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Half-$h/2e$ critical current —Oscillations of SQUIDs

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Abstract. – The current-voltage characteristics of Superconducting Quantum Interference Devices (SQUIDs) are known to modulate as a function of applied magnetic field with a period of one flux quantum $\Phi_0 = h/2e$. Here we report on the fabrication and properties of SQUIDs modulating with a fundamental period of $1/2 \times \Phi_0$. The characteristics of these bicrystal SQUIDs are consistent with either a strong $\sin(2\varphi)$ component of the current-phase relation of the Josephson current, or with an interaction between the Cooper pairs, causing an admixture of quartets to the condensate.

The interference between two superconducting condensates results in a wide range of intriguing, yet easily accessible phenomena. As shown by Josephson [1], interference between the superconducting order parameters controls the current-voltage ($I(V)$) characteristic of weak links. In first approximation, the density of the zero-voltage current traversing such a link is given by

$$J = J_c \sin(\Delta\varphi), \tag{1}$$

where J_c is the critical current density of the junction, and $\Delta\varphi$ describes the difference of the phases of the superconducting order parameter on both sides of the junction. In case a voltage is generated by the junction, it is proportional to $\partial_t \Delta\varphi$:

$$\partial_t \Delta\varphi = e^* V/\hbar, \tag{2}$$

where $e^* = 2e$ is the charge of the charge carriers in the condensate. Embedding one or two Josephson junctions into a superconducting loop that encloses an area A, superconducting quantum interference devices (SQUIDs) are obtained [2,3]. Exploiting the phase shifts

$$\vec{\nabla}\varphi = (e^*/\hbar)\vec{A} + (m^*/\hbar)\vec{v}, \tag{3}$$

induced by the magnetic flux density $\mu_0\vec{H}$ penetrating the SQUID loop, these devices are magnetometers with outstanding sensitivity. Here, $\vec{A}$ is the vector potential associated with

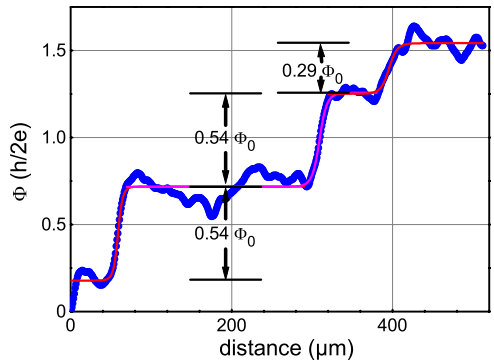

Fig. 1 – Magnetic flux Φ generated at 4.2 K by a (110)/(100) grain boundary in a $YBa_2Cu_3O_{7-\delta}$-film plotted as a function of the position along the boundary line. The measurement was done by scanning SQUID microscopy.

$\vec{H}$, m^* is the mass and $\vec{v}$ the velocity of the charge carriers. According to eqs. (1) and (3), the circulating currents in the SQUIDs, and thereby the SQUIDs' characteristics, periodically modulate with H, the period $\Delta\Phi$ being given by the magnetic flux quantum Φ_0:

$$\Delta\Phi = \Phi_0 = h/\mathrm{e}^* = h/2\mathrm{e}. \tag{4}$$

SQUIDs have been fabricated in huge numbers. In all cases in which the oscillation period was measured as a function of applied flux, it was found to equal $h/2\mathrm{e}$. In fact, rings containing grain boundary Josephson junctions have been used with great success to measure the Cooper-pair charge in $YBa_2Cu_3O_{7-\delta}$ [4]. Therefore, one might believe that the oscillation period of SQUIDs always has to be Φ_0. This belief would be erroneous, because neither is $\Delta\Phi = h/2\mathrm{e}$ required for SQUIDs that use junctions for which eq. (1) is not fulfilled (see, *e.g.*, [5]), nor do eqs. (1) and (3) predict Φ_0-periods for SQUIDs built from hypothetical superconductors with carriers characterized by charges $\mathrm{e}^* \neq 2\mathrm{e}$.

Intriguingly, experiments measuring the impedance of rf-SQUIDs [6] and transport measurements of dc-SQUIDs [7] indicate the contribution of a $\sin(2\varphi)$ component to the Josephson current for (100)/(110) grain boundaries in $YBa_2Cu_3O_{7-\delta}$, such that for these junctions the relation between the Josephson current density and the phase difference is given by $J = J_{c1}\sin(\Delta\varphi) - J_{c2}\sin(2\Delta\varphi)$. Further, in a set of experiments in which the flux-hysteresis loop of rf-SQUIDs patterned on asymmetric 30° grain boundaries was measured, the presence of a temperature-dependent second-harmonic term was found [8]. For faceted (100)/(110) boundaries it was noted that the spatially averaged current density may consist of many harmonics, even if the local tunnel current density has a first-harmonic component J_{c1} only [9]. In case J_{c2} dominates J, SQUIDs built from such junctions are expected to reveal a periodicity $\Delta\Phi = 1/2 \times \Phi_0$. While evidence for a $\sin(2\varphi)$ Josephson current-phase relationship exists, to our knowledge no observation of the expected doubling of the oscillation frequency with applied magnetic field in a SQUID has been reported.

The second possibility to generate non-$h/2\mathrm{e}$ periods, superconducting carriers with charges $\mathrm{e}^* \neq 2\mathrm{e}$, seems to be implausible. Nevertheless, superconductors with such carriers can in principle exist. We considered this idea while measuring the magnetic flux generated by (100)/(110) boundaries in bicrystalline $YBa_2Cu_3O_{7-\delta}$ films [10,11]. As shown by fig. 1, in one of these samples the flux is generated in quantities close to $\Phi \simeq h/4\mathrm{e}$ and $\Phi \simeq h/6\mathrm{e}$ ($\Phi \simeq h/8\mathrm{e}$). Although these data could not be confirmed with other samples so far, we analyzed the possibility that in superconductors such as the cuprates an interaction between the Cooper

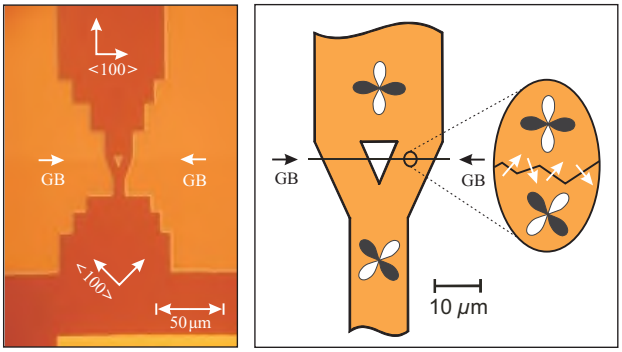

Fig. 2 – Micrograph (left) and sketch (right) of a $YBa_2Cu_3O_{7-\delta}$ SQUID used in the study. The grain boundary is marked by the arrows labelled "GB". The blow-up on the right sketches the facet structure and the corresponding Josephson current crossing the boundary.

pairs binds a fraction of the pairs into particles with charge $e^* = n \times 2e$, $n = 2, 3, 4 \ldots$. These considerations suggested the fabrication of SQUIDs from (100)/(110) boundaries to analyze their oscillation period. The necessity for such measurements is underlined by measurements of Early *et al.*, who found Shapiro steps [12, 13] at (100)/(110) boundaries to occur not only at the standard voltages $V_n = nh/2ef$, but also at voltages $V_n = nh/4ef$ [14]. Here, f is the microwave frequency used to generate the resonance steps. Furthermore, studying the critical current I_c of such a boundary in a $Bi_2Sr_2Ca_1Cu_2O_{8+x}$ film, van Harlingen and his group noted that the width of the $I_c(H)$ zero-field maximum had only half the expected value [15].

While it is possible to attribute these experimental results to effects well known in superconductivity and to peculiar artifacts resulting from inhomogeneities, we saw the need to analyze the oscillation periods of dc-SQUIDs built from asymmetric 45° boundaries to clarify whether they equal $h/2e$. We note that numerous SQUIDs containing such junctions have already been fabricated with the so-called biepitaxial process. To our knowledge, a special oscillation period was not reported for any (see, *e.g.*, [16]). This behavior may be the result of J_c inhomogeneities masking the true oscillation period. SQUIDs with asymmetric 45° junctions have also been built by Lindström *et al.* using the more reproducible bicrystal technology. The dynamics of these SQUIDs demonstrated clearly the presence of strong second harmonics in the current-phase relations of these devices [7], which oscillate with a fundamental period of $\Phi_0/2$.

We fabricated and analyzed 12 bicrystalline dc SQUIDs containing (100)/(110) boundaries, together with reference samples for calibration. The experiments were performed with $YBa_2Cu_3O_{7-\delta}$ films (10 SQUIDs on 5 chips) and with a $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ film (2 SQUIDs on 1 chip) grown by pulsed-laser deposition to a thickness of $\simeq 120\,\mathrm{nm}$. The commercial $SrTiO_3$ substrates contained (100)/(110)-asymmetric grain boundaries, specified to within 1°. The films were patterned into dc SQUID structures by photolithography and ion-beam etching before gold contacts were sputter deposited. As reference samples, nominally identical SQUIDs were grown on symmetric 45° bicrystals. The junctions have widths of $9\,\mu\mathrm{m}$, the SQUID holes are symmetric triangles with base lengths of $8\,\mu\mathrm{m}$ and heights of $9.5\,\mu\mathrm{m}$. A sketch and a micrograph of the devices are given in fig. 2. All measurements were conducted in a magnetically shielded room. The SQUIDs had non-hysteretic current-voltage $I(V)$ characteristics, their critical currents were measured by tracing the $I(V)$-curves, using a voltage criterion of $\simeq 2\,\mu\mathrm{V}$. Because the $Y_{0.6}Ca_{0.4}Ba_2Cu_3O_{7-\delta}$ sample behaved identically to the $YBa_2Cu_3O_{7-\delta}$ SQUIDs, we do not differentiate between the two in the following. The grain

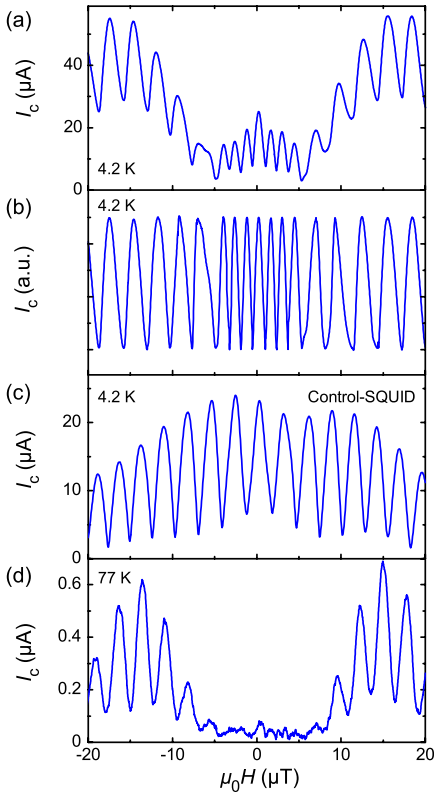

Fig. 3 – (a) Magnetic-field dependence of the critical current of an $YBa_2Cu_3O_{7-\delta}$ SQUID with an asymmetric 45° grain boundary measured at 4.2 K; (b) same data as (a) with the oscillation amplitude numerically normalized; (c) field dependence of the critical current of a control SQUID with a symmetric 45° boundary; (d) same as (a), but measured at 77 K.

boundaries in the bicrystalline $YBa_2Cu_3O_{7-\delta}$ films are faceted, with facet lengths of the order of 10–100 nm. Together with the $d_{x^2-y^2}$ symmetry of the $YBa_2Cu_3O_{7-\delta}$, the facets cause anomalous $I_c(H)$ dependencies [17]. These arise because a large fraction of the facets act as π-Josephson junctions with negative I_c.

Figure 3 displays the $I_c(H)$-dependence of a SQUID with an asymmetric 45° boundary measured at 4.2 K (a), together with the corresponding characteristic of a control SQUID with a symmetric 45° boundary (c). Both curves show the SQUID oscillations superimposed on the $I_c(H)$ curves of the Josephson junctions. These interferences occur because the width of the Josephson junctions is comparable to a side length of the SQUID hole. It is noted that for the SQUID with the asymmetric boundary, I_c is small at small H, which is due to the suppression of I_c by the π-facets. $I_c(4.2\,\mathrm{K})$ at $H = 0$ typically equals 10–100 μA for the $YBa_2Cu_3O_{7-\delta}$ samples, which corresponds to $J_c(4.2\,\mathrm{K}) = 10^3$–$10^4$ A/cm^2, typical values for 45° boundaries [17]. In these respects, the SQUIDs behave as expected. But how large are the oscillation periods? As shown by fig. 3, the SQUID oscillations of the control sample have a period of $\simeq 2.7\,\mu$T. To evaluate the magnetic flux in the loop corresponding, we calibrated the flux focussing of the samples. To do so, 24° and 36° bicrystalline $YBa_2Cu_3O_{7-\delta}$ films were structured into the sample pattern. Their oscillation periods are 2.6–2.9 μT [18], very similar to that of the control sample. Because bicrystal SQUIDs with almost identical geometries

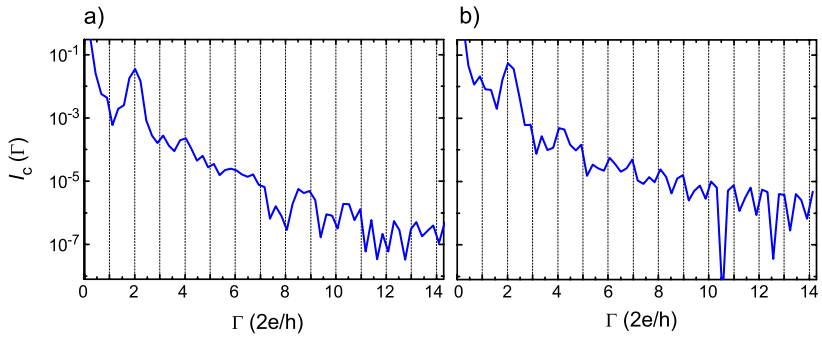

Fig. 4 – a), b) Fourier transforms of the $h/4e$ critical current SQUID oscillations for two different SQUIDs fabricated in different runs for the magnetic-field range of $|H| \leq 5\,\mu$T. a) shows the data presented in fig. 3(a). The data have been taken at 4.2 K.

have been reported to oscillate with $h/2e$ [19], it is evident that the oscillation period of the control SQUIDs is $h/2e$, corresponding to a flux focussing factor of 20.2.

In contrast to the standard SQUIDs, the oscillation periods of the SQUIDs with the (100)/(110) boundaries were found to be $1.44\,\mu$T (fig. 3a,b). Because the flux focussing factor and the loop areas are nominally identical to the control SQUIDs, this periodicity equals $\Phi_0/2 = h/4e$. In the SQUID shown in fig. 3a, the period even changes from $h/4e$ to $h/2e$ when H exceeds $5\,\mu$T (fig. 3). Providing an internal calibration, this curve gives additional evidence that the oscillation period equals $\Phi_0/2$ at small H. The fundamental oscillation period of $\Phi_0/2$ is clearly revealed by the Fourier-transform of the $I_c(H)$-dependence (fig. 4). The spectra even suggest minute signals at $h/8e$, although the higher harmonics die off quickly. The temperature dependencies of both oscillation periods differ. While at 77 K the $\Phi_0/2$ oscillations have almost completely disappeared, the Φ_0 oscillations are still pronounced (see fig. 3d).

$I_c(H)$-oscillations with a period of $\Phi_0/2$ were displayed by nine out of the twelve SQUIDs with asymmetric 45° grain boundaries that we have fabricated. In many cases the oscillation period switched to $h/2e$ when the applied field exceeded ≈ 5–$10\,\mu$T. Three SQUIDs showed $h/2e$ oscillations over the whole field range, two of them with very weak $h/4e$ components even at larger applied magnetic fields. The differences in the behavior of these samples may be caused by differences of the facetted microstructure of the grain boundaries. This may also be the reason why the $I_c(H)$ patterns of the (100)/(110) grain boundary SQUIDs analyzed in ref. [7] differ from the $I_c(H)$ patterns generally observed in our samples.

Clearly, the dc properties of the SQUIDs are highly unusual and do not agree with simple expectations based on the dc-Josephson relation (eq. (1)). To analyze whether the rf properties of the SQUIDs also differ from the standard behavior as predicted by the ac-Josephson relation (eq. (2)), we measured the $I(V)$ curves of the samples under microwave radiation at 11.88 GHz. Since the radiation caused the samples to become noisy at small H, these experiments could only be performed in the large-H regime, for which the samples showed the $h/2e$ period. The $I(V)$ characteristics display conventional, integer Shapiro steps $V_n = nh/2ef$ in case the samples are biased with magnetic fields for which the SQUID oscillation have I_c maxima. Shapiro steps at half the standard frequency $V_n = nh/2ef$ are seen if the junctions are operated in magnetic fields that result in I_c minima.

The SQUID properties strongly suggest that they are caused by a dominating J_{c2}-contribution to the Josephson current, in agreement with refs. [6–8]. For (100)/(110) boundaries, a large J_{c2}/J_{c1} ratio is expected, because J_{c1} is suppressed by the tunneling geometry, which

couples the nodal with the anti-nodal directions of the order parameters. Further, because $\vec{\nabla}\varphi$ is insignificant at small H (eq. (3)), the π-facets have negative current densities in this field range: $J_1 = J_{c1}\sin(\Delta\varphi+\pi) = -J_{c1}\sin(\Delta\varphi)$ and therefore suppress I_{c1}. However, these facets do not suppress I_{c2}, because at the π-facets $J_2 = J_{c2}\sin(2\times(\Delta\varphi+\pi))$. Therefore, the I_{c2}/I_{c1} ratio is enhanced at small H, precisely where the $\Phi_0/2$-period is seen. The suppression of I_{c1} by the asymmetric 45° boundaries also explains why the $h/4$e-period has only been seen for SQUIDs fabricated from such junctions. Because the facet structure of the grain boundaries plays a key role in this mechanism, it is understandable that one of the samples showed $\Delta\Phi = h/2$e. Due to non-identical growth parameters, the devices vary in their facet structures and therefore vary in their $J_{c2}(x)/J_{c1}(x)$ ratios. This may also explain the difference of the basic oscillation period of the samples presented by Lindström *et al.* and ours.

Of course, if the data were interpreted in conventional terms, such that the SQUID oscillations are taken as a measure of e*, the $h/4$e-oscillations would propose that carriers with charge 4e are present at the grain boundary, and that their condensate couples the phases of the two Josephson junctions around the SQUID hole. The possibility that this model was correct seems to be remote, because the BCS ground state is composed solely of Cooper pairs [20]. However, the derivation of this ground state is based on the assumption that all interactions between the charge carriers are two-particle interactions, described by the term $V_{\vec{k},\vec{k}'}$. This mean-field approximation is clearly appropriate for conventional superconductors, in which the coherence length ξ is much larger than the spacing d between Cooper pairs. However, because in the cuprates ξ and d are of the same order, and because correlation effects are predominant, it is not clear why this approximation should be applicable to the high-T_c compounds [21,22].

The possible attractive or repulsive interaction between an electron or a hole of one Cooper pair with an electron or a hole of a second pair, arising, for example, from magnetic interactions, Coulomb interactions or lattice distortions, results in a pair-pair interaction energy $V_{CP,CP}$. Depending on the sign of $V_{CP,CP}$, the two Cooper pairs are correlated or anticorrelated. For a strongly attractive $V_{CP,CP}$ interaction they pair. For weak $V_{CP,CP}$, these pairs dynamically rearrange themselves in a fluctuating manner. The correlations between the pairs result in a non-BCS ground state, expressed by the ansatz

$$\Psi = \alpha_2\Psi_{2e} + \alpha_4\Psi_{4e} + \alpha_6\Psi_{6e} + \ldots, \tag{5}$$

where $\alpha_2\Psi_{2e}$ is the Cooper-pair term and the higher-order terms describe the Cooper-pair multiples, such as quartets or sextets.

We note that an analysis of the temperature dependencies of the penetration depths and of the fluctuation contributions to the conductivity and to the magnetization led to the proposal that the superconducting phase transition of the cuprates is due to interacting pairs [22]. Intriguingly, a related situation exists for the binding of nucleons in light nuclei. While pairs of nucleons condense into a superconducting condensate, they may bind by their higher-order correlations into multiples, in particular into alpha-particle–like quartets [23]. The onset of "quartetting" in nuclear matter has been considered [24] and found to predominate in the low-density regime.

Because the Josephson current crossing weak links also couples the higher-order multiples, SQUIDs built from superconductors in which $|\alpha_4|^2/|\alpha_2|^2$ is non-negligible are expected to show an $h/4$e contribution in their oscillations. For this to occur, the $\alpha_4\Psi_{4e}$ term has to be sufficiently large to provide phase coupling around the SQUID loop. Although this exotic mechanism is consistent with the measured $h/4$e-behavior of the SQUIDs, it has to provide answers for the questions why $h/4$e periods have never been observed for SQUIDs with more conventional junction geometries, and why the Abrikosov vortices carry a flux of $h/2$e. To account for these effects, one would need to conclude that $|\alpha_4|^2/|\alpha_2|^2$ is enhanced at (100)/(110) interfaces.

One might expect to find non-$h/2$e oscillation periods or non-$h/2$e vortices also in other systems and under different circumstances, for example half flux quanta in the bulk, in particular if further mechanisms exist that cause the unusual SQUID properties reported.

In summary, we have fabricated nine high-T_c SQUIDs which display $I_c(H)$ oscillations with a period of $1/2 \times h/2$e. Two mechanisms are found to be consistent with these characteristics. The more conventional and more likely one is based on Josephson currents with a $J = J_c \sin(2\Delta\varphi)$ current-phase relation. The other mechanism is based on interacting pairs that form quartets with charge 4e. Further experiments are required to clarify whether one of these effects, or perhaps another one, underlies the SQUID behavior, and whether these mechanisms are possibly realized in other systems.

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