

## Tunneling spectroscopy of the heavy-fermion superconductor $\text{CeCu}_2\text{Si}_2$

I. Iguchi, T. Yasuda, Y. Ōnuki, and T. Komatsubara

*Institute of Materials Science, University of Tsukuba, Sakura-mura, Ibaraki 305, Japan*

(Received 24 February 1987)

Single-particle tunneling spectroscopy between a  $\text{CeCu}_2\text{Si}_2$  ingot and a Pb film is presented. The observed dynamic tunnel resistance as a function of junction voltage under an applied magnetic field shows an asymmetric behavior typical of valence-fluctuation compounds. Below the transition temperature of  $\text{CeCu}_2\text{Si}_2$  under no magnetic field, a series of small structures related to  $\text{CeCu}_2\text{Si}_2$  gap properties develop. The result suggests that  $\text{CeCu}_2\text{Si}_2$  may be a singlet  $d$ -wave superconductor according to the tunnel calculation of Tachiki, Nakahara, and Teshima.

The heavy-fermion superconductors  $\text{CeCu}_2\text{Si}_2$ ,<sup>1</sup>  $\text{UBe}_{13}$ ,<sup>2</sup> and  $\text{UPt}_3$  (Ref. 3) have attracted considerable recent attention both theoretically and experimentally in the field of solid-state physics. The main feature of these materials is that the electronic specific heat becomes  $10^2$ – $10^3$  times greater than that of usual materials, suggesting heavy-mass fermions. It has been pointed out that the superconductivity induced in these materials is quite anisotropic in that the superconducting energy gap vanishes at points or lines on the Fermi surface as judged from the experimental data on specific heat,<sup>4,5</sup> nuclear magnetic relaxation,<sup>6,7</sup> and ultrasonic absorption.<sup>8</sup> It has been argued also that the superconducting state may be due to even-parity pairing (singlet)<sup>9–11</sup> or odd-parity pairing (triplet).<sup>12,13</sup>

A tunneling study of these materials would certainly provide important information on superconductivity mechanism, but no conclusive data have been reported yet. All the measurements so far performed have relied on point-contact spectroscopy. Poppe<sup>14</sup> reported 15%–80% of Josephson current flow between  $\text{CeCu}_2\text{Si}_2$  and Al. Very recently, Han *et al.*<sup>15</sup> performed the point-contact spectroscopy between Nb and  $\text{UBe}_{13}$  below the transition temperature of  $\text{UBe}_{13}$  and found a proximity effect due to possible  $p$ -wave pairing.

In this Rapid Communication, we present single-particle tunneling measurements between  $\text{CeCu}_2\text{Si}_2$  and Pb. The sample was a polycrystalline  $\text{CeCu}_2\text{Si}_2$  ingot of about 7 mm outer diameter grown by the Czochralski method under Ar pressure of about 10 atm. The sample surface was polished using fine powder, then coated with photoresist for electrical insulation, except for the junction area. Next, the sample was set in a vacuum chamber and its surface was oxidized by the dc glow-discharge technique. Finally, a Pb film of 0.5 mm width was deposited by a resistive heater. The junction area was  $0.4 \times 0.5 \text{ mm}^2$  and its resistance ranged from 0.05–1.0  $\Omega$ . The junction thus fabricated was stable and could be used repeatedly.

The sample surface after oxidation was checked by Auger electron spectroscopy. Figure 1 shows an Auger profile. The oxygen-rich layer was about 3 nm as estimated from a measurement using a step-height stylus, suggesting the formation of an appropriate oxide layer for tunneling. The sample was tested utilizing a  $^3\text{He}$  cryostat.

Figure 2 shows the dynamic tunnel resistance versus junction voltage  $V$  at  $T=0.8 \text{ K}$  under various applied magnetic fields. The normal resistance of this junction was 0.16  $\Omega$ . When  $H=0$ , the gap structure due to the Pb film is clear. As the magnetic field is increased, the Pb gap becomes smaller and disappears at about  $H=1 \text{ kG}$ . Above  $H=1 \text{ kG}$ , the asymmetric resistance structure due to  $\text{CeCu}_2\text{Si}_2$  appears. Although the tunneling structure at  $H=0$  shows a typical  $dV/dI$  peak at  $V=0$  and a  $dV/dI$  minimum at  $V \approx \Delta_{\text{Pb}}/e$ , it has been wholly lowered below the normal resistance, for which the reason is not clear. It may not be a special property of  $\text{CeCu}_2\text{Si}_2$  since a similar structure was also observable for a heavy-fermion single crystal of  $\text{CeCu}_6$ .

Figure 3 shows the dynamic tunnel resistance as a function of junction voltage at  $T=0.8 \text{ K}$  and  $H=30 \text{ kG}$ . The top curve is the measured one, while the bottom and middle curves correspond to the symmetric and asymmetric component of the measured curve, respectively. The observed behavior quite resembles the general features of

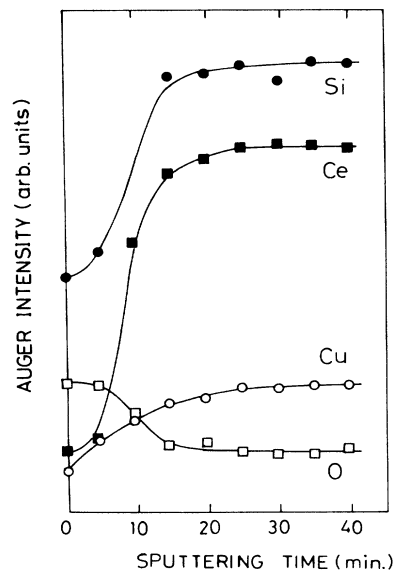


FIG. 1. Auger profile. The solid lines were drawn as a guide for the eye.

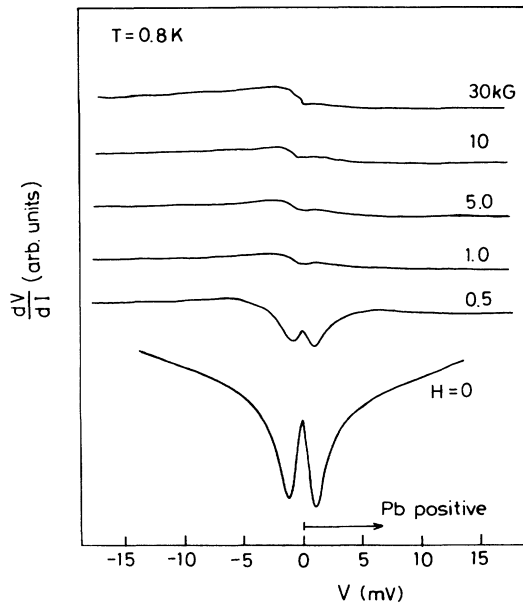


FIG. 2. Dynamic tunnel resistance as a function of junction voltage at  $T=0.8$  K under various applied magnetic field. The curves are each shifted by an appropriate amount.

intermediate-valence compounds.<sup>16,17</sup> The dip at  $V=0$  in the symmetric curve suggests the peaking of the density of states at the Fermi level since the conductance of a tunnel junction becomes proportional to the density of states. This fact is consistent with the presence of Kondo resonant state. The peak in the symmetric curve and the

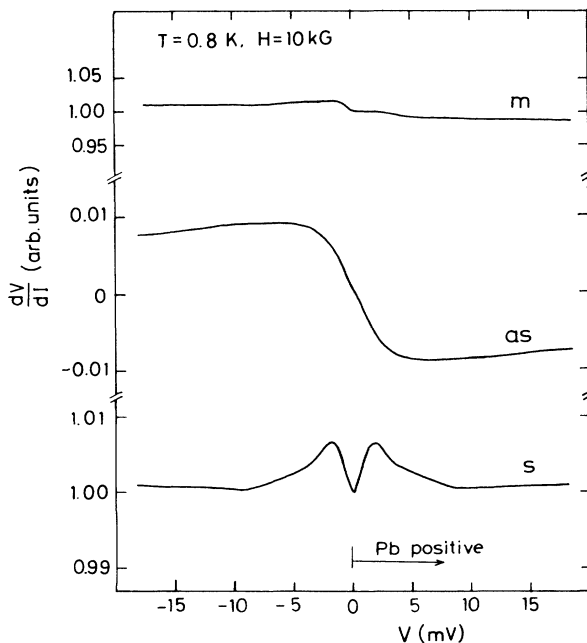


FIG. 3. Dynamic tunnel resistance as a function of junction voltage at  $T=0.8$  K when  $H=10$  kG. *m* is the measured curve, *s* is the symmetric part, and *as* is the asymmetric part.

prominent slope change in the asymmetric curve near 2 mV are probably due to the first excitation energy in the interconfigurational transition according to the work of Bussian, Frankowski, and Wohlleben.<sup>16</sup>

Figure 4 shows the result of the dynamic tunnel resistance plotted against the junction voltage at  $H=0$  and  $T=0.58$  K below the transition temperature of  $\text{CeCu}_2\text{Si}_2$  ( $T_c=0.74$  K). As the bath temperature is lowered below  $T_c$ , a weak modulated structure appears and develops around the Pb gap structure. We believe that this structure is due to  $\text{CeCu}_2\text{Si}_2$ . There exist five small structures as shown by arrows, two below and two above  $(dV/dI)_{\text{min}}$ , and one at the center, which are reproducible. The relatively weak structure may possibly be attributed to predominant tunneling into the nonsuperconducting region where the inclusion of oxygen disturbed the stoichiometry of  $\text{CeCu}_2\text{Si}_2$ . A Josephson current was not observed probably because the oxide layer was too thick to allow Josephson tunneling and/or a very weak gap structure was observed; hence this fact does not preclude possible Josephson-current flow.

Suppose that the observed structure corresponds to the derivative of the conventional gap structure of current rise and cusp between dissimilar superconductors; then we recognize the two peak structures above  $\Delta_{\text{Pb}}$ , i.e., at  $\Delta_{\text{Pb}}+\Delta^*$  (*a*) and  $\Delta_{\text{Pb}}+\Delta^{**}$  (*b*), and the derivative of two cusp structures below  $\Delta_{\text{Pb}}$ , i.e., at  $\Delta_{\text{Pb}}-\Delta^*$  (*a'*) and  $\Delta_{\text{Pb}}-\Delta^{**}$  (*b'*). In addition,  $\Delta_{\text{Pb}}$  peak *c* is also present, suggesting the coexistence of a finite gap and zero gap (gapless). In other words, the result is consistent with the picture that the energy gap in a heavy-fermion superconductor is quite anisotropic and vanishes at points or lines on the Fermi surface. The values of  $\Delta^*$  and  $\Delta^{**}$  are

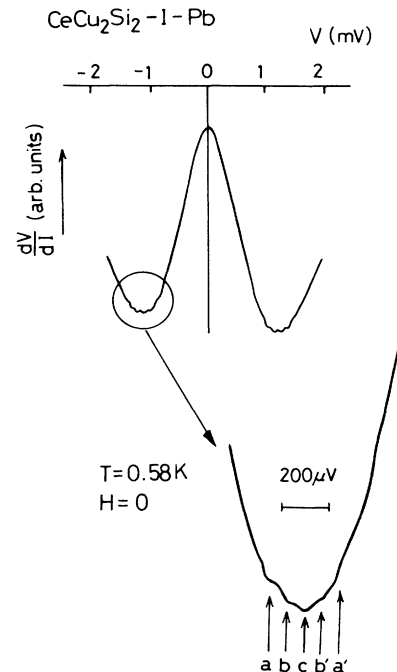


FIG. 4. Dynamic tunnel resistance as a function of junction voltage at  $T=0.58$  K.

about 0.15 and 0.075 meV, respectively ( $\Delta^{**} \approx \Delta^*/2$ ); hence  $\Delta^*/k_B T_c = 2.3 \pm 0.2$  by taking into account the finite resistive transition width, which is already much greater than the BCS value of 1.76. The expected value of  $\Delta_0^*/k_B T_c$ , where  $\Delta_0^*$  is the zero-temperature gap parameter, will be  $3.2 \pm 0.2$  by assuming the BCS temperature dependence.

Recently, Tachiki, Nakahara, and Teshima<sup>18</sup> calculated the tunnel conductance for single-particle tunneling between a normal-metal and a heavy-fermion superconductor, as well as the expected value of the universal constant  $\Delta_0^*/k_B T_c$  for *p*- and *d*-wave superconductors at  $T=0$ . In case of *p*-wave superconductors, all three phases (Anderson-Brinkman-Morel and planar, polar, and Balian-Werthame) have a characteristic peak at  $V = \Delta_0^*/e$ . On the other hand, for *d*-wave superconductors, two phases (type I and type II) were considered. The tunnel conductance of the type-I phase has peaks at  $V = \Delta_0^*/2e$  and  $V = \Delta_0^*/e$ , whereas the type-II phase has only at  $V = \Delta_0^*/e$ . The universal constant  $\Delta_0^*/k_B T_c$  was 2.462 for the polar phase, 2.029 for the ABM and planar phase, 1.764 for the BW phase, 3.087 for type I phase, and 2.563 for type-II phase. In comparison with the data

obtained, we conclude that CeCu<sub>2</sub>Si<sub>2</sub> may be close to a singlet *d*-wave superconductor (type I) because the two structures corresponding to  $\Delta^*/2$  and  $\Delta^*$  were observable, and the estimated universal constant was large.

In conclusion, we have presented the measurements of single-particle tunneling between a CeCu<sub>2</sub>Si<sub>2</sub> ingot and a Pb film. The tunneling characteristics showed strong asymmetry around zero bias under an applied magnetic field, typical for valence-fluctuation compounds. Below the transition temperature of CeCu<sub>2</sub>Si<sub>2</sub>, the gap structure due to CeCu<sub>2</sub>Si<sub>2</sub> was observed, which suggests a possible singlet *d*-wave superconductor according to the theory of Tachiki *et al.*<sup>18</sup>

More experimental measurements under lower-temperature conditions using a single-crystal sample with a junction of better quality are necessary, and we encourage future investigation.

The authors are indebted to Dr. M. Shinoki in the Electrotechnical Laboratory for the Auger-electron spectroscopy measurements. One of us (I.I.) is also grateful to Professor M. Tachiki for helpful discussions and for sending up copies of his work.

- <sup>1</sup>F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Mescheded, W. Franz, and H. Schäfer, *Phys. Rev. Lett.* **43**, 1892 (1979).  
<sup>2</sup>H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, *Phys. Rev. Lett.* **50**, 1595 (1983).  
<sup>3</sup>G. R. Stewart, Z. Fisk, J. O. Willis, and J. L. Smith, *Phys. Rev. Lett.* **52**, 679 (1984).  
<sup>4</sup>H. R. Ott, H. Rudigier, T. M. Rice, K. Ueda, Z. Fisk, and J. L. Smith, *Phys. Rev. Lett.* **52**, 1915 (1984).  
<sup>5</sup>F. Steglich, C. D. Bredl, W. Lieke, U. Rauchschwalbe, and G. Sparn, *Physica* **126B+C**, 82 (1984).  
<sup>6</sup>Y. Kitaoka, K. Ueda, T. Kohara, and K. Asayama, *Solid State Commun.* **51**, 461 (1984).  
<sup>7</sup>D. E. MacLaughlin, Cheng Tien, W. G. Clark, M. D. Lan, Z. Fisk, J. L. Smith, and H. R. Ott, *Phys. Rev. Lett.* **53**, 1833 (1984).  
<sup>8</sup>D. J. Bishop, C. M. Varma, B. Batlogg, E. Bucher, Z. Fisk, and J. L. Smith, *Phys. Rev. Lett.* **53**, 1009 (1984).

- <sup>9</sup>M. Tachiki and S. Maekawa, *Phys. Rev. B* **29**, 2497 (1984).  
<sup>10</sup>F. J. Ohkawa and H. Fukuyama, *J. Phys. Soc. Jpn.* **53**, 4344 (1984).  
<sup>11</sup>T. Matsuura, K. Miyake, H. Jichu, and Y. Kuroda, *Prog. Theor. Phys.* **72**, 402 (1984).  
<sup>12</sup>P. W. Anderson, *Phys. Rev. B* **30**, 1549 (1984).  
<sup>13</sup>C. M. Varma, *Bull. Am. Phys. Soc.* **29**, 404 (1984).  
<sup>14</sup>U. Poppe, *J. Magn. Magn. Mater.* **52**, 157 (1985).  
<sup>15</sup>S. Han, K. W. Ng, E. L. Wolf, A. Millis, J. L. Smith, and Z. Fisk, *Phys. Rev. Lett.* **57**, 238 (1986).  
<sup>16</sup>B. Bussian, I. Frankowski, and D. Wohlleben, *Phys. Rev. Lett.* **49**, 1026 (1982).  
<sup>17</sup>I. Frankowski and P. Wachter, *J. Appl. Phys.* **53**, 7887 (1982).  
<sup>18</sup>M. Tachiki, M. Nakahara, and R. Teshima, *J. Magn. Magn. Mater.* **52**, 161 (1985).