Comparison of the normal- and superconducting-state electromagnetic absorption in Sr_2RuO_4

G. V. Sudhakar Rao and M. Reedyk
Department of Physics, Brock University, St. Catharines, ON, L2S 3A1, Canada

N. Kikugawa and Y. Maeno
Department of Physics, Kyoto University, Kyoto 606-8502, Japan
and CREST, Japan Science and Technology Corporation, Kawaguchi, Saitama 332-0012, Japan

(Received 29 July 2005; published 28 February 2006)

The ratio of the far-infrared reflectance of Sr_2RuO_4 in the superconducting state R_S to that in the normal state R_N has been measured. For light polarized within the *ab* plane, the superconducting state reflectance is enhanced over that of the normal state below $\approx$20 cm^{-1}, with the thermal reflectance ratio R_S/R_N peaking near 9 cm^{-1}. The energy at which the thermal reflectance peaks, and the magnitude of its enhancement are significantly larger than expected due to the formation of a BCS energy gap. The energy scale coincides with the superconducting energy gap of Sr_2RuO_4 determined via tunneling measurements.

DOI: 10.1103/PhysRevB.73.052507 PACS number(s): 74.25.Gz, 74.70.Pq, 78.30.−j, 74.20.Rp

The discovery of superconductivity in Sr_2RuO_4 (Ref. 1) quickly triggered great interest because of its structural similarity to the high T_C cuprates and its exhibition of multiband unconventional superconductivity (for a review, see, e.g., Refs. 2 and 3). The unconventional characteristics of the superconductivity in Sr_2RuO_4 include the temperature independence of the spin susceptibility across its transition temperature T_C for magnetic fields parallel to the conducting plane,[4,5] the spontaneous appearance of a magnetic field on entering the superconducting state which is a signature of broken time-reversal symmetry,[6] evidence for a two-component order parameter,[7] and the absence of a Hebel-Slichter peak in the nuclear spin relaxation rate $1/T_1$, just below T_C.[8] These experimental findings indicate that the ruthenate exhibits spin triplet superconductivity for which a *p*-wave order parameter is the simplest candidate. However, for a *p*-wave order parameter the excitation spectrum should be fully gapped which implies exponential behavior of physical properties such as the penetration depth, the electronic ultrasound attenuation, the nuclear spin-lattice relaxation rate, the electronic specific heat, and the thermal conductivity, whereas experiments showed a power law behavior of the quasiparticle density of states to the lowest temperatures.[9–16]

A number of theoretical models which take into account the quasi-two-dimensionality of the Fermi surface[17,18] and orbital dependent superconductivity[19] were proposed to explain how the power law dependence of the quasiparticle density of states can arise within a system exhibiting spin-triplet superconductivity.[20–25] The Fermi surface of Sr_2RuO_4 crosses three bands which are referred to as the γ or "active" band, and the α and β or "passive" bands.[26] In these models the γ band is the source of the superconducting instability and has a *p*-wave order parameter. The γ band is close to circular, and thus the anisotropic order parameter would lead to a gap function with minima and maxima at points on the Fermi surface. Recent field-orientation dependent specific heat measurements by Deguchi *et al.*[27] resolved a superconducting gap in the active band with a minimum along the [100] direction. Based on these results a *p*-wave order parameter $\mathbf{d}(\mathbf{k})=\hat{\mathbf{z}}\Delta_o(\sin ak_x+i\sin ak_y)$ where Δ_o is the gap amplitude,[25] was proposed for the γ band. For this **d** vector the spin component of the Cooper pairs is parallel to the RuO_2 plane while the orbital moments align along the *c* axis. The orbital symmetry suppresses scattering of Cooper pairs between bands and as a result the passive bands develop only small amplitude gaps which are modeled to go to zero at intermediate temperatures on a line of the Fermi surface, thus giving rise to the power-law behaviors observed. Note that magnetic fluctuations, assumed to be responsible for the anisotropic Cooper pairing,[28,29] have significant orbital dependence,[30] and thus the temperature onset of the superconducting instability in the passive α and β bands can be lower than that of the active γ band.

In conventional BCS superconductors the energy gap Δ can be directly measured through energy resolved tunneling and electromagnetic spectroscopy techniques. Laube *et al.* investigated the gap function of Sr_2RuO_4 with T_C=1.02 K using point contact spectroscopy.[31] They extracted a value for the gap of, $2\Delta/k_BT_C\approx 17$ using an indirect method of analyzing differential resistance as a function of applied bias in terms of Andreev bound states based on the assumption of the order parameter $\mathbf{d}(\mathbf{k})=\hat{\mathbf{z}}\Delta_o(k_x+ik_y)$ for the chiral *p*-wave state. However, the work of Deguchi *et al.*, which suggests that this is not the correct order parameter for Sr_2RuO_4, places a caveat on this result. Laube *et al.* used excess current measurements[32] to explain the unusually large value of $2\Delta/kT_C$=17. An additional pair breaking channel is invoked to reconcile the magnitude of the superconducting gap and the temperature dependent excess current. The scattering between quasiparticles and low energy bosonic fluctuations, which probably originate from spin fluctuations in the superconducting order parameter, is proposed. Upward *et al.* performed a more direct measurement of the gap by means of measurements of the tunneling conductance parallel to the *c* axis via scanning tunneling spectroscopy experiments.[33] Their results show a suppression in the density of states of Sr_2RuO_4 which disappears at temperatures above T_C and in the presence of a field larger than the critical magnetic field. They determined $2\Delta_{max}/kT_C$=8.0 at the surface, where Δ_{max} is the maximum value of the gap in *k* space. The relationship between Δ_{max} and Δ_o is dependent on the geometry of the Fermi surface and the model order parameter; see, for example, Ref. 25. Since the value of the gap at the surface may be reduced from the bulk as a result of surface disorder, $8.0kT_C$ represents a lower bound for $2\Delta_{max}$ in the bulk. They attributed this value, which is still substantially larger than that expected within BCS theory for an isotropic gap within the weak coupling regime, to strong anisotropy of the superconducting gap within the RuO_2 plane. While they question the validity of the analysis of Laube *et al.*, they note that their result, which being a *lower* bound, is not inconsistent with the conclusion of Laube *et al.*

We have taken an electromagnetic approach in an attempt to gain further information regarding the superconducting state of Sr_2RuO_4. An advantage of this technique is that it is not surface sensitive since electromagnetic radiation penetrates on the order of a micrometer into the crystal. We used very-far-infrared spectroscopy to compare the low frequency reflectance of a high quality Sr_2RuO_4 single crystal with a T_C of 1.42 K and transition width δT_C of 24 mK in the normal and superconducting states. This method has been used to extract the magnitude of the superconducting gap in conventional dirty limit superconductors with an isotropic *s*-wave order parameter.[34,35] In such a case, since the density of states is zero within the gap region at $T=0$, absorption of radiation with frequency less than the magnitude of the gap is prohibited, unit reflectance is observed and the real conductivity, σ, vanishes for $0<\omega\leq 2\Delta$, except for a δ-function peak at zero frequency due to the superconducting condensate. The conductivity is expected to increase monotonically from zero just above 2Δ.

In the case of anisotropic unconventional superconductors, measurement of the superconducting energy gap is not straightforward, but has been modeled by Hirschfeld *et al.*[36] They considered an isotropic *p*-wave state as well as model anisotropic states with points and lines of nodes on the Fermi surface. They found that in general the frequency dependence of the electromagnetic absorption in the superconducting state compared to the normal state varied considerably from the singlet isotropic pairing case. They examined both strong and weak impurity scattering strengths and calculated the ratio of the superconducting state to normal state real conductivity, σ_S/σ_N, for various scattering rates. They found that for the isotropic *p*-wave case in the presence of weak scatterers that unlike the BCS case there is a discontinuous jump in absorption from zero to a finite value at 2Δ in the clean limit. In the resonant strong-scattering limit they found a dominant absorption edge at $\omega=\Delta$ with the possibility of some absorption below Δ as well, due to the presence of bound states near $\omega=0$. For nonresonant scattering the threshold shifts to near 1.5Δ due to scattering processes between a gap edge, and the bound state, which has moved away from zero energy. For anisotropic states they found that there is finite absorption to zero frequency, but that the specific frequency dependence depends on the details of the state. For strong (resonant) scattering σ_S/σ_N shows a broad peak centered near Δ, which is absent for weak scattering. In the weak scattering limit the frequency dependence of the electromagnetic absorption obeys a power law which is determined by the symmetry of the order parameter.

The work of Hirschfeld *et al.* shows that in principle, the form of the electromagnetic absorption can be useful for gaining information concerning the gap amplitude, and the structure of the order parameter of the superconducting state in the clean limit. Finite temperature, or increasing magnitude of the scattering rate relative to T_C degrades distinctive structure making it more difficult to distinguish between states.[36] Another complication is that collective modes of the order parameter may also play a role in the electromagnetic response of unconventional superconductors where they can contribute to the power absorption at frequencies below the quasiparticle gap edge at 2Δ.[36–38] Their response will look similar to the bound-state peak centered near Δ that occurs in the strong-impurity scattering limit.[36]

Our measurements were carried out on a single crystal sample of Sr_2RuO_4 grown in an image furnace by the floating zone method. We measured an *ab*-plane face which was 8 mm long and 4 mm wide. In each case the power reflected from the sample was ratioed to that from a reference mirror at temperatures above and below the T_C of 1.42 K using a Helium-3 Cryostat and step-and-integrate Martin-Pupplett-type polarizing interferometer. This instrumentation is efficient for very low frequency measurements. The measurements were repeated with a metallic gold film evaporated *in situ* onto the sample in order to ensure that the temperature dependence observed was intrinsic to the sample. In principle the absolute reflectance R could be determined using this technique[39] however, a reduced intensity of the power spectrum at these low frequencies, and the fact that there is little difference between the high reflectance of the sample and the metallic gold film, resulted in the additional ratioing required by this method of extracting R, increasing the noise to unacceptable levels. Only the reflectance ratio between temperatures below and above T_C is thus presented. We refer to such ratios of the reflectance at two different temperatures as "thermal reflectance" which represents the relative change in the reflectance between the two temperatures.

Figure 1(a) shows the frequency dependence of the in-plane thermal reflectance, defined to be the ratio of the reflectance in the superconducting state at $T=0.5$ K to that in the normal state at $T=1.6$ K for measurements on the *ab* face. Within the scatter of the data beyond $\approx 35\ \mathrm{cm}^{-1}$ the thermal reflectance ratio is approximately one indicating that the reflectance above and below T_C is the same. At lower frequencies the thermal reflectance shows structure. Below $\approx 20\ \mathrm{cm}^{-1}$ the superconducting state reflectance is enhanced by a maximum of $\approx 1\%$ over that in the normal state. This implies that the electromagnetic absorption A in the superconducting state is less than in the normal state since $R=1-A$ for bulk metallic samples in the far infrared. In conventional superconductors a monotonic Drude-like decrease of the reflectance with increasing frequency is expected in the normal state while in the superconducting state there is 100% reflectance below 2Δ, beyond which there is an onset of absorption due to excitation of electrons across the energy gap. Thus, the ratio of the superconducting to normal state reflectance will be enhanced at low frequencies, peaking at 2Δ. A BCS calculation assuming a Hagen-Rubens normal state reflectance corresponding to the *ab*-plane resistivity of Sr_2RuO_4 at 1.6 K of $\approx 2\ \mu\Omega\,\mathrm{cm}^1$ however yields an en-

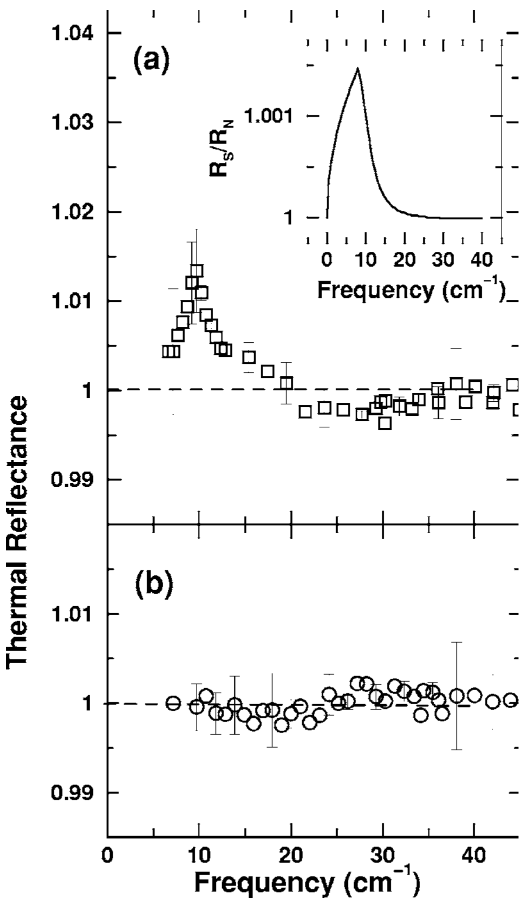

FIG. 1. (a) Frequency dependence of the ratio of the reflectance in the superconducting state to that at 1.6 K in the normal state measured on an *ab* face [squares]. The inset shows the BCS prediction for a normal state resistivity of 2 $\mu\Omega$ cm. (b) The ratio of reflectances for the gold coated sample at low and high temperatures [circles].

hancement that is more than an order of magnitude less than that observed experimentally as shown in the inset to Fig. 1(a). This result is not very sensitive to variation in the *ab*-plane resistivity which ranges from 1–2 $\mu\Omega$ cm.[1,40] While the R_S/R_N enhancement in the BCS calculation increases as the *ab*-plane resistivity increases, an *ab*-plane resistivity of 10 $\mu\Omega$ cm, which is five to ten times larger than literature values for Sr_2RuO_4, yields a peak in R_S/R_N that is five times less than the observed experimental result.

Figure 1(b) shows the ratio of reflectances for the gold coated sample at low and high temperatures. The thermal reflectance ratio for the gold coated sample is within uncertainty approximately one for the frequency range investigated indicating that the reflectance is the same at 0.5 K and at 1.6 K. Note that in order to increase the signal to noise each curve of Figs. 1(a) and 1(b) represents the average result for an experiment which consisted of several days of data collection. While these curves represent the best sets of data obtained, other experiments produced similar results. Error bars calculated using standard deviation are shown at some frequencies.

As discussed above, the experimental challenges of making these measurements at low frequencies preclude our ability to determine the absolute reflectance from which, with Kramers-Kronig analysis, one could obtain the optical conductivity. The optical conductivity could be used to make comparisons with the predictions of Hirschfeld *et al.* for unconventional superconductors, with the caveat that their work is limited to three-dimensional (3D) gaps while the Sr_2RuO_4 system is closer to 2D. What are needed are predictions for the frequency dependence of the experimentally measurable quantity R_S/R_N, from which it would be possible to elucidate information concerning the magnitude of the energy gap and the structure of the superconducting order parameter in unconventional 2D and 3D superconductors. Our measurements bear out that for light polarized within the planes there is increased reflectance in the superconducting state over an interval of $\approx$20 cm^{-1} which peaks near 9 cm^{-1}. The magnitude of the enhancement is significantly larger than expected for a conventional BCS superconductor. Since Sr_2RuO_4 is established to be an unconventional anisotropic superconductor the method of using the peak in R_S/R_N to determine 2Δ, appropriate for conventional isotropic *s*-wave superconductors, is likely inapplicable. The work of Hirschfeld *et al.*[36] shows however that even for unconventional 3D superconductors the changes in electromagnetic absorption due to the onset of superconductivity take place within an interval of order 2Δ. We note that the range of the interval where greater absorption is observed in the normal state relative to the superconducting state in Sr_2RuO_4 is roughly comparable to the size of the gap deduced by Laube *et al.* while the peak location corresponds approximately to the lower limit placed on the gap by Upward *et al.* This suggests that the enhanced reflectance we observe at low temperatures is due to the onset of superconductivity, and we conclude that our results provide independent confirmation (i.e., by a technique distinct from tunneling) that $2\Delta/k_BT_C$ for Sr_2RuO_4 is considerably larger than the BCS result.

ACKNOWLEDGMENT

This work was supported by the Natural Sciences and Engineering Research Council of Canada.

[1] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednortz, and F. Lichtenberg, Nature (London) **372**, 532 (1994).

[2] A. Mackenzie and Y. Maeno, Rev. Mod. Phys. **75**, 657 (2003).

[3] C. Bergemann, A. P. Mackenzie, S. R. Julian, D. Forsythe, and E. Ohmichi, Adv. Phys. **52**, 639 (2003).

[4] K. Ishida, H. Mukuda, Y. Kitaoka, Z. Q. Mao, H. Fukazawa, and Y. Maeno, Phys. Rev. B **63**, 060507(R) (2001).

[5] J. A. Duffy, S. M. Hayden, Y. Maeno, Z. Mao, J. Kulda, and G. J. McIntyre, Phys. Rev. Lett. **85**, 5412 (2000).

[6] G. M. Luke, Y. Fudamoto, K. M. Kojima, M. I. Larkin, J. Merrin, B. Nachumi, Y. J. Uemura, Y. Maeno, Z. Q. Mao, Y. Mori, H. Nakamura, and M. Sigrist, Nature (London) **394**, 558 (1998).
[7] P. G. Kealey, T. M. Riseman, E. M. Forgan, L. M. Galvin, A. P. Mackenzie, S. L. Lee, D. McK. Paul, R. Cubitt, D. F. Agterberg, R. Heeb, Z. Q. Mao, and Y. Maeno, Phys. Rev. Lett. **84**, 6094 (2000).
[8] K. Ishida, Y. Kitaoka, K. Asayama, S. Ikeda, S. Nishizaki, Y. Maeno, K. Yoshida, and T. Fujita, Phys. Rev. B **56**, R505 (1997).
[9] I. Bonalde, B. D. Yanoff, M. B. Salamon, D. J. Van Harlingen, E. M. E. Chia, Z. Q. Mao, and Y. Maeno, Phys. Rev. Lett. **85**, 4775 (2000).
[10] K. Ishida, H. Mukuda, Y. Kitaoka, Z. Q. Mao, Y. Mori, and Y. Maeno, Phys. Rev. Lett. **84**, 5387 (2000).
[11] C. Lupien, W. A. MacFarlane, Cyril Proust, Louis Taillefer, Z. Q. Mao, and Y. Maeno, Phys. Rev. Lett. **86**, 5986 (2001).
[12] S. Nishizaki, Y. Maeno, and Z. Q. Mao, J. Low Temp. Phys. **117**, 1581 (1999).
[13] K. Izawa, H. Takahashi, H. Yamaguchi, Yuji Matsuda, M. Suzuki, T. Sasaki, T. Fukase, Y. Yoshia, R. Settai, and Y. Onuki, Phys. Rev. Lett. **86**, 2653 (2001).
[14] M. A. Tanatar, M. Suzuki, S. Nagai, Z. Q. Mao, Y. Maeno, and T. Ishiguro, Phys. Rev. Lett. **86**, 2649 (2001).
[15] M. A. Tanatar, S. Nagai, Z. Q. Mao, Y. Maeno, and T. Ishiguro, Phys. Rev. B **63**, 064505 (2001).
[16] M. Suzuki, M. A. Tanatar, N. Kikugawa, Z. Q. Mao, Y. Maeno, and T. Ishiguro, Phys. Rev. Lett. **88**, 227004 (2002).
[17] 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).
[18] C. Bergemann, S. R. Julian, A. P. Mackenzie, S. Nishizaki, and Y. Maeno, Phys. Rev. Lett. **84**, 2662 (2000).
[19] D. F. Agterberg, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. **78**, 3374 (1997).
[20] M. E. Zhitomirsky and T. M. Rice, Phys. Rev. Lett. **87**, 057001 (2001).
[21] J. F. Annett, G. Litak, B. L. Gyorffy, and K. I. Wysokinski, Phys. Rev. B **66**, 134514 (2002).
[22] S. Koikegami, Y. Yoshida, and T. Yanagisawa, Phys. Rev. B **67**, 134517 (2003).
[23] T. Nomura and K. Yamada, J. Phys. Soc. Jpn. **71**, 404 (2002).
[24] Y. Yanese and M. Ogata, J. Phys. Soc. Jpn. **72**, 673 (2003).
[25] K. Miyake and O. Narikiyo, Phys. Rev. Lett. **83**, 1423 (1999).
[26] I. I. Mazin and D.J. Singh, Phys. Rev. Lett. **79**, 733 (1997).
[27] K. Deguchi, Z. Q. Mao, H. Yaguchi, and Y. Maeno, Phys. Rev. Lett. **92**, 047002 (2004).
[28] T. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).
[29] I. I. Mazin and D. J. Singh, Phys. Rev. Lett. **82**, 4324 (1999).
[30] T. Imai, A. W. Hunt, K. R. Thurber, and F. C. Chou, Phys. Rev. Lett. **81**, 3006 (1998).
[31] F. Laube, G. Goll, H. v. Lohneysen, M. Fogelstrom, and F. Lichtenberg, Phys. Rev. Lett. **84**, 1595 (2000).
[32] F. Laube, G. Goll, M. Eschrig, M. Fogelstrom, and R. Werner, Phys. Rev. B **69**, 014516 (2004).
[33] M. D. Upward, L. P. Kouwenhoven, A. F. Morpurgo, N. Kikugawa, Z. Q. Mao, and Y. Maeno, Phys. Rev. B **65**, 220512(R) (2002).
[34] P. L. Richards and M. Tinkham, Phys. Rev. Lett. **1**, 318 (1958).
[35] P. L. Richards and M. Tinkham, Phys. Rev. **119**, 575 (1960).
[36] P. J. Hirschfeld, P. Wolfle, J. A. Sauls, D. Einzel, and W. Putikka, Phys. Rev. B **40**, 6695 (1989).
[37] D. S. Hirashima and H. Namaizawa, Prog. Theor. Phys. **77**, 585 (1987).
[38] S. Higashitani and K. Nagai, Phys. Rev. B **62**, 3042 (2000).
[39] C. C. Homes, M. Reedyk, D. A. Crandles, and T. Timusk, Appl. Opt. **32**, 2976 (1993).
[40] N. E. Hussey, A. P. Mackenzie, J. R. Cooper, Y. Maeno, S. Nishizaki, and T. Fujita, Phys. Rev. B **57**, 5505 (1998).