Influence of hydrostatic pressure on the magnetic phase diagram of superconducting Sr_2RuO_4 by ultrasonic attenuation

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(Received 15 December 2007; published 8 February 2008)

The effect of high hydrostatic pressure on the magnetic field and temperature dependence of the attenuation of a longitudinal ultrasonic wave propagated along the ⟨100⟩ direction in superconducting Sr_2RuO_4 has been investigated. The magnetic field dependence of the attenuation suggests the vertical character of the nodes in the superconducting gap. Application of pressure causes a decrease of the critical temperature T_c and critical magnetic field H_{c2}; the width of the transition under pressure increases. The dependence of $H_{c2\|ab}$ on T_c was found to be linear, in contrast to the quadratic one for $H_{c2\|c}$. The slope $dH_{c2\|ab}/dT_c$ has a universal character; its value does not depend on the physical factor causing reduction of T_c, regardless if it is pressure or impurities. The observed effects demonstrate that the pressure application induces an increase in the electron-electron interaction. We show that this increase can be described in terms of a changing electron relaxation time, as in the case of impurities.

DOI: 10.1103/PhysRevB.77.052502 PACS number(s): 74.70.Pq, 74.25.Ha, 74.25.Ld, 74.62.Fj

The persistent interest in layered Sr_2RuO_4 over the past dozen of years (for review see Refs. 1–3) is fueled by the unconventional spin-triplet superconductivity[4] and the rich variety of phenomena, related to this property. Some characteristics of this material, for example, two dimensionality of the electron liquid[5] and the Fermi surface topology,[6] have already been understood. At the same time, there is still no self-consistent superconductivity model for this material. In particular, the question of the gap structure is especially contradictory. Some authors believed that the best explanation of experimental results can be achieved with the assumption of vertical nodes.[7–11] Others insist that the assumption of horizontal nodes is more correct and promising.[1,12–19] There have also been suggestions of point nodes.[20] Observation of a second superconducting phase[21] makes this problem even more complicated. The lack of relevant experimental data, in particular, magnetoacoustic and high pressure, was discussed, for example, in Refs. 1, 22, and 23. Consequently, there is a clear motivation to perform more work on Sr_2RuO_4, diversifying the experimental techniques and conditions in an effort to obtain deeper insight into the nature of its superconductivity. We believe that our research is an important step in clarifying the gap node character. Among the other revealed phenomena, the linear dependence of the upper critical magnetic field, $H_{c2\|ab}$, on T_c is the most interesting one, especially that it is contrasting with the known[24] quadratic dependence for $H_{c2\|c}$.

The Sr_2RuO_4 single crystal was grown by the floating zone technique[25] in oxygen-reduced atmosphere at the rate of 30 mm/h. The as-grown crystal was about 30 mm in length and 4 mm in diameter. Upon 100 h of annealing at 1000 °C in O_2 atmosphere, the crystal was oriented and cut perpendicular to the ⟨100⟩ direction. The superconducting transition occurred at a temperature of T_c=1.2 K, which indicates the good quality of the material. A pair of longitudinal 60 MHz $LiNbO_3$ transducers attached to the two polished optical quality opposite (100) surfaces allowed us to perform the pulse-echo study of the c_{11} ultrasonic mode. The measurements were done in the frequency range of 60–300 MHz using the resonant frequency of the transducers and its odd harmonics. The details of the ultrasonic experimental apparatus were published earlier.[26]

The crystal was placed into a hydrostatic piston-cylinder pressure cell, whose design will be described elsewhere.[27] The value of the pressure inside the cell was monitored by the shift of the 694 nm luminescence line of a ruby crystal,[28] installed next to the sample. During the experiment, the pressure was changed in one direction only, from the low to high values. The cell was mounted in the rotator. Due to the rather large size of the cell and limited space inside the magnet, we could rotate the crystal with respect to the magnetic field for only approximately ±10°. This, however, enabled us to use the angular dependence of the ultrasound signal in Sr_2RuO_4 (Ref. 29) for precise alignment of the ⟨100⟩ direction of the crystal (and the ultrasound wave vector) along the field.

The temperature and magnetic field dependences of the ultrasound attenuation α in the vicinity of the superconducting transition, measured at several values of the hydrostatic pressure, are presented in Fig. 1. The specific construction of the pressure cell deteriorated the noise characteristics of the equipment. Consequently, reliable measurements of the weak changes[30,32] of the sound velocity at the transition could not be made.

Besides the reported earlier[33,34] pressure-induced T_c decrease, Fig. 1(a) reveals a drop in the magnitude of the normal state attenuation (when $T>T_c$, H=0). In the normal state, $\alpha \sim \omega^2 \tau$ (ω is ultrasound frequency, and τ is electron system relaxation time). We will show below that τ drops down under hydrostatic pressure; consequently, α decreases. The evolution of the attenuation when the temperature is fixed to T=0.4 K and the superconductivity is suppressed by an external magnetic field [Fig. 1(b)] shows that with pressure the critical field H_{c2} decreases, but the normal state attenuation (when T=0.4 K, $H>H_{c2}$) increases. The temperature dependence of α is determined by the ultrasound interaction with thermal phonons and with electrons. However, the magnetic field dependence of α is due to the later mechanism only. This explains the difference of the behavior

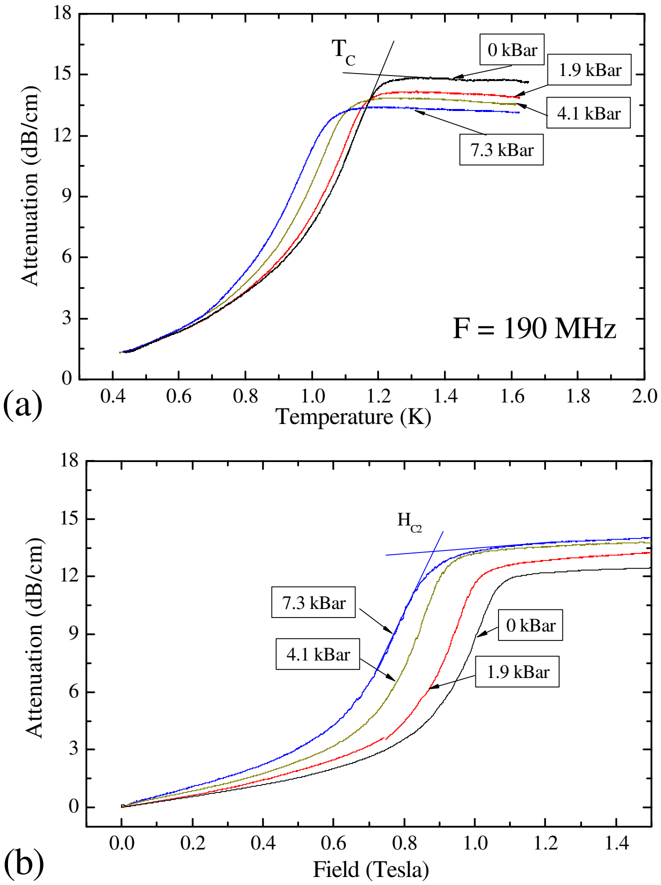

FIG. 1. (Color online) Ultrasonic attenuation in the vicinity of the superconducting transition as a function of temperature (a); magnetic field (b) at the fixed temperature $T=0.4$ K, measured at several values of pressure.

of α, as shown in Figs. 1(a) and 1(b). Quantitative analysis of the normal state behavior of α requires that additional experiments performed.

It is remarkable that in spite of the different T_c values and different growth techniques, the zero pressure normalized ultrasound attenuation in our crystal follows exactly the same temperature curve, as in Ref. 35, plotted versus reduced temperature. A good fit to this dependence was achieved with the assumption of vertical nodes (see Fig. 5 in Ref. 11 and Fig. 3 in Ref. 35) or nodal points (Fig. 3 in Ref. 20). To the best of our knowledge, these ultrasonic data have never been modeled successfully by any theory based on horizontal nodes.

Studying the magnetic field dependence of the ultrasonic attenuation in Sr_2RuO_4 in the neighborhood of the transition can be helpful in determining the gap structure. To the best of our knowledge, Ref. 22 represents so far the only attempt of such a study. This theoretical work considers models, which require as a parameter the value of the impurity scattering rate $\hbar\Gamma/\Delta$, where $\Delta/k_BT_{c0}\approx 1.76$. This rate can be estimated from the Abrikosov-Gorkov equation:

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \psi\left(\frac{1}{2} + \frac{\hbar\Gamma}{2\pi k_B T_c}\right) - \psi\left(\frac{1}{2}\right), \quad (1)$$

where ψ is a digamma function; $T_{c0}\approx 1.50$ K, transition temperature for a perfectly clean crystal.[36] Figure 2 compares

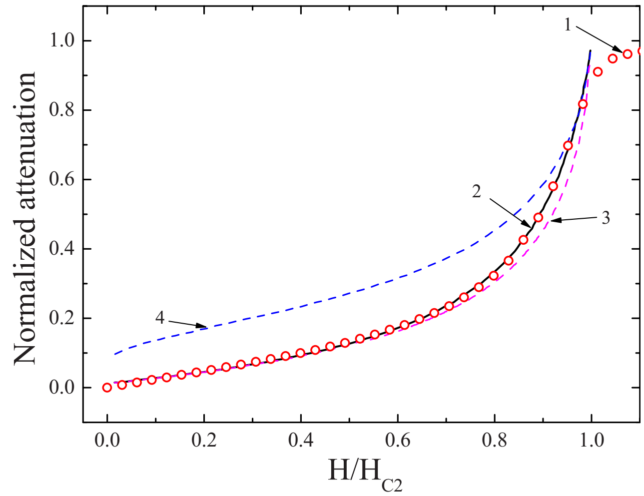

FIG. 2. (Color online) Comparison of the measured ultrasonic attenuation, when superconductivity is suppressed by magnetic field at $T=0.4$ K (curve 1, open circles) with that theoretically predicted (Fig. 1 in Ref. 22) for the cases of vertical nodes with (curve 2, solid) and without (curve 3, dashed) vertices, and horizontal nodes (curve 4, dashed).

our experimental data with theoretical predictions for several models[22] with $\hbar\Gamma/\Delta\sim 0.1$, which should correspond to our case. Clearly, the best overall fit is achieved with assumption of vertical gap nodes with a correction for the vertices. While theorists have proposed several other gap structure models, the lack of calculations of the ultrasonic attenuation based on them does not allow us to compare the theories with our experimental data. Therefore, the question of the nodal topology is still open.

The pressure dependence of T_c is shown in Fig. 3. For comparison, the figure also shows earlier results.[33,34] In all three cases, the transition temperature changes with pressure linearly (in the pressure range studied); what is especially important, the same slope, equal to $dT_c/dP\sim -0.021\pm 0.003$ (K/kbar), holds for all three sets of data points. Figure 3 also shows the pressure evolution of the critical magnetic field value, $H_{c2\parallel ab}$, which is also linear with the slope of $dH_{c2\parallel ab}/dP\sim -0.027\pm 0.003$ (T/kbar).

For the case of simple superconducting metals (Sn, In,

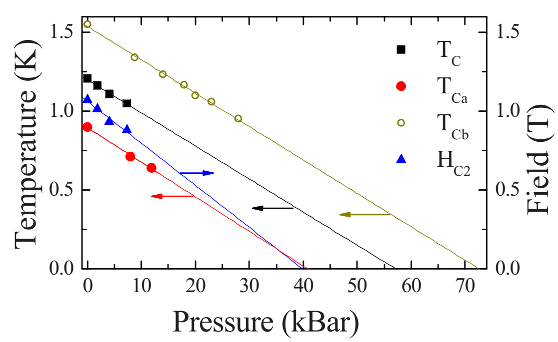

FIG. 3. (Color online) T_c and H_{c2} values of the superconducting transition as a function of pressure. Data from this work (solid squares and triangles) are shown along with those from Refs. 33 (solid circles) and 34 (open circles).

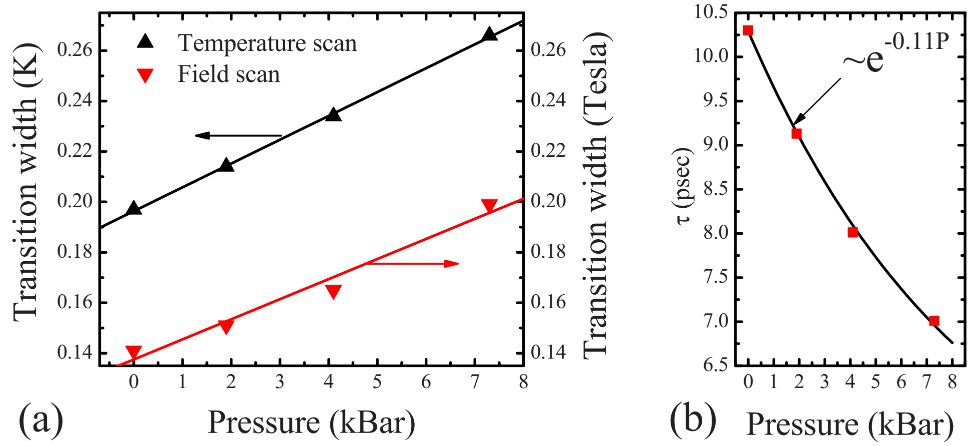

FIG. 4. (Color online) (a) Transition width, measured at half maximum of the derivatives of the curves from Fig. 1, for temperature (up triangles) and magnetic field (down triangles) scans. (b) Pressure dependence of the electron relaxation time; solid line is an exponential fit to the data.

etc.), the reduction of T_c with pressure is a well known phenomenon, where it was explained by modification of the electron-phonon interaction.[37] However, in Sr_2RuO_4, this effect is driven by the electron-electron interaction[34] rather than by the electron-phonon one. We believe that the broadening of the transition [see Fig. 4(a) and Refs. 33 and 34] is another evidence of the above mentioned pressure-induced change of the electron-electron interaction. As we show below, phenomenologically, this change can be described in terms of a decrease in the electron relaxation time [Fig. 4(b)], similar to the previous series of experiments where the relaxation time was reduced by introducing impurities.[24,36]

In Refs. 24 and 38, it was shown that the electron coherence length in the ab plane ξ_{ab} is inversely proportional to the temperature of the superconducting transition. However, perpendicular to the plane, along the c axis, the coherence length ξ_c is determined by the interlayer spacing (actual value is estimated below); consequently, ξ_c does not depend on temperature. At $T=0$, following the Ginzburg-Landau theory for anisotropic superconductors, one can write

$$H_{c2\|ab}(T,T_c)|_{T=0} = \frac{\Phi_0}{2\pi\xi_{ab}\xi_c} \sim \sqrt{\frac{\eta\Phi_0}{2\pi}} \cdot \frac{T_c}{\xi_c}, \qquad (2)$$

where η is an experimentally determined parameter.[24] At a finite but relatively low value of temperature, the function $H_{c2\|ab}(T,T_c)|_{T=\mathrm{const}}$ should have a form, similar to that of $H_{c2\|ab}(T,T_c)|_{T=0}$. Therefore, one can expect a dependence of the upper critical field in the ab plane on the critical temperature to be linear. There are no known reports on any studies of impurity effect on the $H_{c2\|ab}$ value. For this reason, we had to analyze the literature data obtained at the same temperature as that of our measurements, 0.4 K, by different groups on a number of various quality crystals.[7,12,30,31] As it was expected, this analysis showed a linear dependence $H_{c2\|ab}(T,T_c)|_{T=0.4\text{ K}}$ (see Fig. 5). It is remarkable that the data points from our experiment also fall on the same straight line (Fig. 5). This demonstrated that the influence of pressure is similar to the influence of impurities and allows us to use Eq. (1) to estimate the electron relaxation time, whose value drops down exponentially with pressure [Fig. 4(b)]. It is worth noting that the straight line in Fig. 5 intersects the X axis at the point $T \sim 0.4$ K, the temperature of our experiment, confirming the appropriateness of our approach.

The linear dependence $H_{c2\|ab}(T_c)$ differs from the square law dependence of $H_{c2\|c}(T_c)$ explored in Ref. 24, pointing out the layered nature of the material. The least squared straight line fitted over all points (our and taken from literature) gives $dH_{c2\|ab}/dT_c \sim 1.3 \pm 0.1$ T/K at $T=0.4$ K. The value of ξ_c estimated from Eq. (2) is equal to 36 Å in agreement with the literature.[1,38]

In conclusion, we studied the pressure influence on the ultrasound attenuation in Sr_2RuO_4 in the vicinity of the superconducting transition. (i) The magnetic field dependence of attenuation ($H\|\langle 100\rangle$) follows the theoretically predicted

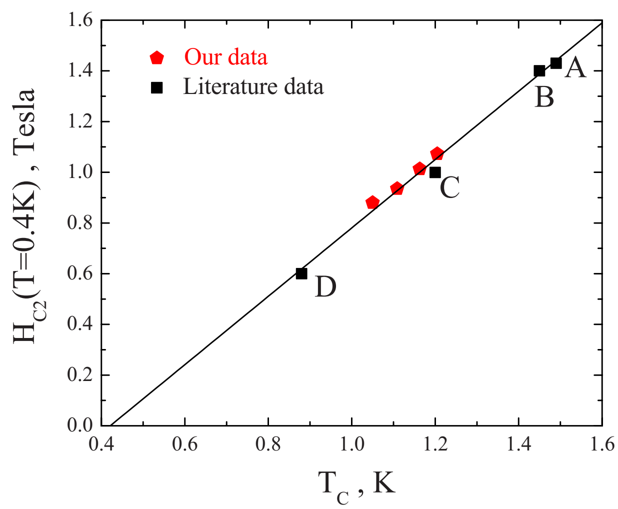

FIG. 5. (Color online) Relationship between $H_{c2\|ab}$ and T_c at $T=0.4$ K. Our data (squares), where T_c changes due to pressure (from right to left: 0, 1.9, 4.1, and 7.3 kbars), are superimposed against the literature data (pentagons), where T_c changes due to impurities (points A, B, C, and D are taken from Refs. 7, 12, 30, and 31, correspondingly). Note that the least squared fitted straight line intersects the horizontal axis at $T \sim 0.4$ K.

curve suggesting the presence of vertical nodes in the superconductive gap. Unfortunately, the absence of theoretical calculations does not allow comparison with other models. (ii) Application of pressure leads to a decrease in T_c and $H_{c2\|ab}$; at the same time, the transition width increases. (iii) We demonstrate that phenomenologically the effect of pressure on T_c can be described in terms of the electron relaxation time decrease and estimate the latter. (iv) Our most interesting finding is the *linear* dependence of $H_{c2\|ab}$ on T_c, unlike the quadratic one for $H_{c2\|c}$.[24] (v) The value of slope $dH_{c2\|ab}/dT_c$ was found to have a universal character, independent of the particular physical factor that causes change in T_c, regardless if it is pressure or impurities.

This work has been performed at the NHMFL, which is supported by the NSF Cooperative Agreement No. DMR-0084173 and the State of Florida. The ultrasonic research at the NHMFL is supported by the In-House Research Program. Also, the work was partially supported by the NNSA/DOE grant No. DE-FG52-03NA00066.

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[1] A. P. Mackenzie and Y. Maeno, Rev. Mod. Phys. **75**, 657 (2003).
[2] C. Bergemann, A. P. Mackenzie, S. R. Julian, D. Forsythe, and E. Ohmichi, Adv. Phys. **52**, 639 (2003).
[3] I. Eremin, D. Manske, S. G. Ovchinnikov, and J. F. Annett, Ann. Phys. **13**, 149 (2004).
[4] T. M. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).
[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, S. R. Julian, A. J. Diver, G. J. McMullan, M. P. Ray, G. G. Lonzarich, Y. Maeno, S. Nishizaki, and T. Fujita, Phys. Rev. Lett. **76**, 3786 (1996); A. P. Mackenzie, S. Ikeda, Y. Maeno, T. Fujita, S. R. Julian, and G. G. Lonzarich, J. Phys. Soc. Jpn. **67**, 385 (1998).
[7] K. Deguchi, M. Tanatar, Z. Mao, T. Ishiguro, and Y. Maeno, J. Phys. Soc. Jpn. **71**, 2839 (2002); K. Deguchi, Z. Mao, and Y. Maeno, J. Phys. Soc. Jpn. **73**, 1313 (2004).
[8] M. J. Graf and A. V. Balatsky, Phys. Rev. B **62**, 9697 (2000).
[9] M. Udagawa, Y. Yanase, and M. Ogata, Phys. Rev. B **70**, 184515 (2004).
[10] R. Arita, S. Onari, K. Kuroki, and H. Aoki, Phys. Rev. Lett. **92**, 247006 (2004); Physica B **359-361**, 584 (2005).
[11] T. Nomura, J. Phys. Soc. Jpn. **74**, 1818 (2005).
[12] M. A. Tanatar, M. Suzuki, S. Nagai, Z. Q. Mao, Y. Maeno, and T. Ishiguro, Phys. Rev. Lett. **86**, 2649 (2001); M. A. Tanatar, S. Nagai, Z. Q. Mao, Y. Maeno, and T. Ishiguro, Phys. Rev. B **63**, 064505 (2001).
[13] M. E. Zhitomirsky and T. M. Rice, Phys. Rev. Lett. **87**, 057001 (2001).
[14] M. B. Walker, M. F. Smith, and K. V. Samokhin, Phys. Rev. B **65**, 014517 (2001).
[15] K. Izawa, H. Takahashi, H. Yamaguchi, Y. Matsuda, M. Suzuki, T. Sasaki, T. Fukase, Y. Yoshida, R. Settai, and Y. Onuki, Phys. Rev. Lett. **86**, 2653 (2001).
[16] Y. Hasegawa and M. Yariyama, J. Phys. Soc. Jpn. **72**, 1318 (2003).
[17] Y. Hasegawa, J. Phys. Soc. Jpn. **72**, 2456 (2003).
[18] J. F. Annett, B. L. Gyorffy, G. Litak, and K. I. Wysokinski, Eur. Phys. J. B **36**, 301 (2003).
[19] G. Litak, J. F. Annett, B. L. Gyorffy, and K. I. Wysokinski, Phys. Status Solidi B **241**, 983 (2004).
[20] P. Contreras, M. Walker, and K. Samokhin, Phys. Rev. B **70**, 184528 (2004).
[21] K. Tenya, S. Yasuda, M. Yokoyama, H. Amitsuka, K. Deguchi, and Y. Maeno, J. Phys. Soc. Jpn. **75**, 23702 (2006).
[22] L. Tewordt and D. Fay, Phys. Rev. B **65**, 104510 (2002); **64**, 024528 (2001).
[23] H.-Y. Kee, Y. B. Kim, and K. Maki, Phys. Rev. B **70**, 052505 (2004).
[24] Z. Q. Mao, Y. Mori, and Y. Maeno, Phys. Rev. B **60**, 610 (1999).
[25] A. M. Balbashov and S. K. Egorov, J. Cryst. Growth **52**, 498 (1981).
[26] A. Suslov, B. K. Sarma, J. Feller, and J. Ketterson, Rev. Sci. Instrum. **77**, 035104 (2006).
[27] S. Tozer (private communication).
[28] G. J. Piermarini, S. Block, J. D. Barnett, and R. A. Forman, J. Appl. Phys. **46**, 2774 (1975).
[29] A. Suslov, O. Svitelskiy, E. Palm, T. Murphy, and D. Shulyatev, in *Low Temperature Physics: 24th International Conference on Low Temperature Physics*, edited by Y. Tanako, S. P. Hershfield, S. O. Hill, P. J. Hirschfeld, and A. M. Goldman (AIP Conf. Proc. Vol. 850, Orlando, 2006), p. 1661.
[30] H. Matsui, M. Yamaguchi, Y. Yoshida, A. Mukai, R. Settai, Y. Onuki, H. Takei, and N. Toyota, J. Phys. Soc. Jpn. **67**, 3687 (1998).
[31] K. Yoshida, Y. Maeno, S. Nishizaki, and T. Fujita, Physica C **263**, 519 (1996).
[32] N. Okuda, T. Suzuki, Z. Mao, Y. Maeno, and T. Fujita, Physica C **388-389**, 497 (2003).
[33] N. Shirakawa, K. Murata, S. Nishizaki, Y. Maeno, and T. Fujita, Phys. Rev. B **56**, 7890 (1997).
[34] D. Forsythe, S. R. Julian, C. Bergemann, E. Pugh, M. J. Steiner, P. L. Alireza, G. J. McMullan, F. Nakamura, R. K. W. Haselwimmer, I. R. Walker, S. S. Saxena, G. G. Lonzarich, A. P. Mackenzie, Z. Q. Mao, and Y. Maeno, Phys. Rev. Lett. **89**, 166402 (2002).
[35] C. Lupien, W. A. MacFarlane, C. Proust, L. Taillefer, Z. Q. Mao, and Y. Maeno, Phys. Rev. Lett. **86**, 5986 (2001).
[36] 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).
[37] J. S. Schilling (private communication).
[38] T. Akima, S. NishiZaki, and Y. Maeno, J. Phys. Soc. Jpn. **68**, 694 (1999).