

Magnetic resistivity in SrRuO₃ and the ferromagnetic phase transition

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We measured the resistivity of the itinerant ferromagnet SrRuO₃ as a function of temperature ($120\text{ K} < T < 180\text{ K}$) and magnetic field ($0 < H < 10\text{ kOe}$) in the vicinity of the ferromagnetic phase transition ($T_c \sim 150\text{ K}$). We find that the magnetic resistivity ρ_m is related to the magnetization M by $\rho_m(M) = \rho_m(0) - aM^2$ over a wide range of M . From the analysis of the resistivity data we determine the critical parameters of the phase transition $\beta = 0.34 \pm 0.02$, $\gamma = 1.14 \pm 0.07$ below T_c , $\gamma = 1.17 \pm 0.14$ above T_c , $C^+/C^- \sim 4$. Here β and γ are the critical exponents of the spontaneous magnetization and the susceptibility, respectively, and C^+/C^- is the amplitude ratio of the susceptibility. Our results are supported by a collapse of the magnetoresistance data on a single curve when an appropriate scaling function is used.

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I. INTRODUCTION

SrRuO₃ is an itinerant ferromagnet ($T_c \sim 150\text{ K}$) with intriguing transport properties: the high-temperature resistivity crosses the Ioffe-Regel limit,¹ the temperature derivative of the magnetic-related resistivity near T_c strongly deviates from the expected specific-heat-like behavior,² and the infrared conductivity indicates incoherent transport near T_c .³ These properties are our main motivation to explore the interplay between magnetism and transport and to determine as precisely as possible the nature of the ferromagnetic phase transition in this compound.

While efforts have been made in the past to study the magnetoresistance in thin films of SrRuO₃,⁴⁻⁷ here we report on measurements of single-phase films where the magnetic field is applied along the (single) easy axis; thus ensuring that the magnetization does not change its axis of orientation when a magnetic field is applied. In addition, we concentrate here on the low field range ($< 10\text{ kOe}$) to allow the investigation of the true critical behavior (namely, beyond mean field). Furthermore, we establish experimentally that a simple relation exists between the magnetic resistivity and the magnetization, which allows us to investigate the magnetic critical behavior by measurements of resistivity. Studying the magnetic phase transition in thin films via transport measurements has special advantages relative to bulk measurements of magnetization that suffer from weak signals on top of large contribution of the substrate (which is temperature and field dependent).

Assuming that the resistivity can be separated into magnetic (i.e., related to spin scattering) and nonmagnetic parts, the application of a magnetic field in the explored temperature range ($120\text{--}180\text{ K}$) is expected to affect the magnetic part while its effect on the nonmagnetic part (through the Lorentz force) is expected to be negligible due to the relatively short mean free path.^{8,19,20} For that reason, the magnetoresistance (i.e., the change in the resistivity upon applica-

tion of field) manifests the interplay between magnetism and transport in this compound.

We show that in a wide range of magnetizations the magnetic part of the resistivity ρ_m is related to the magnetization M by $\rho_m(M) = \rho_m(0) - aM^2$, where a is a constant (which we determine from a comparison with measurements of magnetization). Based on this relation, we use resistivity and magnetoresistance measurements to determine the values of the critical exponents β and γ (which describe the critical behavior of the spontaneous magnetization and the initial susceptibility, respectively). We determine γ independently above and below T_c . With these exponents we successfully scale the entire magnetoresistance data (except for temperatures $|T - T_c| < 3\text{ K}$). We discuss, in view of the determined critical exponents, the universality class of the magnetic phase transition in SrRuO₃.

II. SAMPLES

Our measurements are done on thin films of SrRuO₃ grown on slightly miscut ($\sim 2^\circ$) substrates of SrTiO₃ by reactive electron beam evaporation. These are untwinned single-crystal films⁹ in an orthorhombic phase, with the lattice parameters of $a \approx 5.53\text{ \AA}$, $b \approx 5.57\text{ \AA}$, and $c \approx 7.85\text{ \AA}$. The [001] direction is in the plane of the thin film, and the easy axis of the magnetization is approximately in the [010] direction, which is at 45° out of the plane of the film. The direction of the easy axis changes continuously with temperature,¹⁰ in the (001) plane. The spontaneous magnetization in the zero-temperature limit is nearly $1.4\mu_B/\text{Ru}$ in films and $1.6\mu_B/\text{Ru}$ in bulk, while the paramagnetic moment is $2\mu_B/\text{Ru}$.¹⁰ The thickness of the sample whose measurements are shown in this work is 2000 \AA . The zero-field resistivity near T_c ($\sim 153\text{ K}$) is around $130\text{ }\mu\Omega\text{ cm}$ and the residual resistivity is $\sim 4.6\text{ }\mu\Omega\text{ cm}$.

III. EXPERIMENT

We measured the resistivity of SrRuO₃ as a function of temperature and magnetic field near the ferromagnetic phase transition ($T_c = 153$ K) with temperature stability of ± 0.01 K and field reproducibility of ± 10 Oe. The magnetic field was applied in the (001) plane, close to the easy axis of the magnetization (approximately the [010] direction) and the current was in the [001] direction. In this configuration the magnetization \mathbf{M} is parallel to the easy axis when a field is applied and the current is always perpendicular to \mathbf{M} . The advantages of this configuration are as follows. (a) We avoid changes in the *direction* of \mathbf{M} which could induce changes in resistivity due to the anisotropic magnetoresistance (AMR) effect. (b) Even if small changes in the direction of \mathbf{M} do occur, the angle of \mathbf{M} relative to the current does not change, since both the easy axis and the applied field are in a plane perpendicular to the current. (c) The magnetocrystalline anisotropy does not compete with the applied field, since the field is applied along the easy axis, and it is always the full field which determines the degree of magnetic order.

Measurements below T_c were done after a sufficiently high field (~ 20 kOe) was applied to saturate the sample. Then the sample remains saturated even at low fields. The domains in SrRuO₃ renucleate only when a specific field in the opposite direction is applied, as it was observed in TEM measurements,⁹ except a few degrees below T_c where the domains renucleate at a low positive field. Since the field at which the nucleation starts is clearly observed in the sweeps of resistivity versus field, we were able to assure that our measurements do not involve effects of changes in the domain structure or domain-wall resistivity,¹¹ but reflect solely the changes in the magnetization.

IV. DATA ANALYSIS

A. Zero-field resistivity and the critical exponent β

The zero-field resistivity near T_c is shown in Fig. 1. Since above T_c the magnetization vanishes, the magnetic resistivity induced by spin scattering is expected to become temperature independent far enough above T_c . Therefore, we may assume that the temperature dependence of the resistivity there is only due to nonmagnetic components of the resistivity. Assuming that the behavior of the *nonmagnetic* resistivity is not affected by the ferromagnetic phase transition, its behavior below T_c can be approximated (in some small range of temperatures) by an extrapolation of the resistivity above T_c , which we denote by $\rho^+(T)$. We then subtract the measured resistivity below T_c from the extrapolated $\rho^+(T)$ (based on a linear fit of the range 160–170 K) and denote the difference by $\Delta\rho_{sp}$ (see Fig. 1). This difference is related to magnetic ordering, and in the following we determine its functional dependence on the magnetization M .

Figure 2 shows a plot of $\ln \Delta\rho_{sp}$ as a function of $\ln |t|$, where $t = (T - T_c)/T_c$, and $T_c = 153$ K (see the Appendix). The temperature range used in this fit is 130–149 K. The linearity of the plot in Fig. 2 clearly indicates that $\Delta\rho_{sp}$ exhibits a power-law behavior as a function of $|t|$:

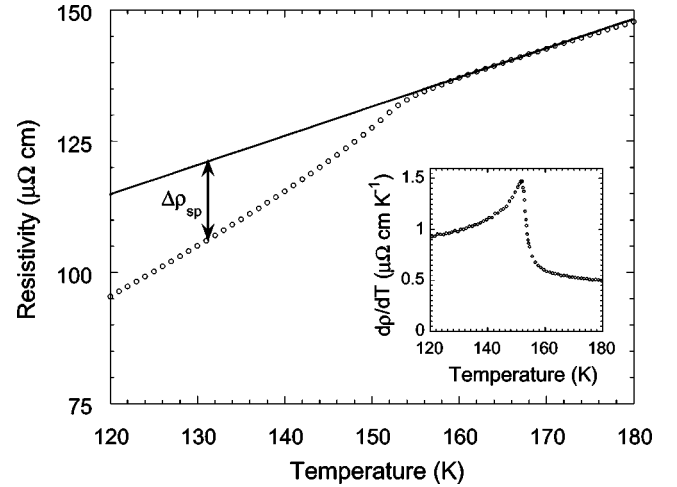


FIG. 1. Temperature dependence of zero-field resistivity ρ near T_c (~ 153 K). The solid line is the extrapolation of the resistivity from $T > T_c$. The definition of $\Delta\rho_{sp}$ is shown. The inset shows the behavior of dp/dT .

$$\Delta\rho_{sp} \propto |t|^s, \quad (1)$$

where $s = 0.68$ is the slope of the plot.

If the magnetic resistivity ρ_m depends only on the magnetic ordering, we can expand the magnetic resistivity in a power series in M around $\rho_m(0)$, which is the magnetic resistivity when no magnetic order exists. Due to symmetry, only even powers of M appear in the expansion

$$\rho_m(M) = \rho_m(0) - aM^2 - bM^4 + \dots \quad (2)$$

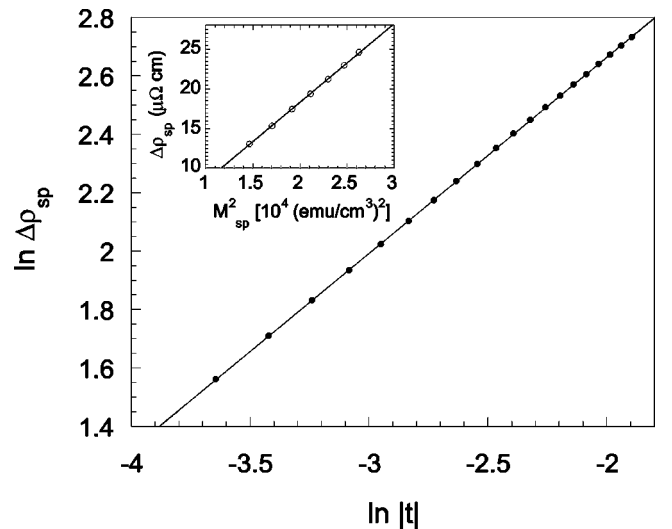


FIG. 2. Critical behavior of $\Delta\rho_{sp}$ with respect to the reduced temperature $t = (T - T_c)/T_c$ from 130 to 149 K with $T_c = 153$ K. A linear fit is shown. The inset shows the correspondence between $\Delta\rho_{sp}$ and the square of the measured spontaneous magnetization M_{sp} .

On the other hand, we assume that $\Delta\rho_{sp}$ is simply

$$\Delta\rho_{sp}(T) = \rho_m(0) - \rho_m[M_{sp}(T)], \quad (3)$$

where $M_{sp}(T)$ is the spontaneous magnetization. Thus we obtain

$$\Delta\rho_{sp} = aM_{sp}^2 + bM_{sp}^4 + \dots \quad (4)$$

In the vicinity of the critical point, M_{sp} is expected to exhibit a power-law behavior of the form $M_{sp} \propto |t|^\beta$. Therefore we suggest, on the basis of the result in Eq. (1), that in our case $\Delta\rho_{sp}$ is well described by the first term alone of Eq. (4) and then it follows that

$$\Delta\rho_{sp} \propto |t|^{2\beta}, \quad (5)$$

where β is the critical exponent of the magnetization.

This implies that β can be found from a plot of $\ln \Delta\rho_{sp}$ as a function of $\ln|t|$, as in Fig. 2. Hence we obtain $\beta=0.34$, which is a reasonable value for this exponent. (Usually β is found to have values between 0.3 and 0.5. See, e.g., Refs. 12–15.)

To determine more accurately the values of T_c and β we plotted $\Delta\rho_{sp}^{1/(2\beta)}$ as a function of T for trial values of β until the best linear dependence was obtained. From the intercept of the line with the T axis we found T_c . In order to check the reliability of the results we followed the same procedure for different ranges of extrapolation of $\rho^+(T)$ and for different ranges of fitting (see the Appendix). From this analysis we find that $T_c = 153.0 \pm 0.5$ K, and $\beta = 0.34 \pm 0.02$.

Comparing these measurements with measurements of spontaneous magnetization on the same film we find that $\Delta\rho_{sp} = aM_{sp}^2$ (see inset to Fig. 2) with $a = (9.5 \pm 1.0) \times 10^{-4} \mu\Omega \text{ cm}/(\text{emu}/\text{cm}^3)^2$. After checking the resistivity of other films, with different thicknesses (down to 100 Å) and different residual resistivities (up to 30 times higher than in the film described here), we find that the value of a is quite insensitive to these parameters (the observed changes were less than 20%).

B. Magnetoresistance and the critical exponent γ

The previous subsection implies that

$$\rho_m(M) = \rho_m(0) - aM^2. \quad (6)$$

This relation enables us to find the magnetic susceptibility from the measurements of magnetoresistance.

The initial susceptibility χ_0 (defined as $\partial M/\partial H$ at $H=0$) is expected to exhibit a power-law behavior near T_c :

$$\chi_0 = C^\pm |t|^{-\gamma}, \quad (7)$$

where C^+ and C^- are the amplitudes above and below T_c , respectively, and γ is the critical exponent. In the following we determine γ separately from measurements above and below T_c and also calculate the value of the amplitude ratio C^+/C^- .

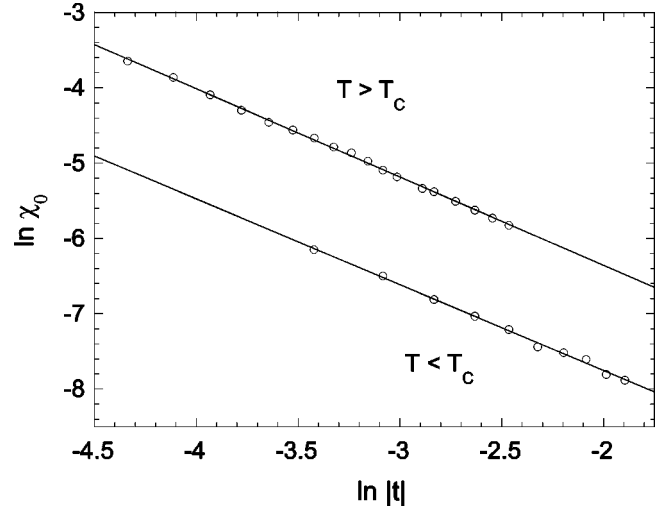


FIG. 3. Critical behavior of the initial susceptibility χ_0 below and above T_c . The critical exponent γ is found from the slopes of the linear fits.

Determination of γ above T_c . We can expand the magnetic field H (at a constant temperature) as a power series in M :

$$H = a_1 M + a_2 M^3 + \dots, \quad (8)$$

where a_1 and a_2 are temperature-dependent constants. For small fields, when the first two terms of the expansion are sufficient, we obtain

$$\frac{H}{M} = a_1 + a_2 M^2. \quad (9)$$

For $M \rightarrow 0$, Eq. (9) reduces to $a_1 = H/M$. Therefore from the definition of the initial susceptibility χ_0 it follows that $\chi_0(T) = 1/a_1(T)$. We determine M from measurements of magnetoresistance according to Eq. (6). When plotting M^2 vs H/M , the values of $a_1(T)$ can be found from the intercept with the H/M axis. Then the critical exponent γ is found from the slope of a log-log plot of χ_0 vs $|t|$, according to the definition of γ in Eq. (7); see Fig. 3. The plot includes the temperatures 155–166 K. The value of γ found by this method is $\gamma = 1.17 \pm 0.14$.

Determination of γ below T_c . At temperatures below T_c a linear response of the resistivity to a low field is observed. Since thin films of SrRuO₃ remain in a single-domain state even at low fields (see Sec. III), it is an intrinsic effect of H on M (not related to changes in the domain structure). Thus $\partial M/\partial H$ at low fields is the initial susceptibility χ_0 . The quantity $\partial M/\partial H$ can be found from the measured $\partial\rho/\partial H$, since using Eq. (6) we can write

$$\frac{\partial\rho}{\partial H} = -2aM_{sp} \frac{\partial M}{\partial H}, \quad (10)$$

where the value of M_{sp} at each temperature can be obtained from the measurements of $\Delta\rho_{sp}$ (based on the relation $\Delta\rho_{sp} = aM_{sp}^2$). We calculated $\partial\rho/\partial H$ from measurements between 130–148 K. At each temperature we used the range of magnetoresistances from 0.08 to 0.16 $\mu\Omega$ cm, where the fields were small enough so that $\partial M/\partial H$ is approximately χ_0 , and high enough so that even at temperatures close to T_c they are higher than the fields where the magnetization reversal starts. The value of γ found from the slope of a log-log plot of χ_0 vs $|t|$ (see Fig. 3) is $\gamma = 1.14 \pm 0.07$.

By comparing the susceptibilities above and below T_c we find that the amplitude ratio C^+/C^- is ~ 4 .

The effect of the demagnetizing field was included in our analyses;¹⁶ however, the correction was usually small.

C. Magnetoresistance data collapse

According to the scaling law hypothesis, the relation between the magnetic field H , the magnetization M , and the reduced temperature t in the critical region has the form

$$\frac{M}{|t|^\beta} = f_\pm \left(\frac{H}{|t|^{\beta+\gamma}} \right), \quad (11)$$

where f_\pm is a function that is different below and above T_c . Using Eq. (6) we obtain for the magnetoresistance

$$\begin{aligned} \Delta\rho(T,H) &= \rho(T,H) - \rho(T,0) \\ &= -a[M^2(T,H) - M^2(T,0)] \\ &= -a|t|^{2\beta} \left[f_\pm^2 \left(\frac{H}{|t|^{\beta+\gamma}} \right) - f_\pm^2(0) \right]. \end{aligned}$$

The expression within the square brackets is a function of $H/|t|^{\beta+\gamma}$ alone, therefore there exists a scaling law for the magnetoresistance of the form

$$\frac{\Delta\rho}{|t|^{2\beta}} = F_\pm \left(\frac{H}{|t|^{\beta+\gamma}} \right) \quad (12)$$

or

$$r = F_\pm(h), \quad (13)$$

where $r = |\Delta\rho|/|t|^{2\beta}$ and $h = H/|t|^{\beta+\gamma}$. When the correct values of β and γ are substituted, plotting the values of r as a function of h should give a smooth curve (with two branches, described by F_+ and F_- , for $T > T_c$ and $T < T_c$, respectively). The correction of the applied field for the demagnetization was not taken into account here.

Figure 4 shows the data collapse obtained with the values of the critical exponents and T_c found in this work ($T_c = 153$ K, $\beta = 0.34$ and $\gamma = 1.15$). This plot includes data for temperatures between 130 and 170 K (excluding the temperatures $|T - T_c| < 3$ K) and fields between 2 and 10 kOe. It is clearly seen that measurements at different temperatures fall on the same curve.

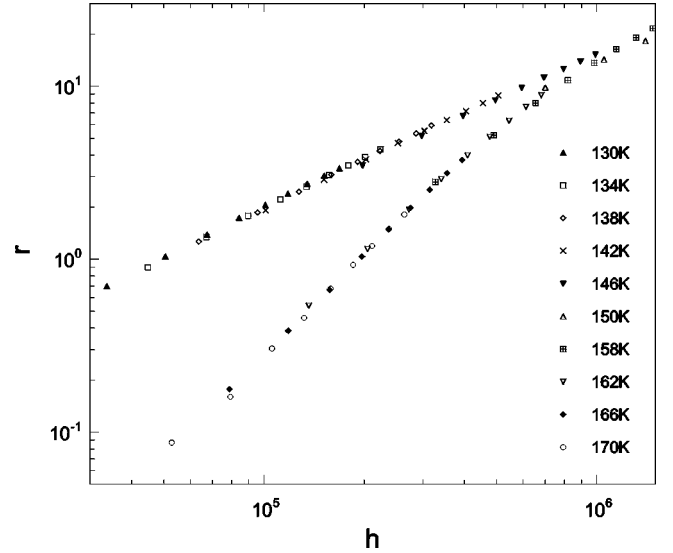


FIG. 4. Scaling of the magnetoresistance-temperature-field data. The quantities r and h are defined in the text. Data for fields between 2 and 10 kOe and temperatures between 130 and 170 K, see legend, is included.

V. SUMMARY AND CONCLUSIONS

From the behavior of the zero-field resistivity near the ferromagnetic phase transition we concluded that the dependence of the magnetic resistivity on the magnetization is $\rho_m(M) = \rho_m(0) - aM^2$. Based on this result and using measurements of resistivity and magnetoresistance we determined the values of the parameters β , γ , and C^+/C^- which describe the magnetic critical behavior in SrRuO₃. From the zero-field resistivity data we determined that $\beta = 0.34 \pm 0.02$. From the magnetoresistance data below T_c we found that $\gamma = 1.14 \pm 0.07$, and using the data above T_c we found that $\gamma = 1.17 \pm 0.14$. The consistency of values of γ obtained below and above T_c supports the validity of our analysis. Comparing the susceptibilities above and below T_c we obtained $C^+/C^- \sim 4$.

The current results are somewhat different from the ones presented in a previous report on the critical indices which relied on direct measurements of magnetization.¹⁰ While the zero-field resistivity below T_c gives a similar result for β , we obtain a different value for γ above T_c (which is determined from field-dependent measurements). We attribute the difference mainly to the fact that previously the field was not applied along the easy axis and therefore unwanted changes in the direction of the magnetic moment were involved.

In Table I we compare the extracted critical exponents with the exponents of the different theoretical models (mean-field, Ising, Heisenberg). The comparison suggests that SrRuO₃ belongs to Ising universality class. This result can be understood in view of the high uniaxial anisotropy of SrRuO₃ which persists also above T_c .¹⁷

It is important to note that the relation $\rho_m(M) = \rho_m(0) - aM^2$ is an approximation which cannot hold arbitrarily close to T_c , since it neglects the influence of spin-spin correlation on the resistivity. This correlation plays an important role very close to T_c as manifested in the divergence of

TABLE I. Comparison of critical parameters of SrRuO₃ with different theoretical models.

	β	$\gamma(T < T_c)$	$\gamma(T > T_c)$	C^+/C^-
Mean field theory	0.5	1	1	2
3D Ising model ^a	0.326	1.24	1.24	4.8
3D Heisenberg model ^b	0.36	1.39	1.39	
SrRuO ₃ (this work)	0.34 ± 0.02	1.14 ± 0.07	1.17 ± 0.14	~ 4

^aReference 13.^bReference 14.

$d\rho_m/dT$ (see inset to Fig. 1), which was discussed by Fisher and Langer¹⁸ (and was found to be anomalous in SrRuO₃, Ref. 2). We can estimate the contribution of this effect to the resistivity by integration of the diverging part of $d\rho/dT$. This contribution is found to be small relative to the resistivity changes considered in our analysis, and it falls rapidly when departing from T_c . Since we did not include in our analyses temperatures which are *very* close to T_c , the omission of this contribution is justified.

In conclusion, the consistent picture of the ferromagnetic phase transition not only supports the obtained values of the critical exponents, but it also reinforces the simple relation between the magnetic resistivity and the magnetization: $\rho_m(M) = \rho_m(0) - aM^2$, which holds for a surprisingly wide range of M .

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APPENDIX: DETERMINATION OF T_c AND β

The critical temperature T_c can be found from the location of the peak of $d\rho/dT$ (denoted hereafter by T_c'), and from the location of the peak of the induced magnetization at a constant magnetic field (hereafter denoted T_c''). Ideally, $T_c = T_c' = T_c''$; however, there is always some smearing of the transition (due to, e.g., defects or small temperature gradients), which may slightly shift T_c' and T_c'' relative to T_c .

We find that $T_c' = 152$ K. However, since $d\rho/dT$ decreases above T_c much faster than it grows below T_c (see

inset to Fig. 1), it is clear that rounding of the ideal $d\rho/dT$ vs T (caused by the smearing) would result in a negative shift of T_c' , namely, $T_c' < T_c$.

We estimate T_c'' to be the location of the maximum of the (negative) magnetoresistance at a constant field. From measurements at different fields we find that $T_c'' = 153.5 \pm 0.7$ K. However, due to the smearing, we expect to obtain $T_c'' > T_c$. This is because the critical susceptibility above T_c has a larger amplitude than it has below T_c (see values of C^+/C^- in Table I). Based on the results presented in the previous two paragraphs we chose $T_c = 153$ K to be the preliminary value for T_c .

In the following we give details on a more accurate determination of T_c and the determination of the critical exponent β , based on our interpretation of $\Delta\rho_{sp}$, as it is described in the text. We plotted $\Delta\rho_{sp}^{1/(2\beta)}$ as a function of T for trial values of β until we obtained the most straight line. Our basic choice is to make the extrapolation of $\rho^+(T)$ upon the range of 160–170 K (not too far from the investigated area and not too close to T_c) and to make the fit of $\Delta\rho_{sp}$ between 140–149 K (not too far from T_c , so that only the leading asymptotic terms are significant, and not too close to T_c , so that the results are not affected by the smearing and by the short-range spin correlations). This results in $T_c = 153.2$ K and $\beta = 0.347$.

To check how a change of the fit range may affect the results we did the same calculation for the ranges 135–145 K and 145–151 K and obtained the results $T_c = 152.5$ K, $\beta = 0.325$, and $T_c = 153.4$ K, $\beta = 0.353$ for those ranges, respectively. Another check was to change the range upon which the extrapolation of $\rho^+(T)$ is done. For a range of 160–180 K we obtained $T_c = 153.4$ K, $\beta = 0.348$. Therefore we conclude that reliable values are $T_c = 153.0 \pm 0.5$ K and $\beta = 0.34 \pm 0.02$.

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⁸From Ref. 1, the Fermi velocity in SrRuO₃ is approximately 2×10^7 cm/s and the mean free path is of order of 10 Å, thus the

quasiparticle scattering time is $\tau \sim 5 \times 10^{-15}$ s. Calculating the cyclotron frequency ω_c [taking the cyclotron mass to be 0.2 electron masses, as found from quantum oscillations measurements (Ref. 19) and from investigation of the extraordinary Hall effect (Ref. 20)], we obtain for our highest field the value of $(\omega_c \tau)^2$ to be 10^{-5} in order of magnitude, which is 3-4 orders of magnitude smaller than the measured relative magnetoresistances $\Delta\rho/\rho$.

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relative to the plane of the film. The in-plane component of the magnetization is perpendicular to the long dimension of the sample. Since the width-to-thickness ratio is large (~ 500), the demagnetization field due to the in-plane component is negligibly small, and the demagnetizing factor for the normal component is 4π . Therefore the demagnetizing field is $4\pi M \sin\theta$. It has a component in the direction of the easy axis and another component perpendicular to it. Magnetic field which is applied perpendicularly to the easy axis, has a very small effect on the magnetization because of the large magnetocrystalline anisotropy of SrRuO₃. Therefore we take only the component parallel to the easy axis as the effective demagnetizing field. Hence the correction should be $H = H_{\text{appl}} - 4\pi M \sin^2\theta$, where H_{appl} is the applied field.

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