Anisotropic Josephson Effects in Point Contacts between the Heavy Fermion Superconductor URu_2Si_2 and Nb

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Point contacts between the heavy-fermion superconductor URu_2Si_2 and Nb are studied. A finite dc Josephson current is found in contacts aligned parallel to the *a-b* directions of URu_2Si_2, whereas it is absent in contacts aligned along the *c* direction. We attribute this extreme anisotropy of the Josephson current to an unconventional superconducting order parameter in URu_2Si_2, with a symmetry leading to destructive interference for Josephson currents along the *c* direction. [S0031-9007(98)06690-3]

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Several classes of new superconducting materials such as the high-T_c and the heavy-fermion superconductors (HFS) are believed to exhibit "unconventional" superconductivity [1,2]. A superconducting order parameter (OP) is denoted as unconventional if, below the transition temperature T_c, additional symmetries are broken besides gauge symmetry. In such a case the OP will, in general, show strong anisotropy, i.e., the magnitude and the phase of the OP vary over the Fermi surface [3]. Considerable interest in unconventional superconductors arises due to their unusual superconducting properties, such as, e.g., the existence of multiple superconducting phases; moreover, a pairing mechanism different from the conventional electron phonon mechanism is likely to be active.

Significant progress in the experimental identification of unconventional superconductivity has been made in the past few years, when it has been realized that direct information on the symmetry of the OP can be obtained from experiments sensitive to the phase of the OP [4]. For example, the Josephson current between two superconductors depends on the phase difference between the superconductors and is thus sensitive to the variation of the phase of the OP in an unconventional superconductor. In the high-T_c superconductor experiments on SQUIDS, interference patterns obtained on single Josephson junctions, the observation of strongly anisotropic Josephson currents as well as Andreev bound states at the surface have provided strong evidence for unconventional superconductivity. An OP with *d*-wave symmetry appears to be established now in these materials [2,5,6].

In heavy-fermion superconductors a variety of indirect experimental evidence exists in favor of an unconventional OP such as, e.g., the observation of multiple superconducting phases in UPt_3 [7] and power-law behavior in the temperature dependence of various physical properties such as the specific heat [3]. Also, the anisotropy of Andreev scattering in normal/superconductor point contacts has provided evidence for an unconventional OP [8]. In contrast, studies of Josephson effects have been reported only rarely, since it has turned out to be extremely difficult to establish Josephson contacts involving heavy-fermion superconductors [9–12]. Accordingly, no direct evidence for an unconventional OP from phase-sensitive experiments has been reported up to now.

We present in this Letter an experimental study of point contacts between Nb and the heavy-fermion superconductor URu_2Si_2. Our main result is that a Josephson current below the transition temperature of URu_2Si_2 is observed in contacts aligned along the *a-b* direction of the tetragonal structure, whereas it is absent in contacts aligned along the *c* direction. Such extreme anisotropy is very unusual in a *metallic* point contact and provides strong evidence for an unconventional OP in URu_2Si_2 with a symmetry such that the Josephson current along the *c* direction is zero because of destructive interference of the currents averaged over the various directions. There are several OP symmetries compatible with this requirement, e.g., the A_{2g}, B_{1g}, or B_{2g} states (see Refs. [13,14]). Odd-parity OP symmetries are excluded because of the large critical current found in our experiments.

The single crystals of URu_2Si_2 used in this study were prepared by a traveling zone flux melting technique [15]. Their superconducting transition occurs at $T_c \simeq 1.3$ K. Point contacts were fabricated by pressing etched Nb needles onto the surface of the single crystals. We obtained good contacts only with surfaces made by cleaving or breaking the single crystals; point contacts on polished surfaces were not superconducting. Contacts along the crystallographic *c* direction were well defined because the samples could be easily cleaved perpendicular to the *c* direction. In contrast, along other directions the URu_2Si_2 samples rather break than cleave. Accordingly, the surfaces were significantly less smooth in such cases and the direction of a point contact could be controlled only roughly. The measurements were carried out in a dilution refrigerator between 0.05 and 9 K. The point

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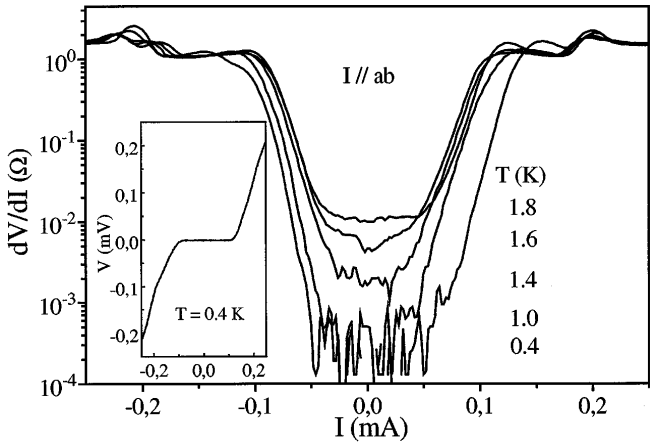

FIG. 1. Differential resistance dV/dI of a point contact between URu_2Si_2 and Nb versus bias current I at various fixed temperatures given in the figure. The current was applied parallel to the a-b direction. Inset: the same data at $T \simeq 0.4$ K shown in a plot of V versus I.

contacts could be adjusted *in situ* at low temperatures using a differential-screw mechanism. We measured I/V and dV/dI versus I characteristics of the contacts, where I is the applied bias current and V is the voltage drop over the contact. The normal state resistance R_N of the contacts varied between 0.1 and 30 Ω. From the Wexler formula [16], which relates R_N to the diameter of the contacts, we obtain a diameter of 100 nm for $R_N = 1\ \Omega$, so that the diameters of our contacts vary between 10 nm and 1 μm.

We show in Fig. 1 a point-contact characteristic obtained for a contact aligned along the a-b direction (denoted as a-b contact in the following). It shows a pronounced structure at about 0.1 mA, which appears below the superconducting transition temperature of Nb of $T_c \simeq 9.2$ K. Below the superconducting transition temperature of URu_2Si_2 the contact resistance drops again at low bias and becomes zero within our experimental resolution (see inset Fig. 1), while, in contrast to the behavior of contacts with normal-metal counterelectrodes [17–19], the shape of the characteristic does not change significantly. The transition to zero resistance below $T_c \simeq 1.3$ K of URu_2Si_2 is evident from the data shown in Fig. 2, where the zero bias resistance R_0 is shown as a function of temperature.

We show in Fig. 3 the differential resistance dV/dI measured at various fixed temperatures and at a bias current close to the critical current as a function of an applied magnetic field. We clearly observe an interference pattern, which confirms the presence of a Josephson current at low bias. The interference pattern is observed only below the superconducting transition of URu_2Si_2, which confirms that bulk superconductivity of URu_2Si_2 is involved in the Josephson effect; we are not studying a "proximity-induced" Josephson effect [20].

We show the temperature dependence of the critical current I_c in Fig. 2. Here I_c is defined as the current corresponding to a resistance of 1 mΩ. At the lowest measured temperature the product of I_c with the normal state resistance $R_N^{ab} \simeq 2$–$3\ \Omega$ (see Fig. 1) of the a-b contact is of order $I_c R_N^{ab} \simeq 150\ \mu$eV. This is somewhat but not drastically reduced compared to the value of 600 μeV obtained from the Ambegaokar-Baratoff formula [21]. We note that the values of $I_c R_N^{ab}$ scatter significantly from contact to contact and are in the range between 1 and 150 μeV. No systematic dependence on the contact resistance was observed, which indicates that it is probably the rather uncontrollable microscopic structure of the contact which determines the critical current.

The contacts obtained for the c direction (denoted as c contacts in the following) show a completely different behavior. An example is shown in Fig. 4. Whereas a structure of dV/dI occurs at about 0.1 mA, similar to the a-b contacts, *no indication* of an additional structure at and below the superconducting transition temperature of URu_2Si_2 was observed in all contacts studied. In particular, dV/dI is always finite at low bias currents. We should note that at very large bias currents additional structures may occur, which can be attributed to heating

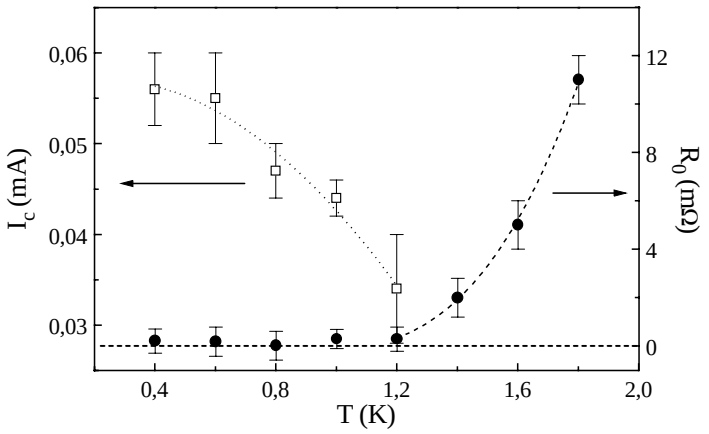

FIG. 2. Zero bias resistance R_0 versus temperature for the a-b contacts of Fig. 1 (solid symbols, right scale) and critical current (open symbols, left scale) as a function of temperature.

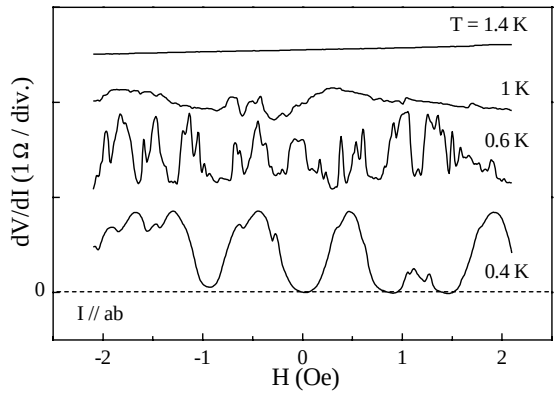

FIG. 3. Differential resistance dV/dI of a point contact between URu_2Si_2 and Nb measured at various fixed temperatures given in the figure versus a magnetic field H applied perpendicular to the contact. The bias current of 0.1 mA was applied along the a-b direction.

effects and are unimportant for the present study. Note, however, that R_N^c should not be determined from a region where heating is important.

Results such as those shown in Figs. 1–3 could be reproduced on several point contacts between URu_2Si_2 and Nb [22]. In particular, we used ten different samples and studied more than 20 a-b and c contacts, each by carefully measuring the temperature and magnetic field dependence of their I-V characteristics. For the c direction even more contact settings have been investigated. Superconductivity never occurred for the c contacts. In contrast, among the a-b contacts superconductivity—including a Josephson interference pattern—has been observed for six times. The general features of the I-V characteristics are well reproducible. Only properties such as the absolute value of $I_cR_N^{ab}$ depend on the particular contact. Therefore we conclude that the extremely anisotropic Josephson current found here is a characteristic and intrinsic feature of URu_2Si_2 contacts against a conventional superconductor.

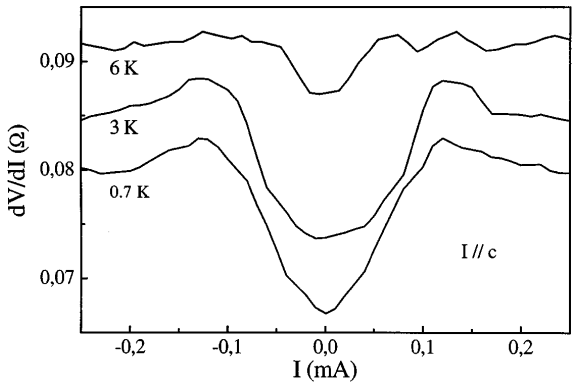

FIG. 4. Differential resistance dV/dI of a point contact between URu_2Si_2 and Nb versus bias current I at various fixed temperatures given in the figure. The current was applied along the c direction.

The current spreading in a metallic point contact is presumably nearly spherical, in particular, in materials with weak resistivity anisotropy, in contrast to tunneling currents, which are strongly peaked in the forward direction because of the exponential dependence on the tunneling barrier. Therefore point-contact spectra do not usually show strong anisotropy so that the extreme anisotropy of the Josephson current found here is very unusual. An explanation therefore requires special circumstances. One scenario is the following: Consider an OP in URu_2Si_2 such that the Josephson current in the c direction,

$$I_C R_N \sim \Delta_{\mathrm{Nb}} \int dk_a dk_b \Delta_{\boldsymbol{k}} , \qquad (1)$$

averages to zero. This requires at least one line node in a plane perpendicular to the a-b plane, which separates regions with a phase difference of the OP of π (for an example, see Fig. 5). Assuming a symmetric current distribution, Eq. (1) then yields zero net Josephson current, since the contributions from regions with phase difference of π cancel each other. Thus, the absence of a Josephson current in the c direction found in our experiments gives strong evidence for an unconventional OP in URu_2Si_2. Note that the vanishing of the OP for the c direction without the destructive interference described above is not sufficient to explain the absence of the Josephson current in a metallic point contact due to the spherical distribution of the current [23].

We note that the cancellation of Josephson currents from different directions can only be complete if the net current in the c contacts flows into the c direction quite accurately. However, firstly, in our experiments the c direction is indeed very well defined, since large flat surfaces perpendicular to the c direction were obtained from cleaving. Secondly, minor deviations from the c direction should be irrelevant, since they correspond to a strongly reduced Josephson current, which might not be detectable within the experimental noise. In contrast to this, for the a-b contacts the current direction is rather poorly defined (with the exception that there cannot be much of the c direction). Via the same averaging as described above this should lead to a strong variation of the critical current in the a-b direction for such contacts, consistent with our experimental observations.

It is possible to put several constraints on the symmetry of the OP from our results. Firstly, the product of

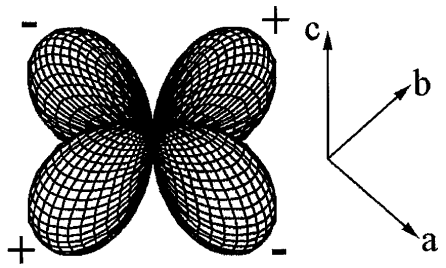

FIG. 5. A possible order parameter for URu_2Si_2 (B_{1g} symmetry). "+" and "−" refer to the phase of the OP.

$I_c R_N^{ab}$ is of the order of the Ambegaokar-Baratoff value. This favors clearly an OP with even parity, since in an odd-parity superconductor I_c is expected to be strongly reduced [3,24]. Secondly, the cancellation of Josephson currents along the c direction discussed above requires an OP which averages to zero in the a-b plane (e.g., with one line node in a plane perpendicular to the a-b plane with a phase difference of π). In principle, the A_{2g}, B_{1g}, or B_{2g} states are compatible with our results. An analysis of OPs allowed by the crystal symmetry has been given by Hasselbach *et al.* [14]. Among their proposals based on a comparison to specific-heat data, the even-parity B_{1g} symmetry (Fig. 5), which shows two line nodes and maximum gap values along the a and b axes, is in best agreement with our results. In contrast, Brison *et al.* [25] have proposed a model based on the interaction between superconductivity and antiferromagnetic order, which yields maximum gap values for the c direction. This model clearly disagrees with our results.

We finally discuss the structure occurring around 0.1 mA in both a-b and c contacts. This structure is most probably related to the superconductivity of Nb. Since the resistivity of URu_2Si_2 is much larger than that of Nb, it is, however, very unlikely that the contact resistance is determined by the resistance of Nb, even if one assumes strong disorder in the contact region. Therefore the large voltage drop indicates proximity-induced superconductivity in URu_2Si_2, as discussed in Ref. [11]. Taking dV/dI slightly above this structure as an estimate of R_N, we find $R_N^{ab} \approx 1\ \Omega$ for the a-b contact and $R_N^c \approx 0.1\ \Omega$ for the c contact. Consequently, the voltage scale for the a-b structure is by about a factor of 10 larger than that of the c structure. Note that this is consistent with the interpretation in terms of proximity-induced superconductivity, since the proximity effect should be more pronounced for the a-b contacts due to the larger coherence length in the a-b direction [26].

In summary, our data show that in point contacts between URu_2Si_2 and Nb a finite dc Josephson effect occurs only in contacts parallel to the a-b direction and is absent in contacts along the c direction. A straightforward explanation of this extreme anisotropy of the Josephson current in a metallic point contact is possible in terms of an unconventional order parameter in URu_2Si_2 with a symmetry leading to destructive interference for Josephson currents along the c direction.

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