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Measured Temperature Dependence of the cosφ Conductance in Josephson Tunnel Junctions

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The temperature dependence of the cosφ conductance in Sn-O-Sn Josephson tunnel junctions has been measured just below the critical temperature, T_c. From the resonant microwave response at the junction plasma frequency as the temperature is decreased from T_c it is deduced that the amplitude of the cosφ term first increases from near zero to positive values and only at $T/T_c \lesssim 0.96$ attains values of order -1.

Observation of the phase-dependent conductance in Josephson junctions was first reported by Pedersen, Finnegan, and Langenberg.[1] It was found that the so-called cosφ term had a negative sign and a magnitude of order 1. This result has recently been confirmed by several authors.[2] According to the tunneling Hamiltonian calculation of the pair current, one would expect a positive sign.[3] Alternative theories introducing a finite relaxation time of the superconducting order parameter do generate a negative sign in agreement with the experiments.[4] Hence, it remains an unsettled question whether the cosφ term predicted by tunneling theory has ever been observed.

In this Letter we report the first measurement of the cosφ term as a function of temperature. The experiments are performed on superconduction-insulator-superconductor (S-I-S) tunnel junctions current biased at zero dc voltage and at temperature just below the critical temperature, T_c. We find that at the highest temperatures ($T/T_c > 0.98$) the cosφ term is in fact *positive* and that at slightly lower temperatures ($T/T_c \lesssim 0.96$) the sign has changed.

The result is deduced from the temperature dependence of the junction plasma resonance. We used two different schemes for detection of the plasma oscillations. Both were based on a measurement of the junction response at 9 GHz as a function of the dc-bias current: (a) detection of the junction-generated second-harmonic voltage at $2f_1$ with an applied signal at $f_1 \sim 4.5$ GHz; (b) detection of the junction response at the difference frequency $f_2 - f_1$ with two signals at $f_1 \sim 9$ GHz and $f_2 \sim 18$ GHz applied simultaneously.

We have interpreted the experimental results within the framework of the shunted-junction model. The total current flowing into the junction is expressed as a sum of four terms:

$$I_{\text{tot}} = C\dot{V} + V/R + \epsilon V \cos\varphi/R + I_c \sin\varphi, \qquad (1)$$

where C is the shunt capacitance and R is the shunt resistance. The third term is the cosφ conductance with amplitude ϵ relative to the phase-independent conductance term. I_c is the critical current, and V the voltage drop across the junction. The voltage drop across the junction. The voltage is related to the time derivative of the phase φ by

$$V = \hbar\dot{\varphi}/2e. \qquad (2)$$

It is not obvious *a priori* that the shunted-junction model is justified in the present experiment. Equation (1) with voltage-dependent coefficients $1/R$, ϵ, and I_c has been derived in the adiabatic approximation and by assuming constant voltage bias. Provided, however, that the junction voltage $V \ll \Delta/e$ and the applied frequencies $f \ll 2\Delta/h$ the voltage dependence may be neglected. This is in accordance with the approach successfully used by others including those of Refs. 1 and 2. With this in mind we accept the model as basically phenomenological.

From Eqs. (1) and (2) we obtain the differential equation determining the evolution in time of the phase:

$$I_{\text{tot}}/I_c = \ddot{\varphi}/(2\pi f_0)^2 + (1 + \epsilon\cos\varphi)\dot{\varphi}/(2\pi f_0 Q_0) + \sin\varphi, \qquad (3)$$

where we have introduced the maximum plasma frequency,

$$f_0 = (2eI_c/\hbar C)^{1/2}/2\pi, \qquad (4)$$

and the corresponding quality factor

$$Q_0 = 2\pi f_0 RC. \qquad (5)$$

In the present context we need the zero-dc-voltage solutions to Eq. (3) with applied currents of the forms

$$I_{\text{tot}} = I_0 + I_1 \sin(2\pi f_1 t + \theta_1) \qquad (6a)$$

and

$$I_{\text{tot}} = I_0 + I_1 \sin(2\pi f_1 + \theta_1) + I_2 \sin(2\pi f_2 t + \theta_2). \qquad (6b)$$

Case (6a) is solved for the second-harmonic voltage generated by the junction, assuming a solution to Eq. (3) of the form

$$\varphi(t)=\varphi_0+\varphi_1\sin(2\pi f_1 t+\psi_1)+\varphi_2\sin(4\pi f_1 t+\psi_2). \tag{7a}$$

The amplitude φ_2 is determined self-consistently to second order in φ_1 using harmonic-balance conditions. The voltage is then found from Eq. (2).

Case (6b) is solved in a similar way. Inserting into Eq. (3) a solution of the form,

$$\varphi(t)=\varphi_0+\varphi\sin(2\pi f t+\psi)+\varphi'\sin(2\pi f' t+\psi')+\varphi_1\sin(2\pi f_1 t+\psi_1)+\varphi_2\sin(2\pi f_2 t+\psi_2), \tag{7b}$$

which includes also a component at the sum frequency, $f'=f_1+f_2$. The voltage component at $f=f_2-f_1$ is found self-consistently to second order in φ_1 and φ_2.

To first order in φ_1, φ_2 the plasma frequency, f_p, depends on the dc-bias current, I_0, as

$$f_p=f_0[1-(I_0/I_c)^2]^{1/4};\quad I_0<I_c. \tag{8}$$

Further details of the straight forward but somewhat tedious calculations will be presented elsewhere.[5]

The experiments were made on overlap tunnel junctions with areas of about 0.2 mm×0.1 mm, normal-state resistances of typically 0.2 Ω, and maximum current densities of order 20 A/cm^2 (at $T=0$ K). The junction was well coupled to the input waveguide of the microwave receiver as previously described.[6] Microwave filters and isolators ensured that only pure monochromatic signals reached the junction and the receiver.

In order to observe the small second-harmonic voltages we used in scheme *A* an *X*-band interferometer with phase-sensitive detection at 70 MHz.[5]

A recording of two orthogonal components of the second-harmonic voltage at $2f_1=8.806$ GHz is shown in Fig. 1(a) as a function of dc-bias current: $I_0<I_c$.[7] With the second-harmonic technique useful recordings of the response could be obtained only in a narrow temperature range ($0.994\geqslant T/T_c\geqslant 0.982$). The upper limit was defined by the condition $f_1<f_0$, and the lower by the switching of the junction to the finite voltage state as the bias current corresponding to resonance approached I_c [Eq. (8)].

Measurements could be made in an additional narrow temperature range ($0.965\geqslant T/T_c\geqslant 0.930$) by doubling the plasma frequency (increasing I_c a factor of 4 by lowering the temperature). Here,

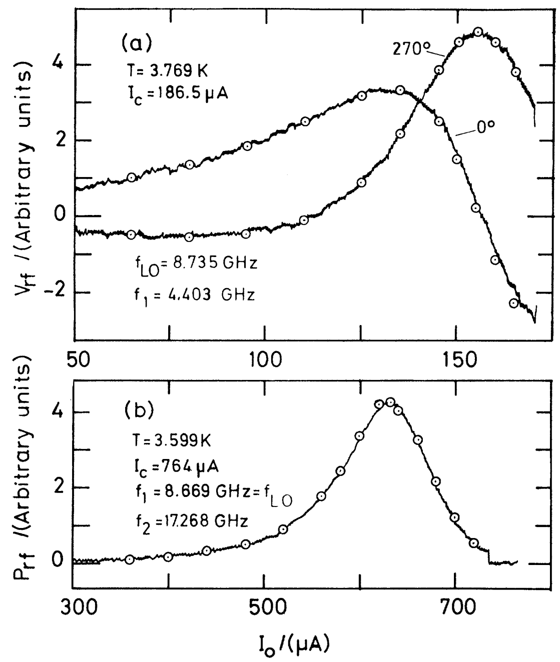

FIG. 1. *X-Y* recorder traces showing the junction plasma resonance response. (a) Two orthogonal components of the second-harmonic voltage at $2f_1=8.806$ GHz. The points are calculated with $f_0=6.381$ GHz, $Q_0=1.744$, and $\epsilon=+0.3$. (b) The microwave power at $f_2-f_1=8.599$ GHz. The points are calculated with $f_0=13.10$ GHz, $Q_0=1.832$, and $\epsilon=-0.8$.

a resonant response was obtained using scheme *B*.[8] The applied singal at $f_1=8.669$ GHz was derived from the local oscillator of the receiver ($f_1=f_{LO}$). The frequency $f_2=17.268$ GHz of the other input signal was adjusted such that $f=f_2-f_1=f_{LO}-f_{IF}$, where f_{IF} is the intermediate frequency of the receiver. A recording of the detected microwave power at the difference frequency $f=8.599$ GHz obtained using a standard superheterodyne spectrometer is shown in Fig. 1(b).

The points shown in Figs. 1(a) and 1(b) are calculated using values of the parameters f_0, Q_0, and ϵ adjusted to minimize the rms deviation between theory and experiment. It is important to point out, however, that this was not sufficient to determine all three parameters. In general it was possible for almost any value of ϵ in the interval ($-1\leqslant\epsilon\leqslant 1$) to fit the experimental trace (within experimental accuracy) by adjusting the parameters f_0 and Q_0. Accordingly, the $\cos\varphi$ amplitude, ϵ, could not be determined accurately without further knowledge of the temperature dependence of either f_0 or Q_0.[9]

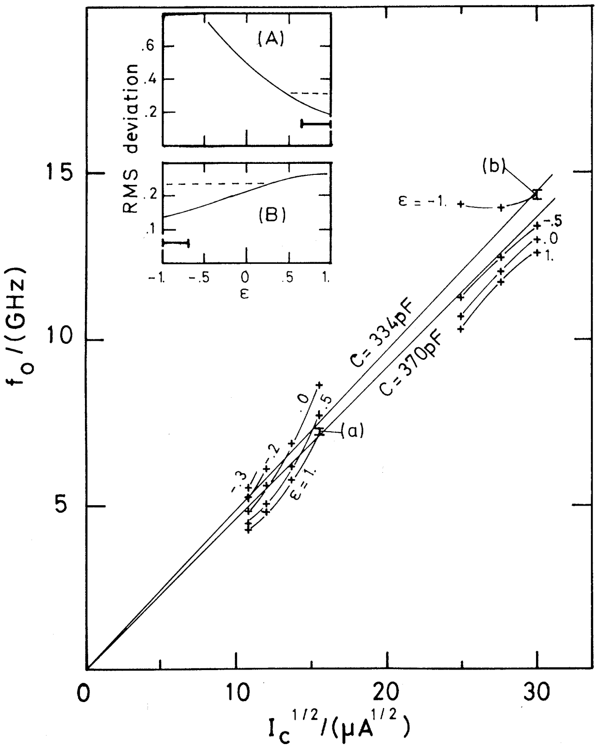

FIG. 2. Maximum plasma frequency, f_0, vs square root of critical current with ϵ as a parameter. The slope of the two straight lines drawn from the origin, determines the junction capacity: $C = 352 \pm 18$ pF. The insets show the rms deviations (minimized with respect to f_0 and Q_0) for (a) $I_c^{1/2} = 15.6\ \mu A^{1/2}$ and (b) $I_c^{1/2} = 30.1\ \mu A^{1/2}$. The dashed horizontal lines in the insets show the experimental uncertainty, and the horizontal bars the values of ϵ consistent with the measured capacity.

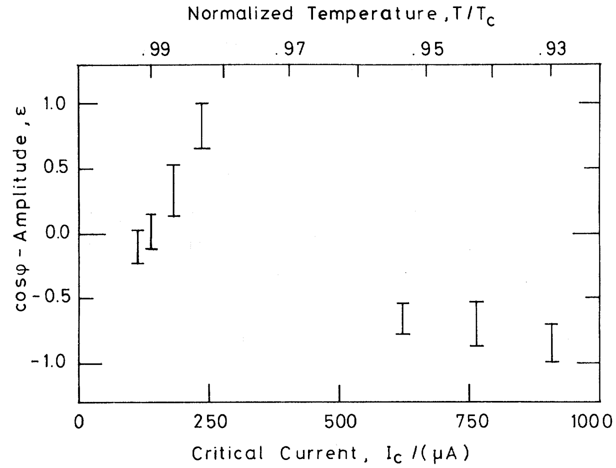

FIG. 3. The measured $\cos\varphi$ amplitude, ϵ, vs junction critical current or normalized temperature. $T_c = 3.815 \pm 0.0005$ K.

We have assumed that $f_0 \propto [I_c(T)]^{1/2}$, which is valid if C is independent of temperature.[10] A least-mean-square fit to each experimental trace was made by variation of f_0 and Q_0 for a number of ϵ values in the interval $-1 \leq \epsilon \leq 1$. Figure 2 shows corresponding values of f_0 and $I_c^{1/2}$ determined in this way with ϵ as a parameter. The points marked (a) and (b) in Fig. 2 and the condition $|\epsilon| < 1$ define the limiting values of $C = 334$ and 370 pF. From Eq. (4) we calculate the corresponding maximum plasma frequencies for each experimental trace, and finally, using the dependence $f_0 = f_0(\epsilon)$ (determined by our curve-fitting procedure) we determine ϵ.

The insets (A) and (B) in Fig. 2 show the rms deviation (minimized with respect to f_0 and Q_0) vs ϵ for the two crucial experimental traces at $I_c^{1/2} = 15.6\ \mu A^{1/2}$ and $I_c^{1/2} = 30.1\ \mu A^{1/2}$. The dashed horizontal lines in the insets indicate our experimental accuracy. It is clearly seen that the dependence of the rms deviation on ϵ is consistent with the value of ϵ determined from the capacity measurement (shown by the horizontal bars). This qualitative agreement was found for all the experimental traces.

The final result is shown in Fig. 3 where the $\cos\varphi$ amplitude, ϵ is plotted vs critical current and normalized temperature. We conclude that the $\cos\varphi$ amplitude is near zero at the highest temperatures and that it increases towards positive values as the temperature is decreased. This is in qualitative agreement with the tunneling Hamiltonian calculation. At still lower temperatures the $\cos\varphi$ amplitude has changed sign and approaches -1 in agreement with previously reported results.

The result strongly suggests that the phase-dependent conductance term may be explained only if competing processes in the tunneling structure are taken into account. We hope that our experimental result will encourage further theoretical work along these lines.

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[7]I_c was measured by recording about 100 I-V curves on a storage oscilloscope. We identified the largest value recorded with the critical current. The reproducibility of the procedure was better than 0.1% and comparable to the long-term stability of the He-bath temperature.

[8]A resonant response was also observed using scheme *A*. Because of the low Q of the plasma resonance in our junctions, this resonance ($f_p = 2f_1$) overlapped strongly with the wing of the $f_p = f_1$ resonance and was not clearly resolved.

[9]The Q may include surface losses in the superconducting films and may depend on the applied microwave power level. No estimate is attempted.

[10]The capacity depends only on geometry and barrier properties which are assumed independent of temperature in the narrow temperature range of the experiment.

Field-Dependent Magnetic Phase Transitions in Mixed-Valent TmSe

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A neutron diffraction study of the field-dependent magnetic ordering in TmSe is reported. The magnetic strucutre in zero field is antiferromagnetic fcc type I with $T_N = 3.2$ K. The magnetic phase diagram may be understood as a successive domain reorientation and metamagnetic transitions for $T < 3$ K with increasing field. We can explain this quantitatively using a simple classical spin Hamiltonian. The mixed-valent character manifests itself mainly in a reduced moment and in a markedly altered crystal field.

The magnetic properties of rare-earth mixed-valent compounds have been a subject of controversy for a number of years.[1,2] Until recently, it was generally believed that conventional magnetic ordering was not possible in mixed-valence materials if the fluctuation energy was large compared with the exchange energy. Varma,[1] however, has predicted that long-range magnetic order be possible provided that both rare-earth valence states are magnetic. The best-known example in which this situation is achieved is the rocksalt chalcogenide TmSe.[3] Early unpublished neutron diffraction experiments by Cox *et al.*[3] in a polycrystalline sample of TmSe indicated complex magnetic correlations but no true long-range order at 1 K. However, more recent macroscopic measurements,[4-8] including susceptibility, thermal expansion, elastic constants, and magnetostriction, on well-characterized single crystals indicate a well-defined phase transition at $T \simeq 3$ K. The magnetic-field dependence of the ordered state appears to be quite anomalous.[5-8] The nature of this magnetic state and its relationship to the mixed-valence fluctuations remains mysterious.

In order to clarify the magnetism of TmSe we have carried out a neutron diffraction study of the magnetic ordering on a well-characterized single crystal[9] of TmSe. We demonstrate that conventional long-range order is indeed achieved ($T_N = 3.2$ K in our sample) thus giving unambiguous configuration of Varma's prediction. The ordering is fcc type I antiferromagnetic (a/f). We show that in a magnetic field there is only one bona-fide phase boundary separating the a/f phase from the paramagnetic (para) phase. The magnetic properties of TmSe may, in fact, be simply understood using a classical spin Hamiltonian. The principal manifestations of mixed-valent effects seem to be a drastically altered