Magnetic-field modulation of the Josephson effect between polycrystalline CeCu$_2$Si$_2$ and Al

A Sumiyama, N Miyakawa, Y Ushida, G Motoyama, A Yamaguchi and Y Oda

Graduate school of Material Science, University of Hyogo, Ako-gun, Hyogo 678-1297, Japan
E-mail: sumiyama@sci.u-hyogo.ac.jp

Abstract. Josephson critical current $I_c$ between a polycrystalline CeCu$_2$Si$_2$ and Al has been measured for the junctions on a CeCu$_2$Si$_2$ surface. A dominant maximum peak at zero magnetic field is observed in the magnetic field $H$ dependence of $I_c$, suggesting that the Josephson currents flowing from variously-oriented crystallites of CeCu$_2$Si$_2$ are in phase at $H = 0$. If CeCu$_2$Si$_2$ is in the $d_{x^2-y^2}$-wave state, in which the sign of the order parameter changes according to the crystallographic orientation, an $s$-wave component induced on the surface is considered as a possible origin of the $s$-wave-like behavior of $I_c(H)$ patterns.

CeCu$_2$Si$_2$, which is the first heavy-fermion superconductor discovered in 1979[1], has attracted much attention in the recent years, since the superconducting transition occurs in the vicinity of a quantum critical point. As expected in superconductors close to an antiferromagnetic instability, a variety of physical properties in CeCu$_2$Si$_2$ suggest the $d$-wave superconductivity: the $d_{x^2-y^2}$ state is proposed to be the most reasonable candidate[2, 3]. Recently, another superconducting state has been found in the pressure-temperature phase diagram of CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ and ascribed to a different pairing mechanism, because it occurs far away from the threshold of magnetism[4].

The Josephson effect between a conventional ($s$-wave) and an unconventional (non $s$-wave) superconductor gives direct information about the order parameter of the unconventional superconductor. For the high-$T_c$ cuprates, the magnetic field dependence of Josephson critical current $I_c$, has given conclusive evidence for the $d_{x^2-y^2}$-wave superconductivity[5]. The Josephson effect of CeCu$_2$Si$_2$ has been investigated in point contacts between CeCu$_2$Si$_2$ and a conventional superconductor by Poppe et al[6]. They observed $I_c$ values between 15 and 80 % of the Ambegaokar-Baratoff limit. In our previous paper we have compared the Josephson effect between S(Nb)-N(Cu)-S’(CeCu$_2$Si$_2$) and S(Nb)-N(Cu)-S(Nb) junctions and found no significant suppression of $I_c$ due to the unconventional superconductivity of CeCu$_2$Si$_2$[7]. In this paper, the magnetic field dependence of Josephson critical currents that come from multiple crystallites of polycrystalline CeCu$_2$Si$_2$ has been investigated and the existence of $s$-wave order parameter is discussed.

A polycrystalline sample of nominal composition CeCu$_2$Si$_2$ was prepared by argon-arc melting the corresponding amounts of the elements. The sample was annealed in Ar at 900°C for 5 days. Figure 1(a) shows the orientation imaging micrograph of the polished surface obtained by EBSD (Electron Backscatter Diffraction) patterns. The colors indicate the crystallographic orientation normal to the surface. Typical crystallites are thin long ones in a width of about 30
μm; crystallites in different longitudinal directions tend to show different crystal faces.

The polycrystal was cut to slices of 1 mm thickness to use as a substrate. The surface was then polished with diamond polish down to a grain size of 1 μm, resulting in a flat mirror-like surface. In order to inspect the grain structure of the junction area, two of six junctions, denoted as J-1 and J-2, were made on the substrate which was lightly etched in dilute nitric acid to produce a microstructural contrast. The other four junctions (J-3, 4, 5, 6) were made on the mirror-like surface.

The CeCu$_2$Si$_2$ (S) substrate was set in a sputtering apparatus and a SNS' junction was prepared; after the surface was sputter cleaned by Ar ion, Cu doped with 5 wt% Al (N: normal metal), SiO$_2$ and Al (S': s-wave superconductor) were deposited successively using rf-sputtering technique, as shown in Figs. 1(b) and (c). Even after the deposition, it is still visible that the junction area contains many crystallites in different directions. The thickness of normal metal $d_N$ was 1.6 μm with the exception of J-2 with $d_N=1.2$ μm. The Josephson effect was measured by a SQUID picovoltmeter. The details of the measuring technique have been described in our previous paper[8].

Figure 2 shows the superconducting transition of the CeCu$_2$Si$_2$ polycrystal observed by ac susceptibility and resistance measurements. As the temperature is decreased, the resistance shows a gradual decrease and then drops to zero at $T_c \sim 0.72$ K. Below $T_c$ the diamagnetic susceptibility appears, rapidly increases and then shows a perfect diamagnetism. The high $T_c$ value above 0.7 K ensures the good quality of the present sample.

The temperature dependence of critical current density $J_c$ is shown for the six junctions in Fig. 3, where $J_c$ is the critical current per unit area of the junction. The inset is the Shapiro steps appeared in the $I - V$ curve, when ac current is superposed on dc current. Since $hν/2e$ is 20 pV at a frequency of 10 kHz, the observed steps indicate that the Josephson current flows by the lowest-order (second-order) tunneling process.

Although CeCu$_2$Si$_2$ is superconducting below $T_c \sim 0.7$ K, the Josephson current becomes measurable well below $T_c$ and increases exponentially at low temperatures, because the use of the "dirty" normal layer Cu(Al) decreases the Josephson coupling between the two superconductors. The weak coupling simplifies the analysis of the Josephson effect as follows.
If the Josephson penetration depth $\lambda_J$, which decreases with an increase in $J_c$, becomes smaller than the width $w$ of the junction, the junction is self-field limited; the current distribution is not uniform and the effective current carrying width becomes of the order of $\lambda_J$. In the present junction, $\lambda_J$ is about 0.5 mm even for $J_c \sim 0.06$ A/cm$^2$; $\lambda_J$ is larger than the junction width and the current flow is uniform throughout the junction area. This ensures that the present results reflect the overall junction area.

The magnetic field dependence of $I_c$ indicates the quality of the junctions, as shown in Fig. 4. If we refer to a coordinate system with the $z$ axis along the current direction and apply an external magnetic field in the $y$ direction, the total Josephson current $I$ through the junction is given by

$$I = \int \int dxdy J_c(x, y) \sin\left(\frac{2\pi d}{\Phi_0} Hx + \psi_0\right),$$

(1)

where $J_c(x, y)$ is the local Josephson critical current density, $\Phi_0$ is the flux quantum, and $\psi_0$ is the phase difference between two superconductors; $d$ is given by $d = d_N + \lambda_S + \lambda_{S'}$, where $\lambda_S$ and $\lambda_{S'}$ are the penetration depth of the superconductors $S$ and $S'$, respectively. In case that the junction is uniform, that is, $J_c(x, y)$ is a constant, the maximum value $I_c$ of $I$ shows a Fraunhofer diffraction pattern;

$$I_c(H) = I_{c0} \left| \sin\left(\frac{\pi H/\Delta H}{\pi H/\Delta H}\right) \right|,$$

(2)

where $I_{c0}$ and $\Delta H$ are constants and $\Delta H$ corresponds to a flux quantum threading through the junction. Even if $J_c(x, y)$ varies spatially, $I$ shows a global maximum $I_c = \int \int dxdy J_c(x, y)$ at zero magnetic field, unless the phase difference also varies spatially.

The solid lines in the figures indicate the Fraunhofer pattern calculated using the global maximum value of $I_c$ as $I_{c0}$ and the $H$ interval between the global maximum and the nearest local minimum as $\Delta H$. The fact that J-1 shows a better fit to the pattern indicates that the Josephson current flows more uniformly as compared with J-2. The other four junctions also showed a modified Fraunhofer pattern like J-2; a global maximum of $I_c$ near $H = 0$ was commonly observed. Similar Fraunhofer-like patterns have already been reported for point contacts on CeCu$_2$Si$_2$, although the size and shape of the junction area that affect $I_c(H)$ patterns were not clear[6].
If the spatial variation of the phase difference \( \tilde{\psi}(x, y) \) exists \((\psi_0 \rightarrow \psi_0 + \tilde{\psi}(x, y))\), the peak at zero magnetic field is expected to become less dominant. Such a magnetic field dependence has been reported for the \( d_{x^2-y^2} \) superconductors; an oscillating pattern is observed for the junctions on the (110) face of YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\)[10] and on the (001) face of CeCoIn\(_5\)[11], because the Josephson current from + and - lobes of the order parameter should cancel out for [110] and [001] directions, while facets, pits and projections on the surface cause a contribution of the Josephson current reflecting a change of the relative sign of the order parameter \((\tilde{\psi}(x, y) = 0 \text{ or } \pi)\).

If CeCu\(_2\)Si\(_2\) is also a \( d_{x^2-y^2} \) superconductor, an oscillating \( I_c(H) \) pattern should be observed, as long as the junction area contain crystallites that face both + and - lobes of the order parameter to the surface. Although the distribution of the crystallographic orientation for each junction area was not determined, the junction area was chosen so that it may consist of crystallites in various directions, as shown in Fig. 1(b). Since such a microstructure leads to the contributions of different crystallographic orientations, the Fraunhofer-like pattern in the present investigation apparently contradicts the \( d_{x^2-y^2} \) superconductivity. One possible explanation is a surface-induced s-wave component[12]. Although such a component also explains the large \( I_c \) values observed for CeCu\(_2\)Si\(_2\)[6, 7], it will be a future issue why CeCu\(_2\)Si\(_2\) behaves differently from other \( d_{x^2-y^2} \) superconductors. It should be noted that unlike CeCoIn\(_5\) CeIrIn\(_5\) has also shown such an s-wave-like behavior[11, 13].

In conclusion, we have investigated the Josephson critical current \( I_c \) between CeCu\(_2\)Si\(_2\) and a conventional superconductor by fabricating junctions on a CeCu\(_2\)Si\(_2\) polycrystal. The magnetic field dependence of \( I_c \) has shown that a global maximum at zero magnetic field is dominant as compared with other peaks. This result may suggest the existence of an s-wave component on the surface of the multiple crystallites.

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References
[1] Steglich F, Aarts J, Broedl C D, Lieke W, Meschede D, Franz W and Schäfer H 1979 Phys. Rev. Lett. 43 1892
[2] Eremin I, Zwicknagl G, Thalmeier P and Fulde P 2008 Phys. Rev. Lett. 101 187001
[3] Lengyel E, Nicklas M, Jeevan H S, Sparn G, Geibel C, Steglich F, Yoshioka Y and Miyake K 2009 Phys. Rev. B 80 140513
[4] Yuan H Q, Grosche F M, Deppe M, Geibel C, Sparn G and Steglich F 2003 Science 302 2104
[5] Wollman D A, Van Harlingen D J, Giapintzakis J and Ginsberg D M 1995 Phys. Rev. Lett. 74 797
[6] Poppe U 1985 J. Magn. Magn. Mater. 52 157
[7] Koyama T, Sumiyama A, Nakagawa M and Oda Y 1998 J. Phys. Soc. Jpn. 67 1797
[8] Sumiyama A, Hata R, Oda Y, Kinura N, Yamamoto E, Haga Y and Ônuki Y 2005 Phys. Rev. B 72 174507
[9] Barone A and Paternò G 1982 Physics and Applications of the Josephson Effect (New York: John Wiley and Sons) p 71
[10] Néils W K and Van Harlingen D J 2002 Phys. Rev. Lett. 88 047001
[11] Sumiyama A, Tsuji Y, Ikeda N, Oda Y, Shishido H, Settai R and Ônuki Y to be published in Physica C
[12] Mößle M and Kleiner R 1999 Phys. Rev. B 59 4486
[13] Sumiyama A, Tsuji Y, Oda Y, Shishido H, Settai R and Ônuki Y 2007 J. Mag. Mag. Mater. 310 599