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Pressure dependence of superconducting critical temperature of Sr_2RuO_4

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We studied electrical resistivity of single crystals of an oxide superconductor Sr_2RuO_4 under hydrostatic pressure up to 12 kbar. The midpoint T_c decreases at the rate of 3%/kbar. Anomalous increase of resistivity along the c axis is observed at room temperature with increasing pressure, whereas that in the ab plane decreased with pressure as normally expected. [S0163-1829(97)07538-3]

The oxide superconductor Sr_2RuO_4 draws our attention with its structural similarity to $La_{2-x}Sr_xCuO_4$ and yet low T_c of ~ 1 K.[1] The anisotropy in resistivity ($\rho_c/\rho_{ab} \sim 900$ at 2 K) is even larger than that of cuprates. It is hoped that it can play a role of a low-T_c counterpart of high-T_c cuprates in studying how cuprates differ from other superconductors.

Shortly after the discovery of superconductivity in Sr_2RuO_4, Rice and Sigrist argued the possibility of triplet-pairing, p-wave superconductivity in this system.[2] The results of specific heat[3] and nuclear quadrupole resonance[4] show that there is a significant portion of conduction electrons which seem to stay in the normal state down to zero kelvin.

In UPt_3, another and older candidate of a p-wave superconductor, a decrease of T_c with increasing pressure was reported by Willis *et al.*[5] They also observed a large decrease of T^2 term (''A'') in resistivity on pressurization, which they ascribed to the suppression of spin fluctuations by pressure.

But if the conduction-electron system can be regarded as a Fermi liquid, which is believed applicable to Sr_2RuO_4,[6,7] and the resistivity at low temperature is governed by the electron-electron interaction, A is proportional to γ^2, where γ denotes the electronic specific heat coefficient. In this case, the decrease of A under pressure is interpreted as the decrease of density of states with pressure. Comparison of the pressure effect on superconductivity in Sr_2RuO_4 with that in UPt_3 is useful in examining their similarity.

There is another topic that this article deals with. In high-T_c cuprates the temperature dependence of the out-of-plane resistivity ρ_c is semiconducting in the so-called ''underdoped'' region. As more holes are doped, ρ_c becomes metallic in the optimally and overdoped region. Maeno *et al.*[1] observed that ρ_c of Sr_2RuO_4 undergoes a crossover from a low-temperature metallic to a high-temperature nonmetallic state. They discussed that a crossover from metallic conduction to the thermally assisted hopping regime occurs when the scattering rate by phonons exceeds $1/\tau_c$, where τ_c stands for the time for an electron to travel between adjacent layers.[8] The pressure effect on ρ_c may shed some light on this interesting behavior.

In this study we have performed measurements of the in-plane and out-of-plane resistivity under hydrostatic pressure up to 12 kbar. The samples used in the measurements were platelike single-crystalline Sr_2RuO_4 grown by the floating-zone method.[9] The typical dimensions were $1.5\times0.6\times0.04$ mm^3. The resistivity was measured by a dc four-probe method. In out-of-plane (c-axis) measurements six gold wires were attached to the sample with heat-cure-type silver paste as shown in the inset of Fig. 1(b) and the terminals numbered 3 and 4 were shorted and used as a $I+$ lead, the terminals 5 and 6 $I-$, and the terminals 1 and 2 voltage leads.

We applied hydrostatic pressure to the samples as follows. The samples were placed in a Teflon cell filled with pressure medium (Idemitsu Daphne No. 7373 oil), which was placed in the Be-Cu clamp cell. Since we clamp the pressure cell at room temperature, the pressure decreases on cooling to low temperature. Previous calibration runs[10] showed that the pressure decrease at the lowest temperatures is about 1.5 kbar irrespective of the starting pressure. The cell was installed either in our ^{3}He cryostat or dilution refrigerator for low-temperature measurements.

The out-of-plane resistivity was measured after the in-plane measurement. A similar pressure dependence of T_c was followed by the out-of-plane measurement to that obtained beforehand in the in-plane experiment, ensuring the reproducibility of the T_c change.

We first present the resistively observed superconducting transition under pressure (Fig. 1). The ρ_{ab} result at 0, 8, and 12 kbar is shown, while the ρ_c result at 0, 4, and 8 kbar. Although the same sample was used throughout, we had to remake electrical contacts on the sample a few times, resulting in different residual resistivity above T_c from one pressure to another. Therefore in these figures we multiplied some data by a scale factor to give a similar residual resistivity. T_c decreases with increasing pressure at least up to 12 kbar.

Second, we show the plot of ρ vs T^2 under several pressure values (Fig. 2). In Fig. 2 no normalization regarding

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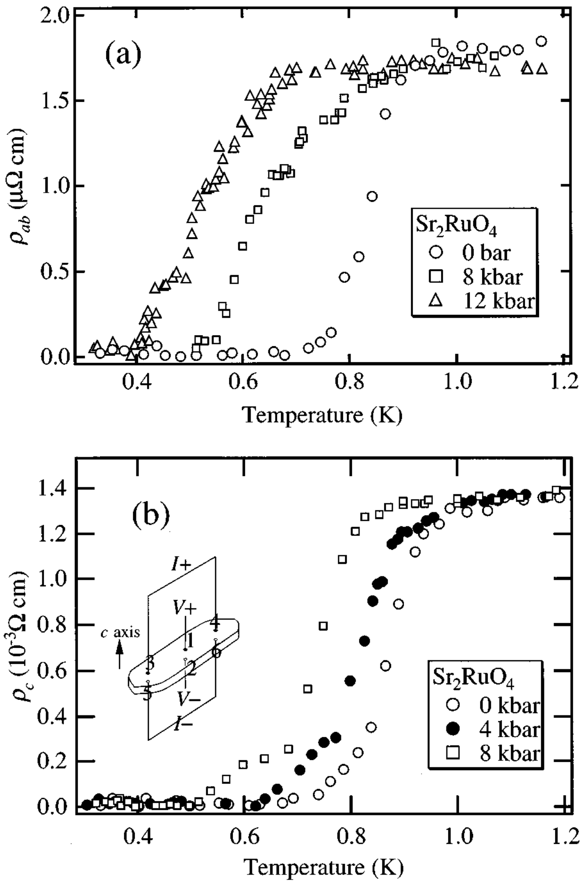

FIG. 1. The temperature dependence of resistivity around T_c under several pressures. (a) and (b) correspond to the in-plane and out-of-plane results, respectively. Note that the applied pressure values are not common to (a) and (b). Inset of (b): a sample with six terminals in c-axis measurements.

residual resistivity is made. Both the residual resistivity and the coefficient of T^2 term (A) depend little on pressure.

During the pressurization at room temperature we monitored the variation of resistivity. The results are shown in Fig. 3, which indicates a monotonic decrease of ρ_{ab} with the increase of pressure, as normally expected. ρ_c, however, increases with increasing pressure. Actual resistivities at room temperature are 160 $\mu\Omega$ cm and 16 mΩ cm for ρ_{ab} and ρ_c, respectively.

From the data depicted in Fig. 1 we obtain a T-P phase diagram (Fig. 4). The transition becomes a little broader with increasing pressure, probably because of the pressure distribution over the sample. By taking the mean of the superconductivity onset temperature and zero-resistance temperature, we deduce that the midpoint T_c decreases at the rate of about 3%/kbar and that superconductivity will be completely suppressed for $P \gtrsim 30$ kbar.

Since some suggest a pairing mechanism other than the usual electron-phonon interaction in this compound, it is worthwhile to see how well (or badly) a BCS-based theory applies. Here we use McMillan's formula for T_c:[11]

$$T_c = \frac{\Theta_D}{1.45} \exp\left[\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right]. \qquad (1)$$

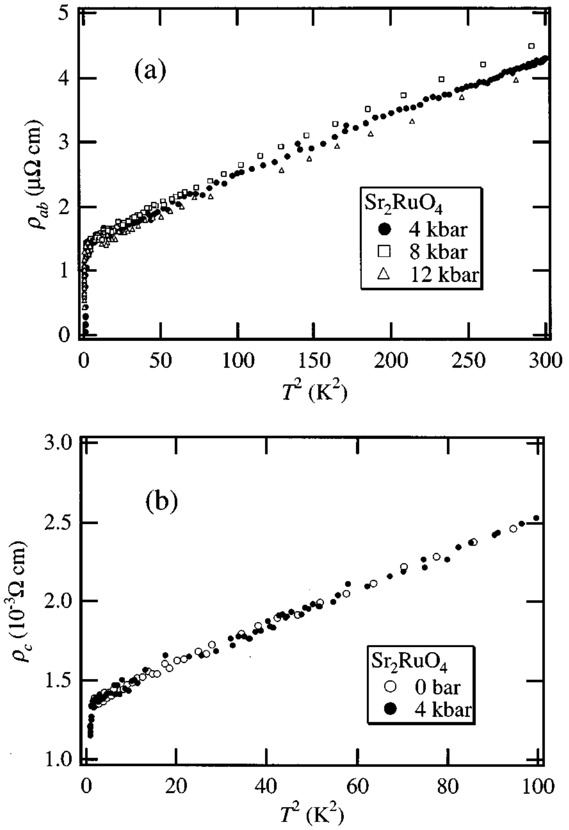

FIG. 2. Resistivity up to 17 or 10 K under various pressures plotted against T^2. (a) is for ab-plane and (b) for c-axis measurements. No normalization regarding the residual resistivity is made.

Θ_D has been determined experimentally as 410±50 K from a specific heat measurement on a single crystal.[12] For many superconducting transition metal elements, μ^* takes a common value of 0.13, as determined from the isotope effect.[13] We take, for instance, 0.13 as the μ^* value here. In ordinary cases λ is estimated from the enhancement of the experimental electronic specific heat coefficient (γ_{expt}) over the band-calculation value (γ_{band}).[14] According to Oguchi's band calculation,[15] λ $(=\gamma_{\mathrm{expt}}/\gamma_{\mathrm{band}}-1)$ is 2.8. If we substitute these quantities into Eq. (1), we obtain $T_c = 56$ K. Surely this is much too high compared with the actual T_c of ~1 K. The reason(s) for this large discrepancy may be the following.

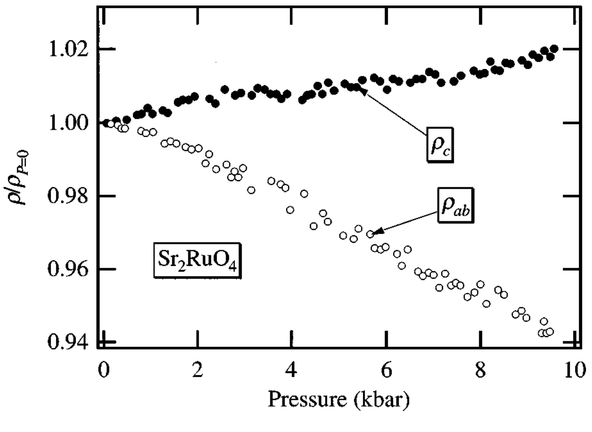

FIG. 3. Room-temperature resistivity as a function of pressure. The resistivity data are scaled by a zero-pressure value.

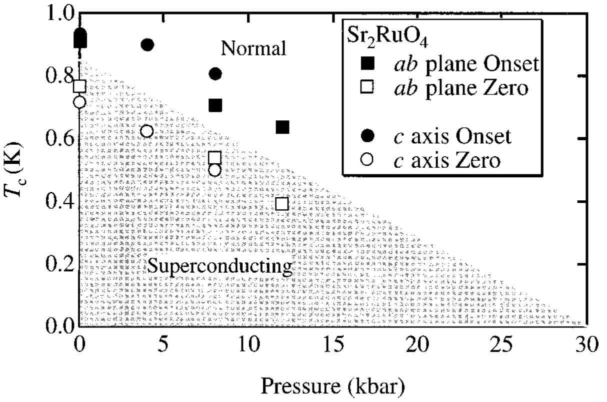

FIG. 4. *T-P* phase diagram at low temperature showing the extent of the superconducting state.

(i) The enhancement of the specific heat is mostly due to another mechanism than the electron-phonon interaction, thereby leaving λ entering in Eq. (1) much smaller than 2.8. For example, spin fluctuations also enhance γ_{expt}.

(ii) T_c is greatly suppressed by spin fluctuations.

(iii) μ^* is anomalously large.

(iv) Superconductivity in this system is all irrelevant to the phonon mechanism.

We point out here that (i) and (ii) are not independent of each other.

Along scenario (i) we can obtain an effective λ relevant to superconductivity ($\lambda_{\text{el-ph}}$) using Eq. (1) and $T_c \sim 1$ K. This $\lambda_{\text{el-ph}}$ is 0.43, which is only 15% of $\lambda_{\text{total}} = 2.8$. We will use these numbers later. Incidentally, aluminum with $T_c = 1.17$ K has nearly the same Debye temperature $\Theta_D = 428$ K as Sr_2RuO_4 and is characterized by $\lambda_{\text{el-ph}} = 0.38$.

Now we compare the pressure dependence of T_c and A in Sr_2RuO_4 with those in UPt_3. Willis *et al.*[5] observed that T_c decrease of UPt_3 is 2.6%/kbar, very close to the decrease rate 3% for Sr_2RuO_4. Note that T_c (~ 0.5 K) of UPt_3 itself is comparable to $T_c \sim 1$ K of Sr_2RuO_4. On the contrary the pressure dependence of A is quite different for the two systems. In UPt_3, A decreases by 30% at 8 kbar. In Sr_2RuO_4 the normal-state resistivity changes so little by pressure that it is difficult to deduce the exact number for the A change. From Fig. 2(a) we can say that the decrease of A from $P = 4$ to 12 kbar, which is expected to be virtually the same as the one from $P = 0$ to 8 kbar, is less than a few percent.

There are two possibilities for the origin of the T^2 dependence of resistivity at low temperature: electron-electron correlation and spin fluctuations. As to these ''exotic'' superconductors there are also two possibilities discussed for the origin of the attractive interaction between carriers: conventional electron-phonon interaction and spin fluctuations. If spin fluctuations dominate both low-temperature resistivity and superconductivity in Sr_2RuO_4, as believed is the case for UPt_3, the observed insensitivity of A in Sr_2RuO_4 is difficult to explain. Therefore, even if an attractive potential is mediated by spin fluctuations in Sr_2RuO_4, the origin of the T^2 dependence is likely to be electron correlation.

If we extend our discussion along scenario (i) and neglect the change of Θ_D by pressure, $\lambda_{\text{el-ph}}$ decreases to 0.41 at 8 kbar from 0.43 at 0 bar. This size of decrease is barely observable compared with λ_{total}. Therefore the insensitivity of A comes naturally if a conventional electron-phonon mechanism applies. In this case we cannot say anything about the origin of the T^2 dependence.

In both cases we may mention that the enhancement factor in Sr_2RuO_4 due to spin fluctuations is much smaller than in UPt_3. In the former case the spin-fluctuation-driven T^2 law is endangered as mentioned above. In the latter case, even if the T^2 dependence is brought about by spin fluctuations, the extent of the T^2 law goes up to much higher temperature in Sr_2RuO_4 than in UPt_3, which indicates a higher scaling temperature T_S in Sr_2RuO_4. T_S is inversely correlated with the enhancement factor due to spin fluctuations. Hence the higher T_S means the smaller enhancement factor.[16]

Therefore superconductivity in Sr_2RuO_4 does not seem to fall into the same category as of UPt_3, and even if spin fluctuations are mediators of attractive force, they should be quite different from those in UPt_3, e.g., having a different shape of $\chi(\mathbf{q},\omega)$.

The pressure dependence of ρ_c (Fig. 3) is very interesting because the resistivity usually decreases under pressure regardless of whether it is a coherent metal or is in a tunneling regime. If the system is a band insulator, the band gap may increase in the modification of the band structure due to pressure. Our ρ_c measurement was done at room temperature and Yoshida *et al.*[8] have suggested that the system crosses over to a thermally assisted hopping regime above 130 K. Surely this broad hump at around 130 K does not look like a metal-to-insulator phase transition and the temperature dependence of ρ_c above 130 K is not an activation type. Thus it is difficult to regard it as a band insulator. The increase of ρ_c has to be understood taking account of the shift of the temperature where ρ_c takes its maximum. We have not yet studied this shift thoroughly.

Since our pressure is hydrostatic, we are not sure that the *c* axis was really shortened on pressurization. Uniaxial stress along the *c* axis is particularly useful to decide whether this increase of ρ_c really occurs with the *c*-axis shortening.

In summary we have observed a fairly rapid T_c decrease by applying pressure on Sr_2RuO_4. The midpoint T_c decreases at ~ 3%/kbar, while A changed little in sharp contrast to UPt_3, for which A decreases drastically. Although relatively stable A is consistent with conventional superconductivity, a different type of spin fluctuation may be in effect in Sr_2RuO_4 from those in UPt_3. Another interesting observation is that ρ_c at room temperature increases under pressure. This behavior has to be considered in the context of a metal-nonmetal crossover of ρ_c.

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