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Published in:
Physical Review B (Condensed Matter and Materials Physics)

Link to article, DOI:
10.1103/PhysRevB.75.060403

Publication date:
2007

Document Version
Publisher's PDF, also known as Version of record

PHYSICAL REVIEW B **75**, 060403(R) (2007)

Competing magnetic fluctuations in $Sr_3Ru_2O_7$ probed by Ti doping

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(Received 21 December 2006; published 7 February 2007)

We report the effect of nonmagnetic Ti^{4+} impurities on the electronic and magnetic properties of $Sr_3Ru_2O_7$. Small amounts of Ti suppress the characteristic peak in magnetic susceptibility near 16 K and result in a sharp upturn in specific heat. The metamagnetic quantum phase transition and related anomalous features are quickly smeared out by small amounts of Ti. These results provide strong evidence for the existence of competing magnetic fluctuations in the ground state of $Sr_3Ru_2O_7$. Ti doping suppresses the low-temperature antiferromagnetic interactions that arise from Fermi surface nesting, leaving the system in a state dominated by ferromagnetic fluctuations.

DOI: 10.1103/PhysRevB.75.060403 PACS number(s): 75.30.Kz, 71.27.+a, 75.40.Cx

The Ruddlesden-Popper-type perovskite strontium ruthenates $Sr_{n+1}Ru_nO_{3n+1}$ show a great diversity of electronic and magnetic ground states. Sr_2RuO_4 ($n=1$), the most widely studied member of the series, is an unconventional superconductor with a spin-triplet pairing.[1–3] $SrRuO_3$ ($n=\infty$) is an itinerant ferromagnet with a Curie temperature of 160 K.[4] The $n=2$ bilayered member $Sr_3Ru_2O_7$ has an intermediate dimensionality between these two; its magnetic ground state was identified as an exchange-enhanced paramagnet.[5] Moderate applied fields induce a metamagnetic transition in this material, with the transition field ranging from 4.9 T (for $H\|ab$) to 7.9 T (for $H\|c$).[6,7]

The metamagnetism in $Sr_3Ru_2O_7$ has generally been interpreted as a field-tuned Stoner transition into a highly polarized magnetic state; since there is no broken symmetry, this transition is expected to be first order.[8,9] In the phase diagram one will thus have a first-order phase boundary line which terminates in a critical end point;[10] there is considerable experimental evidence that the characteristic temperature of this end point is close to zero, indicating the presence of quantum criticality.[7] A variety of unusual features, including non-Fermi liquid behavior and a possible novel phase at the critical point, have been observed in this material.[6,11,12] This finding has generated a great deal of interest and opened new routes to exploring the novel physics of quantum criticality.

Based on the metamagnetic features, we might expect that the relevant magnetic correlations in $Sr_3Ru_2O_7$ are predominantly ferromagnetic in nature.[10] However, inelastic neutron scattering measurements have revealed that the system is dominated by two-dimensional (2D) ferromagnetic (FM) fluctuations only at high temperatures, crossing over to 2D antiferromagnetic (AFM) fluctuations for temperatures below 20 K.[13] Furthermore, recent ^{17}O-NMR measurements have shown that AFM fluctuations dominate the spin fluctuation spectrum near the critical field.[14] This suggests a more complicated magnetic ground state for $Sr_3Ru_2O_7$, in which band FM correlations and AFM correlations due to nesting effects might be in competition. Thus, further experiments to elucidate the low-temperature magnetic ground state are highly desirable.

Ti doping has been used as an effective probe of magnetic correlations in the single-layered ruthenate Sr_2RuO_4. There, Ti was found to enhance the anisotropic, incommensurate AFM fluctuations, ultimately giving rise to a spin-density-wave ordering with the same nesting wave vector seen in the undoped material.[15–19] Thus, to further probe the magnetic correlations in $Sr_3Ru_2O_7$, we have investigated the effects of doping nonmagnetic Ti in $Sr_3Ru_2O_7$. In contrast to Sr_2RuO_4, small levels of Ti suppress the AFM correlations, leaving the system in a state with FM fluctuations. This provides strong evidence for the existence of competing magnetic fluctuations in the ground state of pure $Sr_3Ru_2O_7$.

We have grown a series of Ti-doped crystals $Sr_3(Ru_{1-x}Ti_x)_2O_7$ using a floating-zone technique; growth conditions were similar to those previously reported for pure $Sr_3Ru_2O_7$.[20] Crystals selected for the measurements were characterized by x-ray diffraction and did not include any impurity phase of Sr_2RuO_4 or $Sr_4Ru_3O_{10}$. Magnetization measurements were taken in a superconducting quantum interference device (SQUID) magnetometer and specific heat was measured by a standard thermal relaxation method (Quantum Design, model PPMS). Resistivity measurements were performed with a standard four-probe technique.

In Fig. 1 we present the temperature dependence of susceptibility $\chi(T)=M/H$ for $Sr_3(Ru_{1-x}Ti_x)_2O_7$ for (a) in-plane and (b) out-of-plane field orientations. For the undoped sample, we observe behavior similar to that previously reported by Ikeda *et al.*;[5] susceptibility shows a peak around 16 K for both field orientations. The origin of this peak is likely due to a crossover in the nature of magnetic correlations; inelastic neutron scattering also shows a peak near 16 K for the dynamic susceptibility χ'' near a FM wave vector, while AFM correlations increase rapidly below this temperature.[13] Recent NMR results further suggest that below ~16 K FM correlations are quenched and 2D incom-

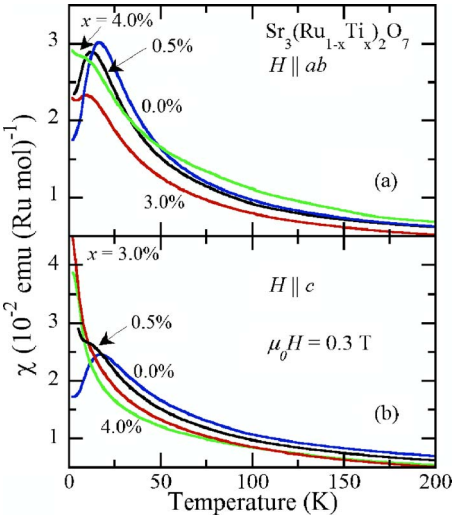

FIG. 1. (Color online) Temperature dependence of the uniform susceptibility $\chi=M/H$ of Ti-doped $Sr_3Ru_2O_7$ for (a) in-plane and (b) out-of-plane fields, taken using an excitation field of $\mu_0 H=0.3$ T.

mensurate AFM fluctuations become dominant.[14]

We now consider the evolution of this feature in susceptibility under Ti doping. For $H\|ab$ [Fig. 1(a)], Ti impurities result in a suppression of the peak near 16 K; specifically, the low-temperature susceptibility increases until, by 4.0% doping, only a small remnant of the peak remains. This trend is even more remarkable for fields $H\|c$, shown in Fig. 1(b); by 4.0% doping the peak is completely suppressed and the susceptibility is reminiscent of a paramagnetic state. These features suggest a change in the magnetic ground state such that 2D AFM correlations are no longer dominant.

Above ~180 K the susceptibility shows Curie-Weiss behavior. Table I shows the result of a fit to the usual expression $\chi(T)=\chi_0+C/(T-\Theta_W)$ for the samples shown in Fig. 1 for an in-plane field, where χ_0 is the temperature-independent term and $C/(T-\Theta_W)$ is the Curie-Weiss term. The Weiss temperature Θ_W increases with Ti doping, consistent with the idea that the system is moving toward a FM ground state. However, the effective moment p_{eff} derived from C does not change remarkably with doping, which may suggest that Ti substitution is acting primarily to suppress the low-temperature AFM correlations rather than directly enhancing the ferromagnetism (see below for further discussion).

The enhanced Stoner model of itinerant metamagnets by Yamada gives a certain criterion for a first-order metamagnetic transition based on a Landau expansion of the free energy.[9] Specifically, given the expansion $H=aM+bM^3+cM^5$, where H is the derivative of the free energy with respect to the magnetization, the ratio ac/b^2 should be less than 0.45 for a first-order metamagnetic transition to occur. This ratio is related to the susceptibility by the expression $ac/b^2=(5/28)[1-\chi(0)/\chi(T_{max})]^{-1}$ where T_{max} is the temperature where the peak in susceptibility occurs. For our samples, this ratio is 0.423 for pure $Sr_3Ru_2O_7$, 0.80 for 0.5% doping, and 6.13 for 3% doping. While the magnetic properties of $Sr_3(Ru_{1-x}Ti_x)_2O_7$ are likely more complex than a simple Stoner picture, this analysis suggests that Ti doping quickly moves the system away from conditions favorable for metamagnetism.

TABLE I. Parameters from Curie-Weiss fits, $H\|ab$, χ_0 in the units of emu/(Ru mol).

x	Currie constant C	χ_0 ($\times 10^{-3}$)	Θ_W (K)	p_{eff} (μ_B)
0.0	0.79	2.7	−21.6	2.5
0.005	0.66	3.0	−12.3	2.3
0.03	0.59	2.2	4.31	2.2
0.04	0.67	3.4	9.43	2.3

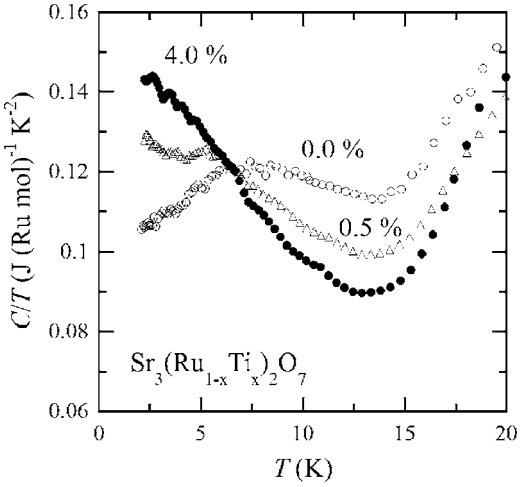

FIG. 2. The specific heat divided by temperature for Ti-doped $Sr_3Ru_2O_7$ at low temperatures.

Measurements of the specific heat give strong evidence that Ti doping leaves the system in a ground state dominated by FM fluctuations. In Fig. 2 we present the specific heat divided by temperature for the identical samples used in magnetization measurements. All curves were taken at zero field. The undoped sample is consistent with previously reported results by Perry *et al.*;[6] C/T shows an upturn close to 15 K, followed by a peak at lower temperatures. Ti doping suppresses this low-temperature peak, such that by 4% doping we observe a rapid increase in C/T as temperature decreases.

While low-temperature behavior consistent with a Schottky anomaly has been previously reported in pure $Sr_3Ru_2O_7$, it occurs only below 0.2 K and is likely not the source of the upturn observed here.[21] Under an applied magnetic field, C/T in pure $Sr_3Ru_2O_7$ also shows a sharp rise at low temperatures, consistent with a $\ln(T)$ divergence due to proximity to a quantum critical point (QCP).[6,21] In our case we have observed no other features which might suggest the presence of a doping-induced QCP; rather, we believe the upturn in specific heat arises from FM fluctuations. Further discussion of the origin of this feature is given below.

We next consider how the features of the metamagnetic transition change with doping. In Fig. 3 we present the magnetization and normalized resistance of Ti-doped $Sr_3Ru_2O_7$ for $H\|ab$. Consistent with previous reports, for the undoped

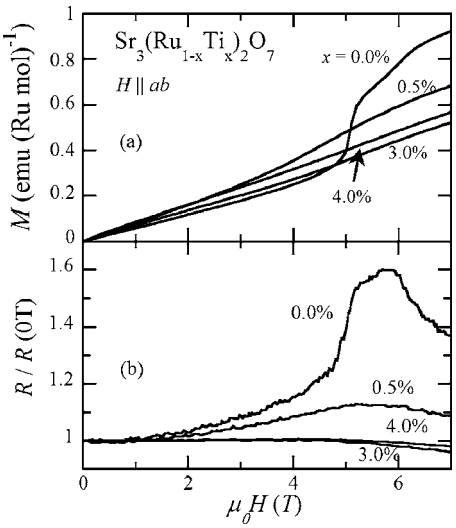

FIG. 3. (a) Magnetization versus field and (b) resistance (normalized to its 0 T value) versus field for the $Sr_3(Ru_{1-x}Ti_x)_2O_7$ system. Magnetization is taken at 2 K and resistance is measured at 0.3 K.

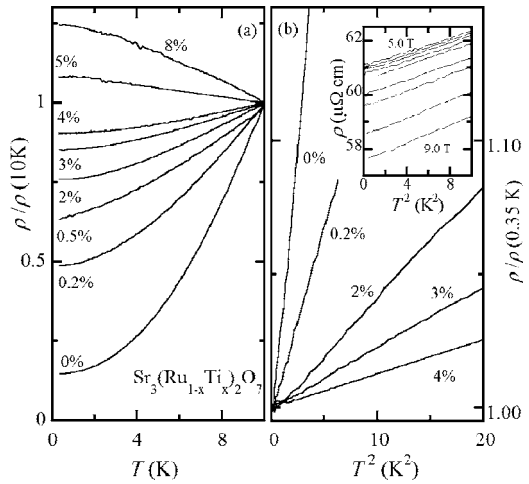

FIG. 4. (a) The temperature dependence of the resistivity for several Ti doping levels, normalized to the resistivity at 10 K. (b) Resistivity (normalized to its 0.3 K value) plotted versus T^2 for metallic doped samples. The data of the x=0.5% sample are not shown here, since they nearly overlap with those of the pure sample. This is likely due to Ti inhomogeneity. Inset: resistivity of the 3.0%-doped sample versus T^2, under a range of applied fields, $\mu_0 H$=5.0, 4.0, 5.5, 3.0, 6.0, 0.0, 7.0, 8.0, and 9.0 T, from top to bottom. $I \| H \| ab$.

sample we observe a superlinear rise in magnetization and a broad peak in resistance around the critical field. However, Ti impurities act to rapidly suppress both of these features; even 0.5% doping has nearly smeared out the metamagnetic features in magnetization and resistance. By doping levels of 3.0% and 4.0%, magnetization is nearly linear in field and only a very small feature is seen in resistance near 5 T. This observation is in agreement with Yamada's criterion for metamagnetism to occur (see above).

One of the most prominent features of the metamagnetic transition in $Sr_3Ru_2O_7$ is the non-Fermi-liquid behavior close to the critical field, manifested as a linear temperature dependence of resistivity.[6] In Fig. 4 we examine the electronic ground state of Ti-doped $Sr_3Ru_2O_7$ by means of bulk resistivity measurements. Figure 4(a) shows the temperature dependence of the resistivity (normalized to its value at 10 K) for several doping levels; the system remains metallic until approximately 5% doping, where it transitions into a localized state. The metallic ground state remains a Fermi liquid, with resistivity proportional to T^2 at low temperatures [see Fig. 4(b)].

At a 3% doping level there is still a small peak in the in-plane susceptibility (see Fig. 1), but the metamagnetic transition is almost completely suppressed and there is only a small downturn in resistivity at 5 T. The inset of Fig. 4(b) displays the resistivity of this 3% sample plotted against T^2 at various applied fields for the configuration $H \| ab$. We can see that the T^2 behavior of ρ at low temperatures is unchanged by an applied field, even close to where the metamagnetic transition occurs in the pure sample; this indicates that quantum criticality is fully suppressed.

We now wish to discuss the origin of these features in $Sr_3(Ru_{1-x}Ti_x)_2O_7$. We begin by noting similarities between our results and previous studies on other itinerant metamagnets; for example, the intermetallic compound $Y(Co_{1-x}Al_x)_2$ has a transition from an itinerant metamagnetic state to a weakly FM one at a doping level of x=0.13.[22] For doping levels approaching 0.13, there is a suppression of the characteristic peak in susceptibility,[23] a strong upturn in C/T,[24] and a significant broadening of the metamagnetic transition.[25] These features have been successfully interpreted quantitatively within the self-consistent renormalized (SCR) theory of spin fluctuations as evidence for proximity to a weakly FM state.[26]. For $Sr_3(Ru_{1-x}Ti_x)_2O_7$, as described above, Ti doping results in similar changes as those seen in $Y(Co_{1-x}Al_x)_2$, clearly suggesting that FM fluctuations become dominant. In $Y(Co_{1-x}Al_x)_2$, the FM enhancement is believed to be associated with doping-induced structural changes which increase the density of states at the Fermi level.[22] However, in the case of $Sr_3(Ru_{1-x}Ti_x)_2O_7$, substituting a small amount of Ti for Ru should have only a very small effect on the structure since Ti^{4+} has a very similar ionic radius as Ru^{4+}. We think that the most likely mechanism for the Ti-doping effect in $Sr_3Ru_2O_7$ is that both AFM and FM fluctuations coexist in the ground state of the undoped sample and that Ti doping primarily suppresses the AFM correlations. This point of view has recently been confirmed by neutron scattering measurements on Ti-doped $Sr_3Ru_2O_7$ samples.[27] In addition, this experiment also reveals that while the FM fluctuations survive in Ti-doped $Sr_3Ru_2O_7$, they are weakened in intensity. This result is consistent with the magnetization data in Fig. 3(a) which shows that the doped samples have smaller magnetization at higher fields than the pure sample.

The existence of competing magnetic correlations in $Sr_3Ru_2O_7$ is also consistent with the results of band structure calculations.[28,29] The bands derived from the Ru $4d_{xy}$ orbitals are close to a Van Hove singularity and thus close to a FM Stoner instability, while bands derived from the $4d_{xz}$ and $4d_{yz}$ orbitals have nesting features which would favor AFM

fluctuations.[13,29] The Ti doping reported here appears to be band selective, primarily affecting the character of the bands derived from $4d_{xz,yz}$ orbitals. This is similar to Ti doping in the related compound Sr_2RuO_4, in which small amounts of Ti have only minimal effects on the rotation of the octahedra and the main γ band, but do remarkably affect the character of the 1D α and β bands in such a way to enhance the nesting features of the Fermi surface and thus enhance the AFM correlations.[15,16,18] In the case of $Sr_3Ru_2O_7$ it appears that Ti impurities result in a reduction of the nesting features, which is reasonable since the characteristics of the bands involved in nesting are different between the two materials.[29,30]

As mentioned above, the standard picture for metamagnetism in $Sr_3Ru_2O_7$ is a field-tuned Stoner transition, which is supported by the de Haas–van Alphen (dHvA) experiment on $Sr_3Ru_2O_7$.[31] Nevertheless, an NMR experiment indicates that the quantum fluctuations near the critical field are AFM in nature,[14] rather than FM as expected for a system near the Stoner instability. It has been suggested that while the metamagnetic transition in $Sr_3Ru_2O_7$ is driven by the field-induced Stoner instability of the Fermi surface, the AFM fluctuations could be enhanced by coupling with the quantum fluctuations of the Fermi surfaces.[14] This argument is reasonable in the frame of orbital-dependent magnetic fluctuations discussed above as long as interband coupling is considered. The suppression of the metamagnetic transition by Ti doping can be ascribed to the localization effect induced by doping. As shown in Fig. 4(a), Ti-doping levels of 5% and above result in a localized state, and this trend towards localization would drive the system away from the Stoner instability, thereby reducing the metamagnetic features.

In conclusion, we have investigated the properties of Ti-doped $Sr_3Ru_2O_7$. We observe evidence that the short-range AFM correlations are quickly suppressed by small amounts of Ti, leaving the system in a state dominated by 2D FM correlations. The most likely mechanism for this is that Ti doping significantly alters the bands derived from the Ru $4d_{xz,yz}$ orbitals, which results in a suppression of AFM fluctuations due to nesting; however, it has less effect on the FM correlations arising from the bands derived from the Ru $4d_{xy}$ orbitals. Ti doping thus confirms previous suggestions about the presence of competing magnetic interactions in pure $Sr_3Ru_2O_7$. This result offers additional insight into the multiband nature of magnetic correlations in this material.

We would like to thank A. Mackenzie, Y. Maeno, and Y. Liu for useful duscussions and D. Niebieskikwiat for technical assistance. This work was supported by the Louisiana Board of Regents support fund LEQSF(2003-06)-RD-A-26 and (2005-06)-ENH-TR-78 and pilot fund NSF/LEQSF(2005)-Pfund-23 at Tulane, the DOE under Grant No. DEFG02-91-ER45439 at the University of Illinois, DARPA Grant No. MDA972-02-1-0012 at UNO, and the National Science Foundation of China (No. 10225417) and the National Basic Research Program of China (No. 2006CB601003) at Z.U. Z.Q.M. thanks the Research Corporation for financial support.

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