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Experiments with d-wave Superconductors

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Abstract

The predominant $d_{x^2-y^2}$-wave pairing-symmetry of most high-T_c superconductors provides the opportunity to fabricate Josephson junction circuits in which part of the junctions are biased by a phase difference of the superconducting order parameter of π. To explore the road to such π-electronics, we have fabricated and studied all-high-T_c dc superconducting quantum interference devices (dc SQUIDs) realized with thin film technology, of which the Josephson junctions consist of one standard junction and one junction with a π-phase shift.

These π-SQUIDs provide clear evidence of the $d_{x^2-y^2}$-wave symmetry of the order parameter, the amount of complex admixtures of other symmetry components being undetectably small. This seems to contradict other experiments, the results of which have been presented as evidence for an s-wave order parameter or for complex admixtures. Possible solutions to resolve this apparent contradiction are presented. In particular it is pointed out that even in the bulk of a superconductor the order parameter symmetry (the admixture of various symmetry components) may be spatially dependent.

The predominant $d_{x^2-y^2}$-symmetry of the order parameter of the high-T_c cuprates offers the possibility to fabricate electronic circuits in which the phase differences of selected Josephson junctions are in equilibrium biased by π [1–5]. For convenience, for the rest of the article such junctions will be termed π-junctions, with the understanding that it is not the microscopic transport across the junction, but the superconducting circuit consisting of the junction and, for example, a SQUID loop, which creates the π-phase shift. The π-junctions provide new opportunities for the design of high-T_c sensors and circuits. As suggested by Terzioglu and Beasley [6], π-junctions are useful for the fabrication of complementary Josephson junction circuits which are composed of standard SQUIDs and SQUIDs that contain a π-junction. These circuits are characterized by small power dissipation and large circuit margins. Likewise, circuits containing π-junctions have been used with outstanding success for analyses of the order parameter symmetry [3–5]. For practical reasons, it is desirable to realize such circuits in an all high-T_c thin film technology using standard growth processes. Therefore we aimed to fabricate thin film high-T_c dc SQUIDs that include one π-junction and one standard junction (see Fig. 1a) and to unambiguously verify their operation. These devices will be referred to as 'π-SQUIDs'. For symmetric π-SQUIDs with small inductance L, the phase shift across the π-junction causes a minimum of the magnetic field dependent SQUID critical current $I_c(H)$ at small applied magnetic fields H, as compared to the maximum shown at zero applied field by the $I_c(H)$-patterns of standard SQUIDs. It is pointed out that first π-SQUIDs have been built by Wollman, van Harlingen and collaborators, who contacted single crystals of $YBa_2Cu_3O_{7-\delta}$ with a conventional s-wave superconductor [3].

The design of the π-SQUIDs is illustrated in Fig. 1(a) [7]. It is based on two symmetric $45 \pm 1°$[001]-tilt grain boundaries fabricated with the bicrystal technology [8,9]. Due to the orientation of the grain boundaries, one of the junctions is a π-junction. For comparison, we also fabricated standard SQUIDs containing conventional Josephson junctions only, as sketched in Fig. 1(b).

The current-voltage, $I(V)$-characteristics of the π-SQUIDs and of the standard SQUIDs follow the behavior expected according to the resistively shunted junction (RSJ) model, with additional self-induced resonances (see Fig. 2). Clear differences between the π-SQUIDs and the standard SQUIDs are revealed by the magnetic field dependencies of the critical currents, which are shown in Fig. 3. Overall, the patterns of all devices show clear SQUID-modulations bounded by the envelopes given by the $I_c(H)$-dependencies of the grain boundary junctions. As is typical for 45° grain boundaries [5], the envelopes deviate significantly from a Fraunhofer pattern, with an unusual periodicity of the minima as well as large side maxima. For all devices, the

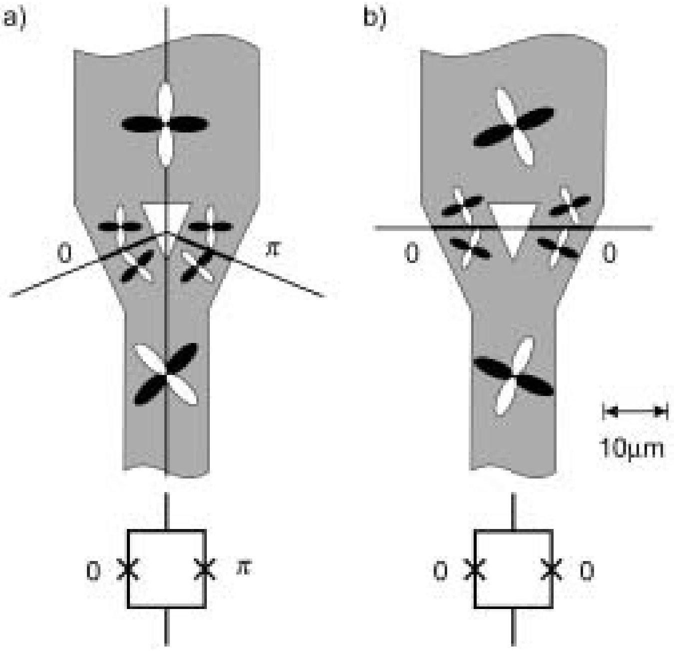

Fig. 1. Schematics of a) the π-SQUID and b) the standard SQUID investigated. For both SQUIDs, the junctions straddle symmetric 45° [001] tilt grain boundaries. As sketched by the vertical line, there is an additional boundary with a nominal misorientation of 0 present in the π-SQUID (from [7]).

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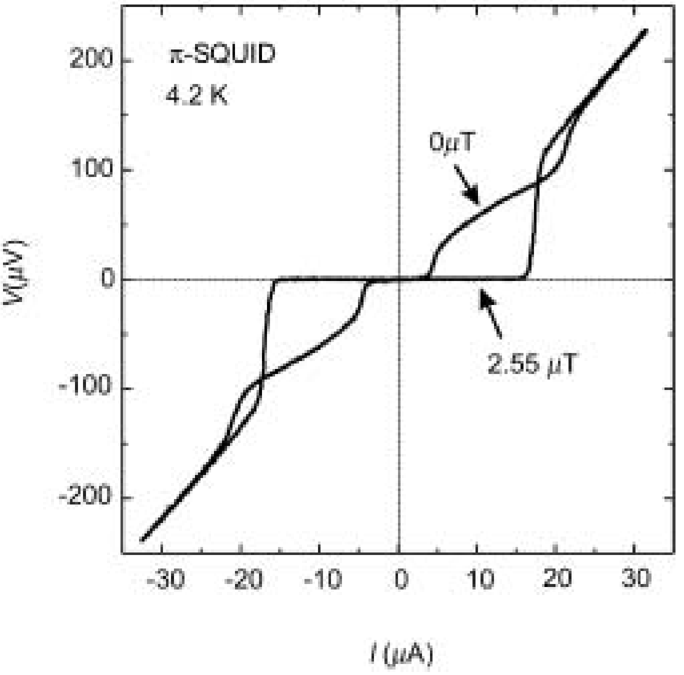

Fig. 2. $I(V)$-characteristic of a π-SQUID at 4.2 K. The resonances are caused by an order parameter symmetry induced half fluxoid oscillating at rf-frequencies (from [10]).

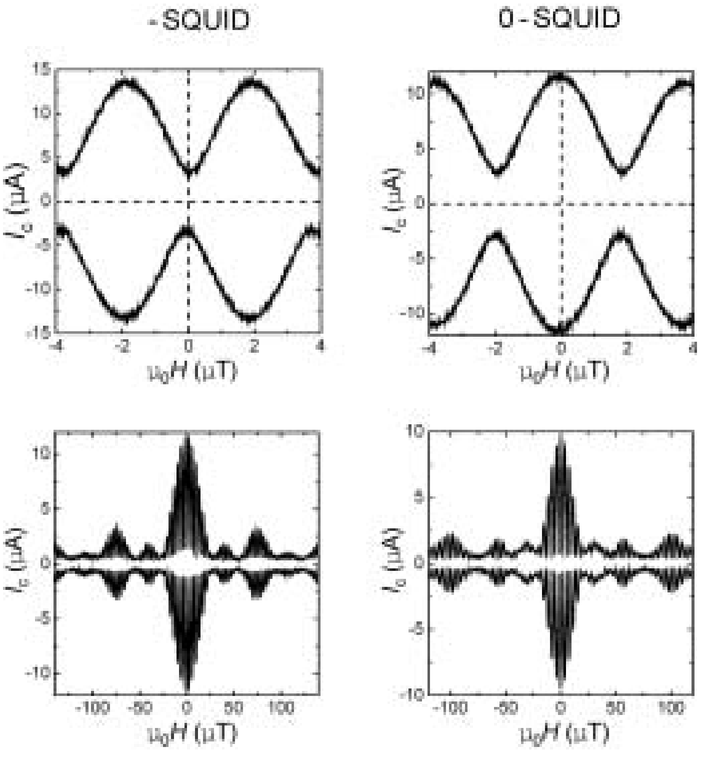

Fig. 3. Dependencies of the critical currents on applied magnetic field of the π-SQUID and a control SQUID measured at 77 K.

SQUID-oscillations and the envelopes are highly symmetric with respect to a small offset field $H_o < 0.2\,\mu T$. The symmetry of the envelopes and the small value of H_o prove that the influence of trapped magnetic flux and of magnetic background fields on the measurements are small. As expected, the $I_c(H)$-patterns of the π-SQUIDs show a minimum at small fields, exactly opposite to the behavior of the standard SQUID's critical current, which goes through a maximum. This effect demonstrates unambiguously that the π-SQUIDs operate correctly. By designing the π-SQUIDs and standard SQUIDs to have small critical currents and inductances, it has been possible to achieve $I_c(H)$-characteristics which are highly complementary.

By generating ac-currents and half magnetic fluxoids oscillating in polarity at tens of GHz, the unconventional pairing symmetry is also the cause for the self induced resonances. The two Josephsons junctions operate by design with a phase difference of π, which induces circulating currents in the SQUID loop. The sense of circulation changes sign with the Josephson frequency corresponding to the voltage across the π-SQUID. These oscillations are detected by their resonance with the LC-resonance of the SQUID loop and the junction capacities, causing the resonance steps of the $I(V)$-characteristics (Fig. 2) [10].

The observation of the $d_{x^2-y^2}$-wave symmetry in $YBa_2Cu_3O_{7-\delta}$ without any detectable complex admixtures agrees with many of the previous phase sensitive studies reported in literature. For example, the experiments by Wollman, van Harlingen, and coworkers [3] and by Brawner and Ott [11] on corner junctions and SQUID-structures fabricated from tunnel junctions between high-T_c compounds and conventional superconductors such as Pb or Nb revealed the presence of a strong $d_{x^2-y^2}$-component. The tricrystal experiments performed by Tsuei, Kirtley, and coworkers [4] gave conclusive evidence for the existence of a $d_{x^2-y^2}$-wave dominated order parameter in a wide variety of high-T_c cuprates. In particular, for $Tl_2Ba_2CuO_{6+\delta}$ films a pure $d_{x^2-y^2}$-wave symmetry was deduced [4]. A very strong $d_{x^2-y^2}$-wave dominated symmetry is also evidenced for $YBa_2Cu_3O_{7-\delta}$ by the characteristic dependence of asymmetric 45° grain boundaries [5].

For another series of experiments, however, in which the charge transport in c-direction has been investigated, different results have been reported in several cases. For such measurements performed on $YBa_2Cu_3O_{7-\delta}$, besides evidence for a $d_{x^2-y^2}$-wave symmetry [12,13], indications for a s-wave (see, e.g., [14]) or a mixed $d_{x^2-y^2}$- and s-wave pairing (see, e.g., [15,16]) were reported. For $Bi_2Sr_2CaCu_2O_{8+\delta}$ samples, the situation is even more confusing. Studies of current flow in c-direction across [001] twist boundaries in $Bi_2Sr_2CaCu_2O_{8+\delta}$ bicrystals were analyzed to reveal a small but finite critical current density, which is independent of the misorientation angle of the grain boundary [17,18]. Based on this finding, the existence of an s-wave or of a $d_{x^2-y^2}$-component of the order parameter has been discussed for $Bi_2Sr_2CaCu_2O_{8+\delta}$ [19] and it has even been argued that the $d_{x^2-y^2}$-component is vanishing [18]. Likewise, measuring a finite dc Josephson current flowing in the c-direction of $Bi_2Sr_2CaCu_2O_{8+\delta}$ /Pb junctions, Mößle and Kleiner report an s-wave component of the order parameter in $Bi_2Sr_2CaCu_2O_{8+\delta}$ [20]. The in-plane torque anisotropy of high-quality $Tl_2Ba_2CuO_{6+\delta}$ films has been measured by Willemin and coworkers, who are explaining their results by approximately equal ratios of s- and d-wave components [21].

Of the above mentioned experiments several have been analyzed quantitatively to derive the value ε of the admixture of an s-wave component to a $d_{x^2-y^2}$-wave dominated order parameter for a given compound: $\Psi = (1-\varepsilon)\Psi_{d_{x^2-y^2}} + \varepsilon i\Psi_s$. These analyses yielded a range of differing values for ε, and therefore no consensus on the admixture coefficients has been reached. Whereas some of the discrepancies may be attributed to experimental deficiencies, in the following the question will be addressed whether at all for the bulk of each superconductor the admixture of various symmetry components has to be described by a fixed number, as typically assumed in the interpretation of the experiments, or whether the admixture of various symmetry components may vary as a function of real space.

Variations of the pairing symmetry have already been discussed for two particular cases. First, it was noted by K.A. Müller that several of the apparently inconsistent experimental results can be accounted for by the possible existence of two superconducting order parameters in the high-T_c cuprates [22]. The two presumed order parameters, having the same transition temperature, are of s- and d-wave symmetry, respectively. As was pointed out in this work, in this case one may naturally expect that experiments probing the *ab*-direction yield a larger $d_{x^2-y^2}$-wave component than experiments relying on current flow in *c*-direction do. Second, the influence of interfaces and surfaces on the behavior of systems with one superconducting order parameter has been investigated theoretically by several groups. They find that by breaking the symmetry of the crystal lattice, interfaces in general cause a spatially dependent change of the admixture of symmetry components of the superconducting order parameter close to the interface [23,24].

However, even in the bulk of the superconductor, the admixture of various symmetry components of a superconducting order parameter does not have to be constant. In the general case, the mixing ratio of the symmetry components is a function of the position in the crystal lattice. To demonstrate this point, we introduce the model system shown in Fig. 4. It consists of two superconducting compounds C_A and C_B, which, as separated bulk materials, are characterized by order parameters Ψ_A and Ψ_B with $\{|\Psi_A|^2\} \gg \{|\Psi_B|^2\}$. Here the parentheses {} denote the spatial average. We consider the case that C_A is dominated by a $d_{x^2-y^2}$-wave, and C_B by an s-wave symmetry component: $\Psi_\alpha = (1-\varepsilon_\alpha)\Psi_{\alpha,d_{x^2-y^2}} + \varepsilon_\alpha i \Psi_{\alpha,s}$; $\alpha = A$ or B; $0 < \varepsilon_A \ll 10 \ll \varepsilon_B < 1$. The materials C_A and C_B form a superlattice in *c*-direction with layer thicknesses *d*, with $d \gg \xi$ (0 K), where ξ is the coherence length. For clarity it is assumed that the *c*-axis coherence lengths of C_A and C_B are equal at all temperatures $\xi_A(T) = \xi_B(T) = \xi(T)$. As the layers are closely coupled, the superconducting properties of the entire heterostructure are characterized by a new order parameter $\Psi(\rightleftharpoons)$. Several interesting features of such heterostructures are noted. At low temperatures, for example, the properties of tunnel junctions fabricated from these superlattices depend crucially on the direction of the surfaces used as electrodes of the tunnel junctions:

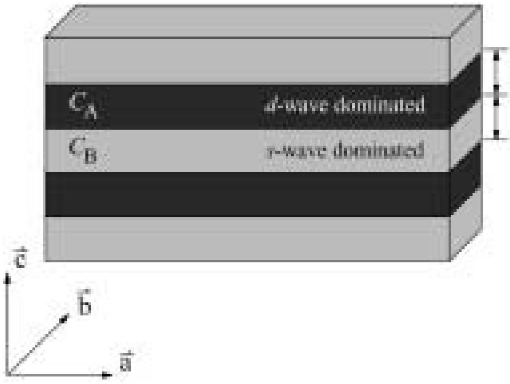

Fig. 4. Sketch of the superlattice with varying symmetry components as introduced in the text. The system consists of a superlattice formed by two superconducting compounds C_A and C_B, which are dominated by $d_{x^2-y^2}$-wave and s-wave symmetry components of the superconducting order parameter, respectively.

In tunnel junctions built from two such superlattices, *SL 1* and *SL 2*, which are connected in the *ab*-direction, the tunneling current *I* consists primarily of two components I_A and I_B tunneling between the layers C_{1A} and C_{2A} as well as between the layers C_{1B} and C_{2B}, respectively. As $\{|\Psi(C_A)|^2\} \gg \{|\Psi(C_B)|^2\}$, the tunneling current *I* is in most cases dominated by the current flowing between the layers C_{1A} and C_{2A} with the large $d_{x^2-y^2}$-wave component. Only for specific misorientation angles, in which a node of at least one $d_{x^2-y^2}$-wave order parameter is facing the junction barrier, a significant contribution from the s-wave admixture to the tunneling current will be notable.

The behavior of junctions in which the tunneling current is flowing in *c*-direction is different. In case the tunneling barrier is located between C_{2B} and C_{1B}, the tunneling shows the s-wave admixture characteristic for C_B. Remarkably, if the tunneling barrier is located between the layers C_{1A} and C_{2A}, the behavior of the tunnel junction resembles the behavior of junctions between superconductors with $d_{x^2-y^2}$-wave order parameters. For temperatures close to T_c, a rather different behavior of the system is expected. Due to the divergence of the coherence length the order parameter and ε are independent of *z*, the *c*-axis coordinate.

The model presented is aimed to point out with one example the remarkable behavior of superconductors in which the admixture of the symmetry components of the order parameter varies. Practical systems, which may be realized by using suitable thin film technologies may differ in several respects, such as the types of symmetries involved, their mixing ratios, or the sequence of the layering. Generally these systems are expected to be characterized by spatially dependent admixtures of the symmetry components of the superconducting order parameter as discussed above, with spatially constant admixture coefficients occurring in special cases only.

We propose similar symmetry variations also to be considered as intrinsic properties of the high-T_c cuprates and to resolve some of the experimental contradictions described above. In the highly anisotropic high-T_c materials such as $Bi_2Sr_2CaCu_2O_8$ the *c*-axis coherence length is in a large temperature range significantly smaller than the unit cell size in *c*-direction. At 4.2 K, ξ_c equals 1.6 Å, as compared to a unit cell length of 15.5 Å. Due to the small value of ξ_c, the magnitude of the superconducting order parameter varies significantly along the *c*-direction, providing the basis for the intrinsic Josephson effect [25].

As discussed above, in such a system the admixture of the various symmetry components does not have to be constant. The *z*-dependence of the symmetry components is controlled by the pairing mechanism and by the microstructure of the compound. Referring to the unit cell of $Bi_2Sr_2CaCu_2O_{8+\delta}$, which is sketched in Fig. 5, we point out that the CuO-planes are favoring an order parameter symmetry with lobes

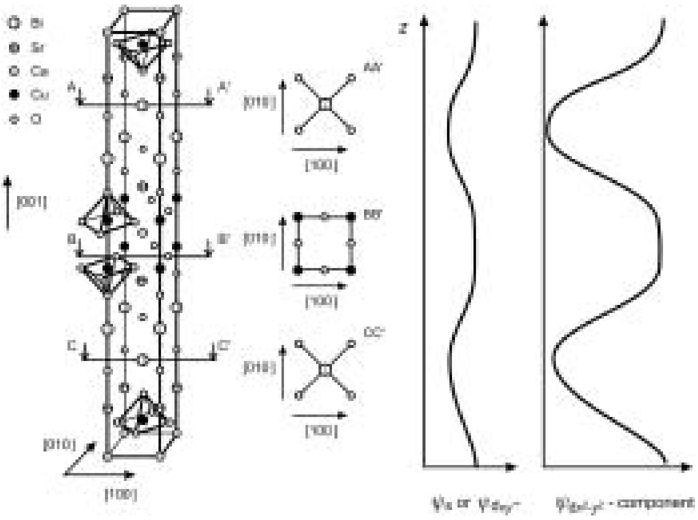

Fig. 5. Sketch of the unit cell of $Bi_2Sr_2CaCu_2O_{8+\delta}$ and cartoon of a possible scenario for the spatial variation of the admixture of an s-wave (or d_{xy}-wave) and a $d_{x^2-y^2}$-symmetry component to the superconducting order parameter.

oriented along the $\langle 100\rangle$ directions, as these directions are the ones with a large density of states at the Fermi level provided by the $\langle 100\rangle$ oriented CuO-bonds. On the other hand, in the BiO-planes, considered to be possibly superconducting due to the proximity effect [24], the Bi-O bonds are oriented along $\langle 110\rangle$. Comparably, it has been suggested that the HgO-layers in $HgBa_2CuO_{4+\delta}$ are not acting as charge reservoir layers only, but that they may be active elements in the pairing [26]. As the lobes of the $d_{x^2-y^2}$-symmetry are oriented along $\langle 100\rangle$, the Bi-O or the Hg-O bonds favor a non-$d_{x^2-y^2}$ symmetry component. In this case, a reduction of the $d_{x^2-y^2}$-order parameter component from the maximum value in the CuO-planes accompanied by a relative increase of the s-wave or of the d_{xy}-component are anticipated to occur in the BiO- or HgO planes.

For superconductors showing such variations of the admixtures of the order parameter symmetries, measurements of the order parameter symmetry will yield different values of ε, dependent on the measurement technique used, in agreement with the large body of experimental results reported.

In summary, we have fabricated all high-T_c dc π-SQUIDs in thin film technology. These π-SQUIDs operate robustly and demonstrate the potential of π-Josephson electronics. They provide clear evidence of the $d_{x^2-y^2}$-wave symmetry of the order parameter, the amount of complex admixtures of other symmetry components being undetectably small. The π-SQUIDs also reveal the presence of symmetry induced ac-currents and half fluxoids, which flip with oscillating frequencies of tens of GHz.

Trying to reconcile the results of these measurements with apparently conflicting results reported in literature, we have shown that in superconductors the admixture of various symmetry components of the order parameter does not have to be constant, but is characterized in general by spatial variations. It is found that in such cases tunnel junctions display a tunneling behavior which is characteristic for s- or $d_{x^2-y^2}$-wave dominated superconductors, depending on whether the Josephson current is flowing in the c-direction or whether it is restricted to the ab-plane.

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