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Observation of Negative *s*-Wave Proximity Effect in Superconducting UBe_{13}

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The Josephson I_c between a Ta-wire probe and an induced, surface, singlet, superconducting state in UBe_{13} decreases with decreasing temperature below the bulk UBe_{13} T_c, in contrast to the increase seen in comparison Mo samples. This shows that the bulk UBe_{13} superconductivity suppresses the induced singlet superconductivity. Such suppression is evidence of a triplet superconducting state in UBe_{13}. Evidence is presented for phase slip between weakly coupled singlet and triplet order parameters.

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The nature of the superconducting state in the heavy-fermion metal UBe_{13}[1] is a question which has generated considerable current interest and experimental activity. The resistivity and specific heat,[1] ultrasonic attenuation,[2] and other properties[3] in UBe_{13} are anomalous and have supported suggestions that its pairing is odd-parity (OP) spin-triplet,[4–6] as in superfluid ^{3}He, or even-parity *d* wave.[5] Several authors have proposed experiments to detect a characteristic "negative proximity effect" between conventional and OP superconductors.[7–9]

Measurements of the Josephson I_c[7,10] and the quasiparticle tunneling spectrum[11] are also pertinent. Interpretation of such experiments is complicated by inherent surface breaking of OP pairs and by the effects of spin-orbit interaction near an interface.[6] It has also been proposed that order parameters of different symmetry will weakly suppress each other by competing for phase space.[8] In this Letter we describe an experiment on UBe_{13} which gives evidence of such suppression, and we argue that this suppression indicates that the superconductivity in UBe_{13} is odd parity.

We have previously reported[12] *s*-wave superconductivity induced in the surface of UBe_{13} above its T_c by exchange of pairs from an *s*-wave probe and observed by the Josephson effect. A surface, singlet, order parameter Δ_s is thus established, extending a coherence length ξ into the bulk. This singlet state, whose magnitude can be monitored by the Josephson I_c, represents a probe of the bulk order parameter as in the proposed proximity-effect experiments.

New comparative measurements of the dc and ac Josephson effects have been carried out on ingots of UBe_{13} and Mo contacted by Ta wires. The samples are mounted outside a hole in the wide face of a *K*-band microwave guide, and the wire driven across its interior by an externally controlled screw. Four wires separately contact the ingot and the Ta wire. Contact is made at 4.2 K, with only millivolt bias. The apparatus can discriminate between a milliohm resistance and a short; the latter is observed on Mo below 0.92 K. Electropolished surfaces of UBe_{13} give significantly clearer Shapiro steps than were obtained previously.[12] Nb and Ta tips on 1-mm wire are mechanically ground and result in typical contact diameters of 1–10 μm. Study of the surface region of UBe_{13} contacted by the probe in a scanning Auger microprobe (microscope) reveals no damage.

Figure 1 shows the $I_c(T)$ curves. As previously re-

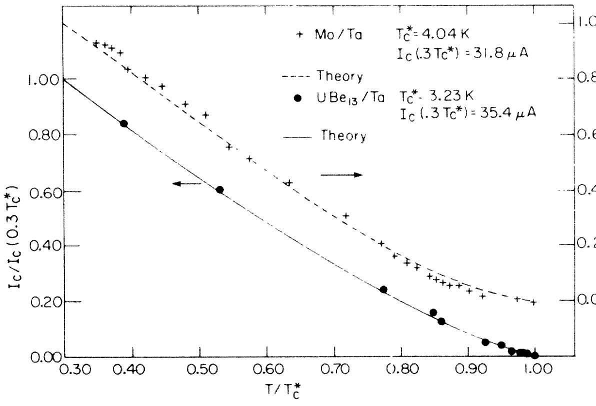

FIG. 1. High-temperature Josephson $I_c(T)$ for Mo-Ta (crosses) and UBe_{13}-Ta (filled circles) contacts. Proximity-induced T_c^* values are 4.04 and 3.32 K; lines are derived from Ginzburg-Landau model of Refs. 13 and 14, which is a more accurate treatment for $T > T_c^T$ than the model which yields Eq. (1) in text.

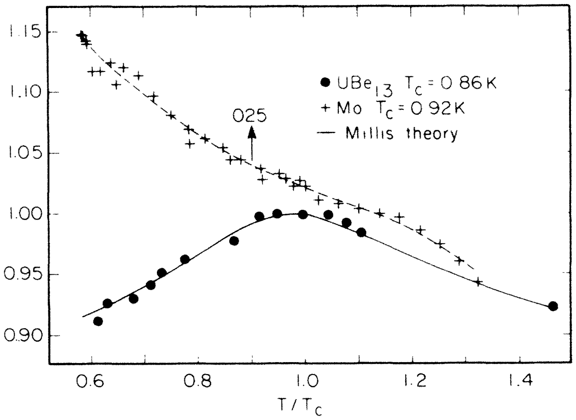

FIG. 2. Low-temperature $I_c(T)$ for nonhysteretic junctions of Fig. 1. T is normalized to ingot T_c. Fall of $I_c(T)$ in UBe$_{13}$ contact indicates s-wave pair breaking by the bulk superconductivity. Dashed curve is a guide to the eye, while solid curve is obtained from Eq. (3), with $\lambda = 2.8$, on the assumption $I_c \propto \Delta_s \Delta_{\rm Ta}$.

ported, the T_c of the contact, T_c^*, exceeds the bulk T_c and approaches that of the Ta wire, 4.47 K. A proximity-induced Josephson effect, characterized by a series-spreading resistance $R_S = \rho/2a$, with a the contact radius and ρ the bulk resistivity, is observed. The $I_c(T)$ curves are reasonably approximated by a Ginzburg-Landau model[13,14] (solid and dashed curves) assuming $I_c = \text{const} \times \Delta_s \Delta_{\rm Ta}$, where Δ_s and $\Delta_{\rm Ta}$ are the pair potentials at the surface of the ingot (UBe$_{13}$ or Mo) and the Ta, respectively.

The low-temperature $I_c(T)$ data are shown in Fig. 2. The Mo I_c rises faster below T_c corresponding to the appearance of an intrinsic pair potential $\Delta_{\rm Mo}$, resulting in an increase in Δ_s. $I_c(T)$ for UBe$_{13}$ *falls* below T_c, indicating suppression of Δ_s by about 10% between T_c and $0.6T_c$, and also the absence of direct Josephson coupling between the bulk UBe$_{13}$ order parameter and the Ta probe. All I-V curves are nonhysteretic. In Fig. 2, the solid curve is obtained from a model (below) based on a triplet bulk order parameter for UBe$_{13}$.

Figure 3 illustrates the effect of microwave irradiation on the I-V curves of the UBe$_{13}$-Ta contact at 0.51 K. The spacing of the Shapiro steps is accurately $h\nu/2e$ as expected for an induced-singlet state. The steps have been carefully observed and do not change as T crosses T_c. The data thus confirm that the surface singlet state Δ_s persists to $0.6T_c$ and is weakly suppressed as the distinct interior pair potential Δ_T develops.

The measured series residual resistances $R(T) = dV/dI|_{V=0,T}$ are plotted in Fig. 4. The expected behavior for a conventional superconductor (crosses) is $R = \rho/2a$ for $T > T_c$, and $R = 0$ for $T < T_c$, as singlet superconductivity expands from radius a to fill the sample. The crosses in Fig. 4 actually represent $R(T)$ measured on a Nb-Ta contact,[15] but are believed representative also of the Ta-Mo contact.

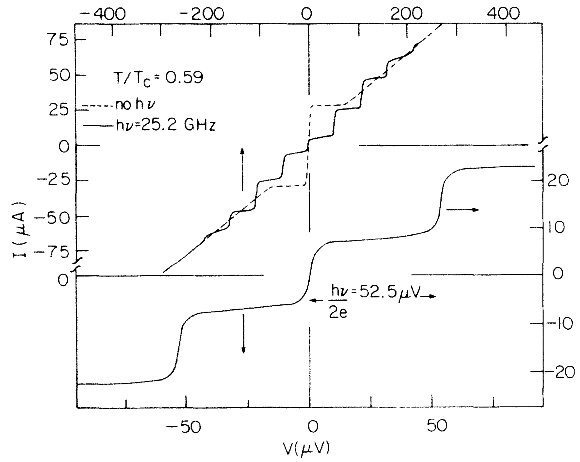

FIG. 3. Conventional Shapiro steps observed in UBe$_{13}$-Ta contact at 0.51 K, indicating Josephson effect between induced-s-wave state Δ_s and $\Delta_{\rm Ta}$. Direct coupling between $\Delta_{\rm Ta}$ and intrinsic UBe$_{13}$ pairing is absent, as seen from detail of steps in bottom panel. This is consistent with different parity (triplet) bulk order Δ_T in UBe$_{13}$.

The anomalous behavior of UBe$_{13}$ is shown by the solid circles in Fig. 4 which represent $R(T) = dV/dI|_{0,T}$ of the UBe$_{13}$-Ta contact, normalized by R^*

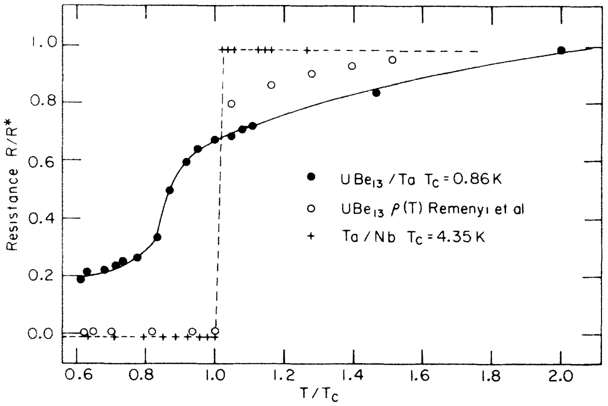

FIG. 4. Residual junction resistance $R(T) \equiv dV/dI|_{V=0,T}$ normalized by $R^* \equiv dV/dI|_{0,2\,\rm K}$ follows dashed curve (disappears at T_c) for induced-s-wave state on s-wave bulk superconductor. Anomalous behavior of UBe$_{13}$-Ta contact (filled circles, solid line) follows trend of UBe$_{13}$ bulk $\rho(T)$ (open circles, Ref. 16) above T_c, but nonzero R persists to $0.6T_c$. Orthogonality of singlet and triplet order parameters overlapping in a region of volume $\sim a^2\xi$ implies weak interaction, and allows phase-slippage between Δ_T and Δ_s.

$=dV/dI|_{0,2\,\mathrm{K}}$. The residual resistance at 2 K is $R^* = 1\ \Omega = \rho/2a$, which, using $\rho(2\ \mathrm{K}) = 200\ \mu\Omega$-cm, implies $a = 1\ \mu$m. $R(T)$ falls in a manner similar to the bulk resistivity (open circles), after recent measurements of Remenyi *et al.*[16] $R(T)$ below T_c drops more sharply, but returns to a reduced slope and reaches $0.2R^*$ at the lowest temperature, 0.5 K.

The observations on $\mathrm{UBe_{13}}$ may be understood if the bulk superconductivity in $\mathrm{UBe_{13}}$ is odd parity.[17] Then one expects, even near an interface, the following: (1) the direct Josephson coupling between tantalum and the odd-parity superconductivity is negligible[7–10]; (2) the odd-parity bulk superconductivity competes with the proximity-induced singlet superconductivity for electrons,[8] thus causing the magnitude of the proximity-induced singlet order parameter Δ_s to decrease as the temperature decreases below the T_c of $\mathrm{UBe_{13}}$; (3) phase slippage occurs in the region where the induced and bulk order parameters overlap, leading to a finite resistance in series with the Josephson junction even though the $\mathrm{UBe_{13}}$ is superconducting in bulk.

A complete discussion of these effects would involve formulating and solving a nonlocal, nonlinear equation. However, the essential question is the validity of point (2) above, and this may be demonstrated within the same simple model used for the $T > T_c$ data.[12]

We assume that the magnitude of the induced-singlet order parameter is fixed by balancing two energies: the free-energy cost δF_1 to impose *s*-type superconductivity on the $\mathrm{UBe_{13}}$ and another energy δF_2 which represents coupling to the Ta wire. We have

$$\delta F_1 = \Delta_s^2 \xi \left[\ln\frac{T}{T_c^s} + \left(\frac{\xi_0}{\xi}\right)^2 + \lambda \frac{\Delta_T^2}{(\pi k_\mathrm{B} T_c^T)^2} \right], \tag{1}$$

$$\delta F_2 = \eta \xi_0 (\Delta_\mathrm{Ta} - \Delta_s)^2, \tag{2}$$

where δF_2 is phenomenological; η is a measure of the transmissivity of the interface; Δ_Ta and Δ_s are the order parameters at the interface in the Ta and $\mathrm{UBe_{13}}$, respectively; and ξ_0 is the coherence length in $\mathrm{UBe_{13}}$. For $T > T_c^T$, where $T_c^T = 0.86$ K is the bulk transition temperature of the $\mathrm{UBe_{13}}$, one has $\xi_0 = (D/T)^{1/2}$. For $T << T_c^T$, ξ_0 goes over to $\xi_0 = (D/\Delta_T)^{1/2}$. Here Δ_T is the magnitude of the triplet gap near the interface.

δF_1 is obtained[8] by following the standard derivation of the Landau-Ginzburg equation for *s*-type superconductivity, but assuming (for $T < T_c^T$) the presence of a triplet gap of magnitude Δ_T. One assumes $\Delta_s(z) = \Delta_s e^{-z/\xi}$, and computes the free energy, δF_1. The parameter $\lambda = \lambda_0(\Delta_T/\Delta_\infty)^2$, where λ_0 is approximately 3.5, depending slightly on the form of triplet state assumed.[8] Δ_T/Δ_∞ measures how much the triplet gap is suppressed from its bulk value Δ_∞ by the presence of the interface. $T_c^s < T_c^T$ is the singlet T_c that $\mathrm{UBe_{13}}$ would have if the triplet-pairing interaction $V_T = 0$. We assume $T_c^s > 0$, but this is not a crucial assumption.

Now ξ is chosen by minimizing δF_1. Then $\delta F_1 + \delta F_2$ is minimized with respect to Δ_s, yielding

$$\Delta_s = \eta \Delta_\mathrm{Ta} \Big/ \left\{ \eta + 2\left[\ln\frac{T}{T_c^s} + \lambda \left(\frac{\Delta_T}{\pi k_\mathrm{B} T_c^T} \right)^2 \right]^{1/2} \right\}. \tag{3}$$

Since Δ_Ta is essentially independent of temperature below 1 K, while $(\Delta_T/\pi k_\mathrm{B} T_c^T)^2 \sim (1 - T/T_c^T)$ for $T < T_c^T$ we find $d\Delta_s/dT \sim +(\lambda - 1)$ which can be positive below T_c^T if $\lambda > 1$. Thus, since the Josephson current is proportional to $\Delta_s \Delta_\mathrm{Ta}$ (for $\Delta_s << \Delta_\mathrm{Ta}$), we obtain our main result: The observed I_c suppression is due to the mechanism described in point (2) above if $\lambda > 1$. The solid curve in Fig. 2, obtained from Eq. (3) with $\lambda = 2.8$, provides the observed behavior.[18] The data for $\mathrm{UBe_{13}}$ from four other contacts (not shown) can also be fitted by Eq. (3), by use of the same material parameter λ, altering only the contact-transmissivity parameter η. Further work is in progress in improving and generalizing this treatment.

We note that were the superconductivity in $\mathrm{UBe_{13}}$ spin-singlet (e.g., "*d* wave") our analysis would not apply.[8] Direct Josephson coupling between the Ta wire and the bulk $\mathrm{UBe_{13}}$ superconductivity would be possible. Also, within $\sim \xi_0$ of the interface, i.e., the region where the presence of the barrier is expected to destroy rotational invariance, the linearized gap equation would couple *s*- and *d*-symmetry gap functions.[19] The suppression of the induced *s*-wave gap caused by the competition for electrons would therefore be much weaker. Therefore, one may obtain a suppression of I_c below the $\mathrm{UBe_{13}}$ T_c from a model assuming *d*-wave superconductivity only if[8] the Josephson coupling to the bulk *d*-wave order parameter is anomalously small, or if the induced-*s*-wave order parameter extends a distance $\xi >> \xi_0$ into the bulk. We therefore believe the superconductivity in $\mathrm{UBe_{13}}$ is odd parity.

In summary, a negative proximity effect has been observed between the bulk superconductivity of $\mathrm{UBe_{13}}$ and a proximity-induced surface singlet state. This effect has been accounted for by a model of triplet-singlet phase competition in $\mathrm{UBe_{13}}$ below its T_c. As we have argued, these new results support an odd-parity superconducting ground state in $\mathrm{UBe_{13}}$.

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[15] For high-purity annealed Mo, $R_s = \rho/2a$ (for $a = 1$–10 μm and residual resistance ratio 100) is below our resolution of 10^{-3} Ω. The Ta foil was cold rolled and had a larger ρ.

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[17] We will briefly consider below the possibility of an even-parity *d*-wave state.

[18] The theory curve (solid) in Fig. 2 has been smoothed to match the experimental temperature resolution.

[19] Note that the odd parity of the *triplet* gap function ensures that the linearized gap equation cannot couple singlet and triplet gap functions, even near the interface.