PHYSICAL REVIEW B **81**, 180510(R) (2010)

Higher-T_c superconducting phase in Sr_2RuO_4 induced by uniaxial pressure

Shunichiro Kittaka,[1,*] Haruka Taniguchi,[1] Shingo Yonezawa,[1] Hiroshi Yaguchi,[1,2] and Yoshiteru Maeno[1]
[1]*Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*
[2]*Department of Physics, Faculty of Science and Technology, Tokyo University of Science, Chiba 278-8510, Japan*
(Received 16 April 2010; published 18 May 2010)

We have investigated uniaxial pressure effect on superconductivity of *pure* Sr_2RuO_4, whose intrinsic superconducting transition temperature T_c is 1.5 K. It is revealed that a very low uniaxial pressure along the c axis, only 0.2 GPa, induces superconductivity with the onset T_c above 3 K. The present results indicate that pure Sr_2RuO_4 has two superconducting phases with $T_c=1.5$ K and with varying T_c up to 3.2 K. The latter phase exhibits unusual features and is attributable to anisotropic crystal distortions beyond the elastic limit.

DOI: 10.1103/PhysRevB.81.180510 PACS number(s): 74.70.Pq, 74.62.Fj

The application of uniaxial pressure (UP) can be a powerful tool to control superconductivity (SC) as well as the electronic structure through anisotropic distortions in the crystal lattice.[1–4] The layered perovskite ruthenate Sr_2RuO_4, for which convincing evidence has been accumulated in favor of spin-triplet SC,[5,6] has an undistorted tetragonal structure. It has been experimentally and theoretically revealed that the electronic states of Sr_2RuO_4 and its related materials are expected to drastically change with an anisotropic distortion.[7–13] Thus, Sr_2RuO_4 is one of the materials expected to have a prominent UP effect on its SC.

The intrinsic superconducting transition temperature T_c of Sr_2RuO_4 was revealed to be 1.5 K for crystals with the best quality.[6] Hydrostatic pressure,[14,15] as well as a small amount of impurities or defects,[16,17] is known to suppress T_c. In contrast, the enhancement of T_c was reported in the Sr_2RuO_4-Ru eutectic system (the onset $T_c \sim 3-3.5$ K) (Refs. 18–20) and a submicron Sr_2RuO_4 single crystal (the onset $T_c \sim 1.8$ K).[21] At present, the mechanisms of the enhancement of T_c remain unresolved. In the eutectic system, lamellae of Ru metal ($T_c=0.49$ K) with approximate dimensions of $10\times10\times1$ μm^3 are embedded with a stripe pattern. It has been established that the 3-K SC with a tiny volume fraction occurs in the Sr_2RuO_4 region in the vicinity of the Sr_2RuO_4-Ru interface.[22,23] One possible scenario is that anisotropic distortions in Sr_2RuO_4, e.g., induced by the presence of Ru, enhance its T_c significantly.[19]

Although UP experiments on Sr_2RuO_4 have not been reported, the change in T_c with UP along the interlayer c and in-plane a axis, $P_{\|c}$ and $P_{\|a}$, in the elastic limit was predicted from the ultrasonic experiments combined with the Ehrenfest relations:[24] $(1/T_c)(dT_c/dP_{\|c})=(0.7\pm0.2)$ GPa^{-1} and $(1/T_c)(dT_c/dP_{\|a})=-(0.85\pm0.05)$ GPa^{-1}. Qualitatively the same UP effects were theoretically predicted based on the change in the density of states at the Fermi level in the band mainly responsible for the SC.[25] On the basis of these predictions, T_c is expected to increase under $P_{\|c}$.

Recently, the UP effects on interfacial 3-K SC in the eutectic system have been investigated.[26,27] It was revealed that the volume fraction of the 3-K SC drastically increases by the application of UP in *any* directions while its onset T_c is almost invariant. These findings urge the UP effect on *pure* Sr_2RuO_4 to be investigated as well.

In this Rapid Communication, we report the effect of $P_{\|c}$ on pure Sr_2RuO_4. We reveal that the onset T_c of pure Sr_2RuO_4 is immediately enhanced to 3.2 K by $P_{\|c}$ of only 0.2 GPa. By comparing the effects of $P_{\|c}$ between pure Sr_2RuO_4 and Sr_2RuO_4-Ru eutectic crystals, it is revealed that the higher-T_c SC in the pure Sr_2RuO_4 sample is not induced by the development of interfacial 3-K SC originating from a tiny amount of Ru inclusions but is indeed an intrinsic property of pure Sr_2RuO_4.

Single crystals used in this study were grown by a floating zone method with Ru self-flux.[28] Because RuO_2 evaporates from the surface of melt during the growth, excess Ru tends to be left in the center of the melt. Therefore, most of single crystalline rods with the best T_c (no defect at the Ru site) contain Sr_2RuO_4-Ru eutectic solidification in its core region and pure Sr_2RuO_4 only around the thin surface part.[19] This feature, as well as the tendency to cleave easily, makes it difficult to obtain a large crystal which only contains Sr_2RuO_4. In particular, it is extremely difficult to prepare a pure sample suitable for the application of $P_{\|a}$. For the application of $P_{\|c}$, we have succeeded in preparing one large sample of practically pure Sr_2RuO_4 with almost no Ru inclusions (sample 1) from a crystalline rod with relatively lower T_c (small amount of defects at the Ru site). The dimensions of sample 1 are 1.5×1.4 mm^2 in the ab plane and 0.22 mm along the c axis. In order to reduce the amount of lattice defects and oxygen deficiencies as well as the number of Ru inclusions, sample 1 was annealed in oxygen at 1 atm and 1050 °C for a week. Then, sample 1 exhibits a very sharp transition at 1.34 K, as determined from the ac susceptibility.

In addition to sample 1, two Sr_2RuO_4 samples containing a small amount of Ru inclusions and five Sr_2RuO_4-Ru eutectic samples have been used to investigate the $P_{\|c}$ effect. Representing those samples, magnetic susceptibility results for a Sr_2RuO_4 sample with a small amount of Ru inclusions (sample 2) and a Sr_2RuO_4-Ru eutectic sample (sample 3) are compared with the results for sample 1 in this Rapid Communication. The approximate dimensions of samples 2 and 3 were 1.5×1.5 mm^2 in the ab plane and 0.3 mm along the c axis. The amount of Ru inclusions was identified from the polarized-light optical microscope images of the ab surfaces, as exemplified in Figs. 1(a)–1(c). The number density of Ru inclusions on the ab surfaces for sample 1 is less than 3 mm^{-2} and about 40 mm^{-2} after and before annealing, respectively while the number densities are about 200 mm^{-2} for sample 2 and about 4000 mm^{-2} for sample 3.

UP was applied parallel to the shortest dimension along

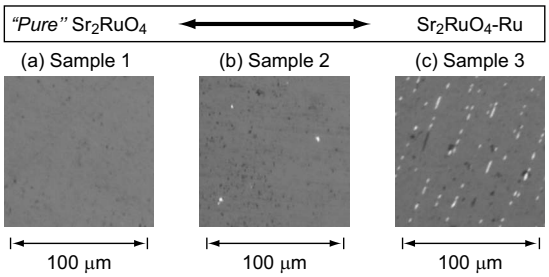

FIG. 1. Polarized-light optical microscope images of the *ab* planes of samples (a) 1, (b) 2, and (c) 3. The dark and bright parts correspond to Sr_2RuO_4 and Ru, respectively.

the *c* axis using a piston-cylinder-type pressure cell made of Cu-Be alloy with a cylindrical outer body made of hard plastic (polybenzimidazole). The room-temperature pressure value was confirmed to be in a reasonable agreement with low-temperature pressure value determined by superconducting transitions of tin and lead.[29] The side surfaces of the samples were covered with thin epoxy (Emerson-Cuming, Stycast 1266) to prevent a breakdown of the sample, as described in Ref. 26. The magnetization was measured down to 1.8 K with an applied dc magnetic field $\mu_0 H_{dc}$ of 2 mT using a superconducting quantum interference device magnetometer (Quantum Design, model MPMS). The background magnetization of the pressure cell was subtracted. The ac susceptibility $\chi_{ac}=\chi'-i\chi''$ was measured down to 0.3 K by a mutual-inductance technique using a lock-in amplifier (LIA) with a ^{3}He cryostat (Oxford Instruments, model Heliox VL). All χ_{ac} data presented here were taken at 293 Hz. The values of χ_{ac} were obtained from the relation $\chi_{ac}=iC_1V_{LIA}/H_{ac}+C_2$, where $V_{LIA}(=V_x+iV_y)$ is the read-out voltage of LIA and H_{ac} is the magnitude of the applied ac magnetic field. The values of C_1 and C_2 were chosen so that $\chi'(4\ \mathrm{K})=0$ and $\chi'(0.3\ \mathrm{K})=-1$ at 0 GPa for each sample; thus $|\chi'|$ corresponds to the ac shielding fraction. For the χ_{ac} curves under $P_{\|c}$, C_1 and C_2 determined at 0 GPa were used.

Figure 2(a) represents the temperature dependence of the superconducting dc susceptibility $\Delta\chi_{dc}=\Delta m/\mu_0 H_{dc}$ for each sample at 0 GPa, where Δm is the observed magnetization change associated with the superconducting transition divided by the sample volume. Here, the ideal value for the full Meissner state without the demagnetization correction corresponds to $\Delta\chi_{dc}=-1$; thus $|\Delta\chi_{dc}|$ is equal to the dc shielding fraction. At 0 GPa, there is no sign of a dc shielding signal for sample 1 above 1.8 K, confirming that it does not contain any eutectic part. A weak shielding signal with the onset T_c slightly above 3 K was observed in samples 2 and 3, consistent with the recent study.[20] At 0 GPa, the dc shielding fraction at 1.8 K is clearly larger in sample 3, containing more Ru inclusions.

Surprisingly, even at relatively low $P_{\|c}$ of 0.2 GPa, SC with the onset T_c of 3.2 K, at which $\Delta\chi_{dc}$ deviates from zero [see Fig. 2(c)], is induced in sample 1. As shown in Fig. 2(b), the shielding fraction in the field-cooling (FC) process is nearly half of that in the zero-field-cooling (ZFC) process at $P_{\|c}=0.3$ GPa. The relatively large FC shielding fraction indicates that the screening area does not contain normal-state regions largely. Figure 2(d) demonstrates the variation in the

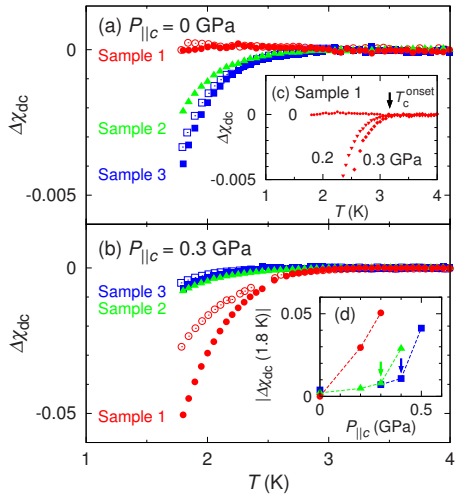

FIG. 2. (Color online) Temperature dependence of the dc susceptibility $\Delta\chi_{dc}$ of samples 1 (circles), 2 (triangles), and 3 (squares) measured with 2 mT at (a) $P_{\|c}=0$ and (b) 0.3 GPa. The applied dc field has been corrected for the remanent field in the sample space. Open and closed symbols indicate data taken in the FC and ZFC processes, respectively (samples 1 and 3 only). Note the vertical scale changes between (a) and (b). (c) Enlarged view near the onset for sample 1 at different $P_{\|c}$. (d) Dependence of the dc shielding fraction on $P_{\|c}$ at 1.8 K. The arrows indicate critical pressure $P^*_{\|c}$.

dc shielding fraction at 1.8 K, $|\Delta\chi_{dc}(1.8\ \mathrm{K})|$, under $P_{\|c}$ for each sample. Unexpectedly, as the amount of Ru inclusions is *larger*, the enhancement of the shielding fraction by $P_{\|c}$ becomes *smaller*; the order of increasing shielding fractions among different samples is reversed between Figs. 2(a) and 2(b). Below 0.3 GPa, the slope of $|\Delta\chi_{dc}(1.8\ \mathrm{K})|$ versus $P_{\|c}$ for sample 1 is much greater than those for samples 2 and 3. These results strongly indicate that the presence of Ru inclusions does not play a positive role in the rapid enhancement of the shielding fraction under $P_{\|c}$. Moreover, it should be noted that the slope for samples 2 and 3 abruptly changes at a critical pressure, which we designate as $P^*_{\|c}$, of around 0.4 GPa. Interestingly, the steep slope above $P^*_{\|c}$ for samples 2 and 3 is almost the same as the slope for sample 1.

We have also measured the temperature dependence of χ_{ac} using the identical samples under $P_{\|c}$ with $\mu_0 H_{ac}$ of 2 μT-rms. These χ_{ac} measurements cover low temperatures down to 0.3 K and enable us to observe the full Meissner state. As presented in Fig. 3(a), sample 1 exhibits a very sharp transition and no dissipation χ'' above 1.5 K at 0 GPa. This sharp transition again supports that sample 1 can be treated as a pure Sr_2RuO_4. By contrast, as shown in Figs. 3(b) and 3(c), broad transitions, typical of the eutectic system,[20] were observed above 1.5 K for samples 2 and 3 at 0 GPa.

By the application of $P_{\|c}$, a broad signal of ac shielding with the onset T_c of up to 3.2 K grows in all samples; the enhancement of the ac shielding fraction was clearly observed above 1.5 K even for pure Sr_2RuO_4. These results are both qualitatively and quantitatively consistent with the results of the dc magnetization measurements in Fig. 2. By contrast, at low temperatures, the ac shielding signal is reduced in magnitude and becomes broader with increasing

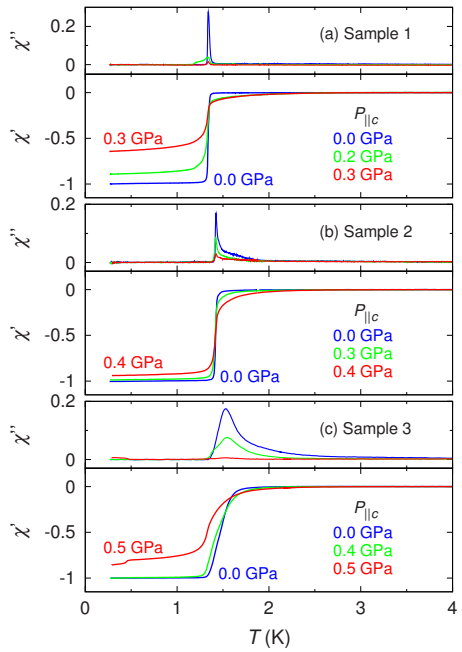

FIG. 3. (Color online) Temperature dependence of the real and imaginary parts of χ_{ac} under different $P_{\|c}$ for (a) sample 1, (b) sample 2, and (c) sample 3 measured with $\mu_0 H_{ac}$=2 μT-rms and 293 Hz.

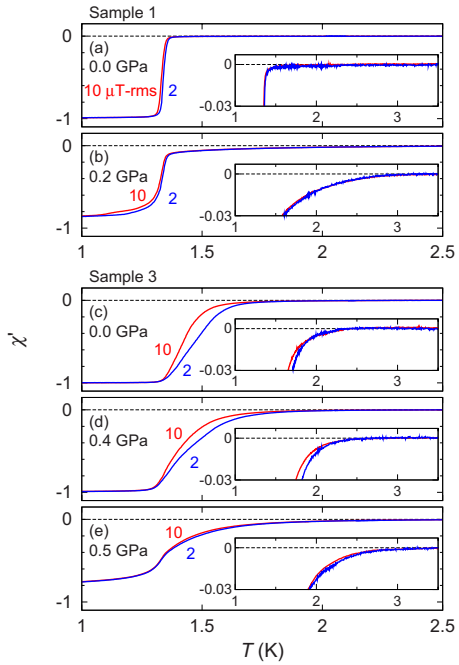

FIG. 4. (Color online) Temperature dependence of χ' for $\mu_0 H_{ac}$=2 and 10 μT-rms at (a) $P_{\|c}$=0 and (b) 0.2 GPa for sample 1 and at (c) $P_{\|c}$=0, (d) 0.4, and (e) 0.5 GPa for sample 3. The insets are enlarged views near the onset. The dashed lines mark χ'=0.

$P_{\|c}$. It should also be noted that the primary 1.5-K SC part in samples 1 and 2 is sustained with little change of its T_c under $P_{\|c}$. These features imply that a region with widely distributed T_c from zero up to above 3 K emerges, replacing some parts of the 1.5-K SC (i.e., two superconducting phases coexist). Although the shielding signal near the onset T_c becomes somewhat smaller after removing uniaxial pressure [e.g., Fig. 4(a) of Ref. 26], the recovery was not complete. Such irreversibility suggests that the enhancement of T_c is accompanied by distortions exceeding the elastic limit.

As evident in Fig. 3(c), the reduction in the shielding fraction and the broadening of the transition below 1.5 K become particularly significant for $P_{\|c} \geq P^*_{\|c}$ [Fig. 2(d)]. Above $P^*_{\|c}$, the interfacial 3-K SC around Ru inclusions also seems suppressed because the screening signal of the superconducting transition of Ru (T_c=0.49 K) becomes substantially strong. Such screening signal of Ru SC was reproducibly observed in all the eutectic samples at higher $P_{\|c}$.

Another important feature of the UP-induced higher-T_c SC is that the screening is insensitive to H_{ac}. As shown in Figs. 4(a) and 4(b), the ac shielding fraction in pure Sr_2RuO_4 was not affected by H_{ac} at any $P_{\|c}$ investigated. In contrast, the ac shielding fraction in sample 3 was sensitively suppressed by H_{ac} of as small as 10 μT-rms below $P^*_{\|c}$ $\sim$0.4 GPa [Figs. 4(c) and 4(d)]. This "weak" SC in the eutectic system is attributed to the formation of a Josephson network;[20] interfacial 3-K SC occurring near the Sr_2RuO_4-Ru interface penetrates into the normal-state Sr_2RuO_4 region due to the proximity effect and forms Josephson-type weak links among the Ru lamellae. In this case, because of the small critical current in the Josephson network area, the ac field penetration is expected to become significantly deeper with increasing H_{ac}. Therefore the spatial distribution of superconducting regions is different between the two systems.

Interestingly, as shown in Fig. 4(e), the H_{ac} dependence almost disappears in sample 3 above $P^*_{\|c}$. This disappearance, in conjunction with the observation in Figs. 2(d) and 3(c), can be consistently understood if the nature of the higher-T_c SC induced by UP in pure Sr_2RuO_4 and that of SC in the eutectic crystal *above* $P^*_{\|c}$ is qualitatively the same.

From the present study, it is most likely that UP-induced higher-T_c SC is an intrinsic property of Sr_2RuO_4 because the enhancement of the higher-T_c SC by $P_{\|c}$ is more striking in samples with smaller amount of Ru inclusions. Let us here discuss possible mechanisms of the occurrence of the higher-T_c SC. While hydrostatic pressure destroys SC of Sr_2RuO_4,[14,15] UP induces SC with T_c above 3 K. These facts indicate that anisotropic distortions caused by UP in the crystal structure play essential roles in enhancing T_c. Nevertheless, the present results are essentially different from the UP effect predicted from the ultrasonic experiments, $dT_c/dP_{\|c}$ $\sim$1 K/GPa. The observed onset T_c increases up to 3.2 K even at $P_{\|c}$ of as small as 0.2 GPa but does not exceed 3.2 K at higher $P_{\|c}$ [see Fig. 2(c)].

This disagreement with the hydrostatic pressure effect and the elastic-limit expectation suggests that a qualitative change in the electronic structure of Sr_2RuO_4 arises at relatively low $P_{\|c}$ and generates SC with enhanced T_c. It is known that the electronic states of $Sr_{n+1}Ru_nO_{3n+1}$ series are significantly affected by the rotation, tilting, and flattening of the RuO_6 octahedra.[8–10] For example, $P_{\|c}$ of about 0.4 GPa changes the electronic state of the bilayer system $Sr_3Ru_2O_7$ from a metamagnetic metal to a ferromagnetic metal.[13] In the case of Sr_2RuO_4, its crystal structure is close to an instability

against the in-plane RuO_6 rotation, as evidenced by the softening of the rotational phonon mode observed in inelastic neutron-scattering experiments.[7] Theoretically, it was predicted that the RuO_6 rotation leads to the decrease in the band widths and to the increase in T_c through the enhancement of the density of states associated with the van Hove singularity of one of the bands.[8,30] Also, the tilting and flattening of the RuO_6 octahedra are expected to make the band widths narrower.[9]

The observed distribution of T_c in UP-induced higher-T_c SC may be explained by lattice imperfection. If T_c depends on the magnitude of local anisotropic distortion, T_c would be distributed because of experimentally unavoidable inhomogeneity of the distortion. In addition, T_c should be suppressed by the pair-breaking scatterings due to lattice imperfection, e.g., the dislocations or rapid spatial variation in the distortion, leading to the reduction in the mean-free path. Note that SC is completely destroyed in pure Sr_2RuO_4 if the mean-free path is less than about 50 nm.[16] Therefore, the higher T_c is expected only in the Sr_2RuO_4 region with a coherent distortion. This scenario is consistent with the recent report on the eutectic system[31] suggesting that the enhancement of T_c occurs in the Sr_2RuO_4 region at least 18 nm away from the interface rather than at the immediate vicinity of the interface with a lot of dislocations.

To summarize, we present a report on the UP effect on Sr_2RuO_4. We have found a remarkable increase in the onset T_c from 1.5 to 3.2 K induced by $P_{\|c}$ below 0.2 GPa. This strong enhancement of T_c cannot be explained by the effect of $P_{\|c}$ in the elastic limit deduced from the Ehrenfest relation for Sr_2RuO_4. We have revealed that the dc shielding fraction associated with the UP-induced higher-T_c SC and the amount of Ru inclusions are anticorrelated. This fact clearly indicates that the higher-T_c SC is not associated with the presence of Ru inclusions.

There remain some important issues to be clarified. Although the spatial distribution of superconducting regions is revealed to be different between the UP-induced higher-T_c SC and the eutectic 3-K SC, the values of their T_c's are surprisingly similar. It should be clarified whether or not the origin of the enhanced T_c is similar between UP-induced SC and the eutectic SC. Another important issue is to resolve the discrepancy between the results of previous hydrostatic-pressure study and of the present UP experiments. Additional experiments under in-plane UP and the crystal-structure analysis under UP may provide important clues although it is technically difficult at present to perform such experiments.

We thank F. Nakamura, N. Takeshita, T. Nakamura, H. Takatsu, M. Kriener, Y. Nakai, and K. Ishida for their support and M. Sigrist for useful discussions. This work is supported by a Grant-in-Aid for the Global COE program "The Next Generation of Physics, Spun from Universality and Emergence" from the MEXT of Japan and by a Grant-in-Aid for Scientific Research from the MEXT, and from the JSPS. S.K. is supported by the JSPS.

*Present address: Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan.

[1] O. M. Dix, A. G. Swartz, R. J. Zieve, J. Cooley, T. R. Sayles, and M. B. Maple, Phys. Rev. Lett. **102**, 197001 (2009).
[2] C. E. Campos *et al.*, Phys. Rev. B **52**, R7014 (1995).
[3] M. Maesato, Y. Kaga, R. Kondo, and S. Kagoshima, Phys. Rev. B **64**, 155104 (2001).
[4] D. S. Jin, S. A. Carter, B. Ellman, T. F. Rosenbaum, and D. G. Hinks, Phys. Rev. Lett. **68**, 1597 (1992).
[5] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednorz, and F. Lichtenberg, Nature (London) **372**, 532 (1994).
[6] A. P. Mackenzie and Y. Maeno, Rev. Mod. Phys. **75**, 657 (2003).
[7] M. Braden, W. Reichardt, S. Nishizaki, Y. Mori, and Y. Maeno, Phys. Rev. B **57**, 1236 (1998).
[8] R. Matzdorf, Z. Fang, Ismail, J. Zhang, T. Kimura, Y. Tokura, K. Terakura, and E. W. Plummer, Science **289**, 746 (2000).
[9] Z. Fang and K. Terakura, Phys. Rev. B **64**, 020509(R) (2001).
[10] O. Friedt, M. Braden, G. Andre, P. Adelmann, S. Nakatsuji, and Y. Maeno, Phys. Rev. B **63**, 174432 (2001).
[11] S. Nakatsuji and Y. Maeno, Phys. Rev. Lett. **84**, 2666 (2000).
[12] F. Nakamura, T. Goko, M. Ito, T. Fujita, S. Nakatsuji, H. Fukazawa, Y. Maeno, P. Alireza, D. Forsythe, and S. R. Julian, Phys. Rev. B **65**, 220402(R) (2002).
[13] S.-I. Ikeda, N. Shirakawa, T. Yanagisawa, Y. Yoshida, S. Koikegami, S. Koike, M. Kosaka, and Y. Uwatoko, J. Phys. Soc. Jpn. **73**, 1322 (2004).
[14] N. Shirakawa, K. Murata, S. Nishizaki, Y. Maeno, and T. Fujita, Phys. Rev. B **56**, 7890 (1997).
[15] D. Forsythe *et al.*, Phys. Rev. Lett. **89**, 166402 (2002).
[16] A. P. Mackenzie, R. K. W. Haselwimmer, A. W. Tyler, G. G. Lonzarich, Y. Mori, S. Nishizaki, and Y. Maeno, Phys. Rev. Lett. **80**, 161 (1998).
[17] Z. Q. Mao, Y. Mori, and Y. Maeno, Phys. Rev. B **60**, 610 (1999).
[18] Y. Maeno, T. Ando, Y. Mori, E. Ohmichi, S. Ikeda, S. NishiZaki, and S. Nakatsuji, Phys. Rev. Lett. **81**, 3765 (1998).
[19] T. Ando, T. Akima, Y. Mori, and Y. Maeno, J. Phys. Soc. Jpn. **68**, 1651 (1999).
[20] S. Kittaka, T. Nakamura, H. Yaguchi, S. Yonezawa, and Y. Maeno, J. Phys. Soc. Jpn. **78**, 064703 (2009).
[21] H. Nobukane *et al.*, Solid State Commun. **149**, 1212 (2009).
[22] H. Yaguchi, M. Wada, T. Akima, Y. Maeno, and T. Ishiguro, Phys. Rev. B **67**, 214519 (2003).
[23] M. Kawamura, H. Yaguchi, N. Kikugawa, Y. Maeno, and H. Takayanagi, J. Phys. Soc. Jpn. **74**, 531 (2005).
[24] N. Okuda, T. Suzuki, Z. Q. Mao, Y. Maeno, and T. Fujita, J. Phys. Soc. Jpn. **71**, 1134 (2002).
[25] T. Nomura and K. Yamada, J. Phys. Soc. Jpn. **71**, 1993 (2002).
[26] S. Kittaka, H. Yaguchi, and Y. Maeno, J. Phys. Soc. Jpn. **78**, 103705 (2009).
[27] H. Yaguchi, S. Kittaka, and Y. Maeno, J. Phys.: Conf. Ser. **150**, 052285 (2009).
[28] Z. Q. Mao, Y. Maeno, and H. Fukazawa, Mater. Res. Bull. **35**, 1813 (2000).
[29] T. F. Smith and C. W. Chu, Phys. Rev. **159**, 353 (1967).
[30] M. Sigrist and H. Monien, J. Phys. Soc. Jpn. **70**, 2409 (2001).
[31] Y. A. Ying *et al.*, Phys. Rev. Lett. **103**, 247004 (2009).