ are more strongly divergent, indicating that critical spin fluctuations affect the transport much more strongly in this material. One may ask whether the data in figure 3(a) should be scaled by ρ_m. For iron the estimated ρ_m is $\sim$80 $\mu\Omega$ cm [33], and while we do not have a reliable estimate of ρ_m in $SrRuO_3$ for reasons discussed above, it is clearly less than the total resistivity at T_c. Therefore, a scaling factor cannot be larger than 2. Hence it cannot account for the striking qualitative difference. That the magnetic phase transition itself is normal supports the view that the abnormal behaviour is related to unusual transport properties.

The strength and beauty of critical phenomena is their insensitivity to many details of the systems studied. Therefore, a clearly different behaviour as exhibited by $SrRuO_3$ implies the invalidity of some of the basic assumptions of the theoretical treatment. What is commonly assumed is that the resistivity is $\rho = \rho_0 + \rho_{ph} + \rho_m$ where ρ_0 is the residual (impurity) resistivity, ρ_{ph} is the resistivity due to electron–phonon scattering, which appears in the Bloch–Grüneisen equation, and ρ_m is the magnetic resistivity due to spin scattering of electrons; all of these contributions to the resistivity are calculated using the Boltzmann equations. However, one may question the validity of the Boltzmann equations. Such an assertion was made by Emery and Kivelson [11] regarding 'bad metals' in general, based on the non-saturating behaviour of the resistivity at high temperatures and the lack of any crossover as the resistivity passes through the Ioffe–Regel limit. If this is the case, then one should not wonder why the Fisher–Langer predictions [5] for $\mathrm{d}\rho_m/\mathrm{d}T$ (that starts with the Boltzmann equations) do not apply to $SrRuO_3$, but rather explore both experimentally and theoretically whether a specific universal critical behaviour takes place as a result of a different transport mechanism. Another possibility is that there are other contributions to ρ which are disregarded and are strongly affected by the ferromagnetic transition, a possibility which may be related to the fact that $SrRuO_3$ is an itinerant ferromagnet and that Stoner splitting clearly affects the states of the conduction electrons. While the breakdown of Boltzmann equations near T_c is not obvious in such a scenario, it still implies a strong non-phononic origin of the high-temperature resistivity.

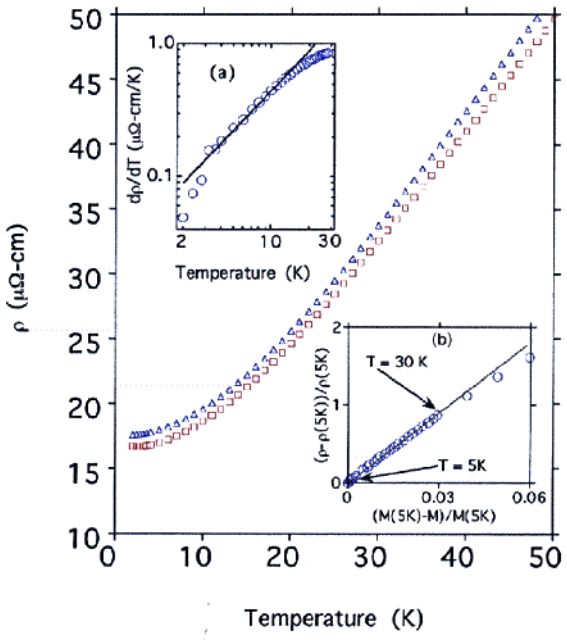

Figure 4. The low-temperature resistivity versus temperature in the [001] (red) and [$\bar{1}$10] (blue) directions of film (b) from figure 1. Inset (a) shows a log–log plot of $\mathrm{d}\rho/\mathrm{d}T$ assuming $\rho = \rho_0 + aT^2$, in comparison. Inset (b) shows the resistivity change versus the magnetization change.

4.3. $\rho(T)$ at low temperatures

Figure 4 shows $\rho(T)$ (of the film (b) from figure 2) at low temperatures where ρ rapidly increases with increasing temperature [29]. From figure 2 it is apparent that while the low-temperature resistivity of the high-quality films is very similar, the low-temperature resistivity of the poorer films is qualitatively different. This difference is mainly due to the occurrence of resistivity minima (discussed in the next section) that affects the resistivity of the lower-quality films over an extended temperature range.

The common temperature dependence of the resistivity ρ in good metallic ferromagnets is $\rho = \rho_0 + aT^2$ where a is of the order of $\sim 10^{-11}$ Ω cm [3]. This dependence is attributed to electron–electron scattering, which yields a T^2-dependence due to Pauli's exclusion principle. For a spherical Fermi surface this scattering contributes to resistivity only via Umklapp processes and thus this term is practically undetectable for simple monovalent alkali metals. However, in ferromagnetic metals different groups of electrons on the Fermi surface have different Fermi velocities, and scattering between 'heavy' and 'light' electrons contributes to the resistivity even without Umklapp processes. $SrRuO_3$ has a non-spherical Fermi surface [10, 21], so electron–electron scattering can be important (even without magnetism), but the change in resistivity below 30 K in $SrRuO_3$ is three orders of magnitude larger than that observed in good metallic ferromagnets. The resistivity due to electron–magnon scattering is expected to be proportional to $\int q^3[\exp(Dq^2/kT) - 1]^{-1}\,\mathrm{d}q$, where two powers of q come from the small-angle scattering factor and the rest is the number of magnons which undergo a collision, with D the spin-wave stiffness coefficient. This mechanism also yields a T^2-dependence. However, in clean ferromagnetic systems the

magnon contribution is usually considered negligible compared to the electron–electron scattering. On the other hand, in ferromagnetic alloys magnons are important and the resistivity was shown to be proportional to the total number of magnons [35]. Inset (a) of figure 4 clearly demonstrates that the expected T^2-fit has very limited success, whereas inset (b) shows that the change in resistivity is well correlated with the change in magnetization. Note, however, that since the relative change in M is only of few per cent, correlations with higher powers of M are not excluded.

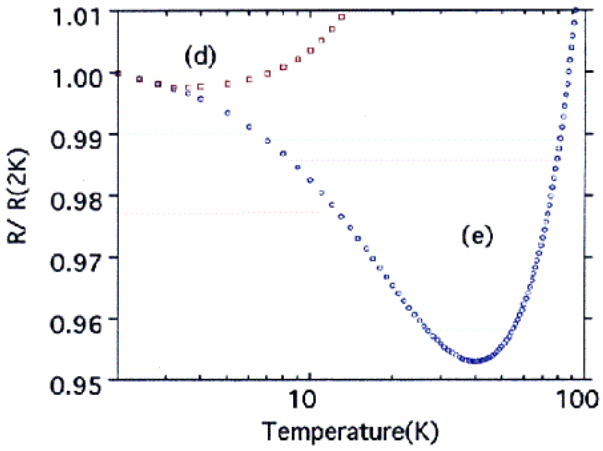

Figure 5. Resistivity minima of films (d) and (e) from figure 2.

The apparent similarity between disordered ferromagnetic alloys and the high-quality crystalline $SrRuO_3$ raises the possibility of an intrinsic sensitivity to disorder, which may find a theoretical basis in the recent extension of the coherent potential approximation to interacting electron systems [36].

4.4. Resistivity minima

Figure 5 shows a blow-up of the low-temperature resistivity of the films (d) and (e) shown in figure 2, plotted in a semilog format. Since the resistivity minima shift to higher temperatures as the residual resistivity increases, they are more conspicuous in the films with higher resistivity. However, down to 2 K resistivity minima were also observed in the high-quality films (c) and (b) (for the latter only in one current direction). The temperature dependence in the zero-temperature limit is slower than logarithmic, and while the resistivity of the highly disordered film is near the Mott limit, it is very unlikely that the resistivity minima of the better films is related to a metal–insulator transition.

Resistivity minima are common in paramagnetic metals that host localized magnetic impurities in low concentrations (the Kondo effect). There are reports on resistivity minima in ferromagnetic alloys which are suspected to be due to the Kondo effect [3], but very little is known about these cases theoretically. The resistivity minima in $SrRuO_3$ are correlated with disorder which may suggest that non-magnetic disorder localizes the otherwise extended magnetic states. This may also explain the shift of the resistivity minima to higher temperatures as the residual resistivity (which reflects disorder) increases.

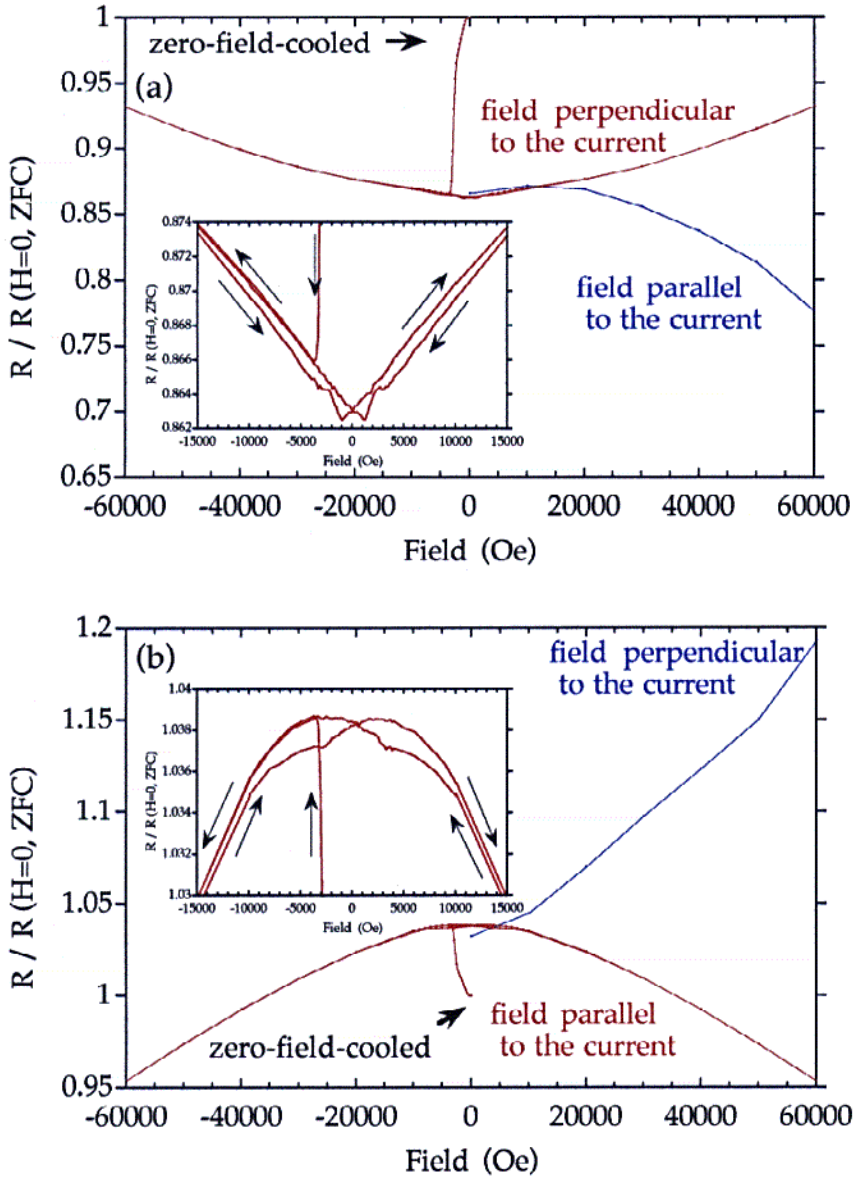

Figure 6. The magnetoresistance of film (a) from figure 2: the data for the field-dependent magnetoresistance are taken after cooling the film in zero field; thus, the initial curve is mostly due to removal of domain walls. The temperature-dependent magnetoresistance is relative to zero-field-cooled resistivity, so part of the magnetoresistance, particularly at low temperatures, is due to the removal of domain walls. (a) The field-dependent magnetoresistance at 5 K with the current in the *c*-direction. The inset shows the hysteretic low-field magnetoresistance due to domain formation. (b) The field-dependent magnetoresistance at 5 K with the current perpendicular to the *c*-direction. The inset shows the hysteretic low-field magnetoresistance due to domain formation. (c) The temperature-dependent magnetoresistance with the current in the *c*-direction. (d) The temperature-dependent magnetoresistance with the current perpendicular to the *c*-direction.

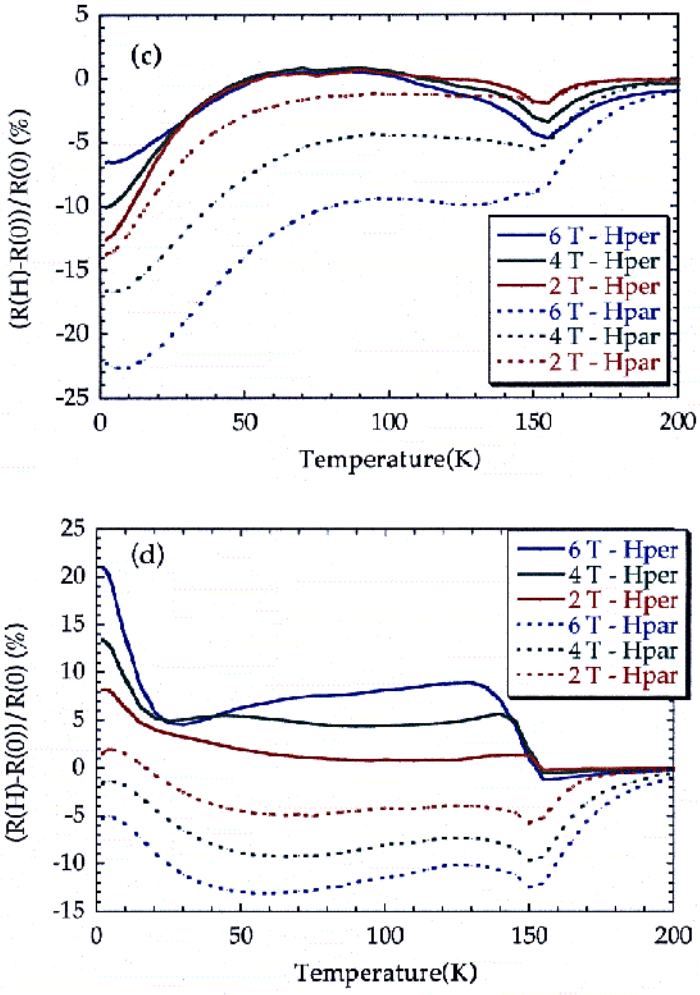

Figure 6. (Continued)

4.5. Magnetoresistance

Figure 6 shows the magnetoresistance of film (a) from figure 2 for currents in the [001] and [$\bar{1}$10] directions as a function of field and temperature. We address two main features:

- the high-field magnetoresistance which is large, anisotropic and non-saturating, and
- the low-field domain-wall-related magnetoresistance at low temperatures which is very large and anisotropic.

The origin of high-field negative magnetoresistance near T_c can be qualitatively understood as being related to the suppression of spin fluctuations [37]. On the other hand the high-field negative magnetoresistance which is observed for fields parallel to the current is more difficult to understand. The irreversibility field at low temperatures is $\sim$2 T and for higher fields the magnetization is uniform and (due to the low temperature) very close to the saturation value. Therefore, the question is what is changing in this high-field range that yields the non-saturating negative magnetoresistance? As the magnitude

of the magnetization does not change much, we believe that this effect is produced by magnetization rotation together with a strong anisotropic magnetoresistance. As shown in figure 1(a), the easy axis is at an angle with the plane of the film where the current flows. Since the anisotropy field is at least several teslas, it is expected that the angle between the current and the moment will continue to change for fields much higher than 2 T. Such a scenario requires, however, unusually large anisotropic magnetoresistance (commonly of the order of a few per cent), which is probably related to the above-mentioned large spin–orbit coupling.

Another intriguing property is revealed in the low-field magnetoresistance after cooling the sample in zero field. The initial change in resistivity is clearly related to removal of domain walls. When the field is swept back and forth we do not recover the initial resistivity, since sweeping the field does not introduce as many domain walls as were initially in the zero-field-cooled sample. Two interesting features are noted: the unusually large domain-wall-related resistivity and the strong dependence on the direction of the current—that is, the removal of domain walls reduces the resistance for current in the c-direction and increases the resistance for current in the $[\bar{1}10]$ direction. Preliminary transmission electron microscopy measurements [18] reveal stripe domain walls which run perpendicular to the c-axis. Thus, the anisotropic behaviour of the resistivity correlates with the anisotropic domain structure. The magnitude of the negative magnetoresistance for currents crossing the domain walls may be connected to the high polarization of the conduction band [10, 21]; and the positive (albeit smaller) magnetoresistance for currents running parallel to the domain walls may be related to the anisotropic magnetoresistance and to an averaged smaller resistivity for such currents, due to the change of the angle between the current and the magnetic moments in the domain walls.

5. Discussion

On the basis of the critical behaviour of the resistivity, we clearly see that it is very different from that of good metallic ferromagnets; and by inspecting the magnetic phase transition at the same time, we have deduced that this is a transport and not a magnetic anomaly. Is the anomalous transport behaviour restricted to the vicinity of T_c? Probably not. The non-saturating resistivity through the Ioffe–Regel limit at high temperature combined with the Kondo-like resistivity minima and apparent high sensitivity to disorder at low temperatures suggest that the transport is unusual over the entire temperature range.

What makes these results even more intriguing is their relevance beyond itinerant ferromagnetism in 'bad metals', which is an interesting issue by itself. An entire family of 'bad metals' appears to exist, which includes HTS, fullerenes and organic conductors. All of them are 'bad metals' in the $k_F l = \mathcal{O}(1)$ limit and are characterized by high-temperature resistivity which is almost linear in T and does not saturate as it passes through the Ioffe–Regel limit of $k_F l = 2\pi$. Therefore, assuming that this similarity is not accidental, our conclusions regarding $SrRuO_3$ may be generalized to other 'bad metals' as well.

The high-temperature resistivity is not only a characteristic feature, but is the focus of controversy, since its microscopic origins are unclear. Adding to the confusion is the difficulty of unequivocally excluding the validity of a Bloch–Grüneisen fit. For $SrRuO_3$, however, the situation is clearer due to the anomalous behaviour at T_c. As we discussed above, deviation from the expected critical behaviour is strong evidence for the inapplicability of the Boltzmann equations and/or for a contribution of a non-phononic source (beyond those expected from impurities and single-spin scattering). This observation reinforces the opinion that also in HTS, for example, a Bloch–Grüneisen fit for the high-

temperature resistivity is inadequate.

The sharp drop in resistivity near T_c and the large negative magnetoresistance are reminiscent of properties of manganites [8] and Eu-rich EuO [9] for which the ferromagnetic phase transition is associated with a metal–insulator transition. If this is relevant it may suggest high sensitivity to alignment of neighbouring moments. On the other hand, recent experiments show a dramatic increase in the scattering time of quasiparticles just below T_c in HTS [38], and this has been interpreted as evidence for an electronic origin for the normal-state resistivity. Since in our case the electronic states are also affected at T_c due to Stoner splitting, the possibility of a similar scenario is intriguing.

The low-temperature resistivity, and particularly the presence of resistivity minima, implies a tendency of states to become localized. Here, the magnetic nature of the localized states and the finite resistivity at low temperatures enables their clear detection due to the Kondo effect, but it may reflect an inherent tendency of states to become localized in other 'bad metals'. The commonly observed Schottky anomaly in specific heat measurements of HTS may be a similar manifestation of this tendency.

Currently, $SrRuO_3$ appears unique in the possibility that it offers, the possibility of studying itinerant magnetism in the $k_F l = \mathcal{O}(1)$ limit in a clean and ordered compound with well characterized magnetic properties. While still much more experimental and theoretical work on this compound is warranted, clearly identifying other magnetic systems with similar properties is important in looking for common characteristics of bad metallic ferromagnets in particular and bad metals in general.

Acknowledgments

We acknowledge useful discussions with P B Allen, V J Emery, and S A Kivelson. This work was supported by the DOE Grant No DE-FG03-94ER-45528 and by the AFOSR Grant No F49620-95-1-0039. The films were characterized in the Center for Materials Research at Stanford University.

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