Microscopic model for multiple flux transitions in mesoscopic superconducting loops

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A microscopic model is constructed which is able to describe multiple magnetic flux transitions as observed in recent ultra-low temperature tunnel experiments on an aluminum superconducting ring with normal metal - insulator - superconductor junctions [Phys. Rev. B 70, 064514 (2004)]. The unusual multiple flux quantum transitions are explained by the formation of metastable states with large vorticity. Essential in our description is the modification of the pairing potential and the superconducting density of states by a sub-critical value of the persistent current which modulates the measured tunnel current. We also speculate on the importance of the injected non-equilibrium quasiparticles on the stability of these metastable states.

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When a superconducting loop is exposed to a perpendicular magnetic field the energy $E_L$ of the $L$-th state is given by:

$$E_L \sim \frac{1}{S} \left( \frac{\Phi}{\phi_0} + L \right)^2,$$

where $L = 1/2\pi \oint S \nabla \theta ds$ is the vorticity, $\theta$ is the coordinate-dependent phase of the superconducting order parameter, and the integration is made along the contour of the loop. $S$ is the loop’s circumference, $\Phi$ is the magnetic flux through the area of the loop, and $\phi_0 = \hbar/2e$ is the superconducting flux quantum. If the system can relax to its ground state, then sweeping the magnetic field causes a periodic variation of the kinetic properties with period $\Delta \Phi = \phi_0$ corresponding to transitions $\Delta L = \pm 1$. The persistent current in the loop is proportional to the derivative of the energy $I \sim dE_L/d\Phi$, and shows the characteristic sawtooth behavior with the same period. The ‘switching’ supercurrent density corresponding to transitions from the state with vorticity $L$ to the nearest state $L \pm 1$ is $I_{\text{switch}} \sim 1/S$. This $\Delta \Phi = \phi_0$ behavior is commonly observed at temperatures close to the critical one.

Recently, low temperature experiments ($T \ll T_c$) were performed on superconducting loops leading to jumps with vorticity changes of $\Delta L > 1$. A phenomenological explanation, based on a numerical solution of the time-dependent Ginzburg-Landau equations, was given in Ref. The basic idea of that theory was as follows: at sufficiently low temperature and when sweeping the magnetic field up the system can ‘freeze’ in a metastable state with vorticity $L_1$. When the circulating persistent current reaches the corresponding critical density $j_c \sim 1/\xi$ this metastable state relaxes to the low-energy level $L_2 \gg L_1$. Thus, in loops with perimeter $S \gg \xi$ one may observe vorticity changes $\Delta L \sim j_c/I_{\text{switch}} \sim S/\xi \gg 1$. One should notice that the explanation based on the Ginzburg-Landau formalism is strictly valid only at temperatures close to the critical one, and the extrapolation to the low-temperature limit requires further justification. Therefore, of particular interest are ultra-low temperature experiments where Al loops with circumference from a few $\mu m$ to a few hundred $\mu m$ were studied, and a well-defined periodic structure with very large vorticity changes $\Delta L$ up to $\sim 50$ were observed. In the present paper we will construct a microscopic model based on the Usadel equations to analyze the experiments of Ref.

Within the Usadel formalism, superconducting correlations between electrons forming a Cooper pair are described by two complex functions: the pairing angle $\theta$ and the phase $\chi$. In the most general case, both functions depend on the coordinate $r$ and the energy $E$. The pairing angle and the phase are linked through the coupled system of equations:

$$\frac{\hbar D}{2} \nabla^2 \theta + [iE - \frac{\hbar}{2D} v_s^2 \cos \theta] \sin \theta + \Delta \cos \theta = 0, \quad (2a)$$

$$\nabla (v_s \sin^2 \theta) = 0, \quad (2b)$$

where the superfluid velocity is $v_s = D(\nabla \chi - (2e/\hbar) A)$, $D$ is the diffusion coefficient and $A$ is the vector potential. The relation between the normal state density of states at the Fermi level $N(0)$ and the superconducting density of states is given by $N(E) = N(0) Re[\cos(\theta(E))]$. The pairing potential $\Delta(r)$ should be defined self-consistently from
the integral equation:

$$\Delta(r) = N(0)V_{eff} \int_0^{\hbar \omega_D} \tanh(E/2k_B T)Im(\sin \theta) dE,$$

(3)

where $V_{eff}$ is the pairing interaction strength and $\omega_D$ is the Debye frequency. These equations are valid for diffusive superconductors with a mean free path $\ell$ much less than the 'pure limit' coherence length $\xi_0 = 0.18 \hbar v_F/\Delta_0$ (where $v_F$ is the Fermi velocity of the electrons on the Fermi surface and $\Delta_0$ is the gap at zero temperature, zero magnetic field and in the absence of transport current).

For loops consisting of a sufficiently narrow wire $\sqrt{\sigma} \leq \xi$ one may neglect the variation of the superconducting properties in the transverse direction, where $\sigma$ is the cross section of the wire. As a first approximation one may also neglect the influence of the tunnel contact on the properties of the superconducting loop. Within these approximations we obtain from Eqs. (2) and (3) a single algebraic equation:

$$E + i \Gamma \cos \theta = i \Delta \cos \theta \sin \theta$$

(4)

where $\Gamma = \hbar \langle v_s^2 \rangle / 2D$ is the depairing energy averaged across $\sigma$. One may neglect the self-induced magnetic field (for the largest loop with $S = 100 \mu m$ it leads to an error less than 1%), and consider the vector potential $A$ as entirely due to the applied field $A = (H_y/2, -H_x/2, 0)$. For arbitrary vorticity $L$ in a rectangular loop the depairing energy $\Gamma$ is:

$$2\Gamma/\Delta_0 = \left( \frac{\pi L}{2} \right)^2 \frac{1}{(w-2d)w} - \frac{\pi LH}{4} + \frac{H^2}{16} (w^2 - 2wd + 4d^2/3)$$

(5)

where the magnetic field $H$ is measured in units of $H_{c2} = \Phi_0/2\pi \xi(0)^2$, $w$ is the side of the loop and $d$ is the width of the wire the loop is made of and both are measured in units of $\xi(0) = \sqrt{\hbar \Delta_0 / D}$.

FIG. 1: Dependence of the gap (a), pairing potential $\Delta$ (a), current density (b) and jump in vorticity $\Delta L$ (inset in Fig. 1(b)) on the applied magnetic field in a superconducting loop with parameters: $T_c = 1.2 K$, $\xi(0) = 150$ nm, $w = 5 \mu m$, $d = 120$ nm, $T = 106$ mK.

It was shown that for quasi-one-dimensional superconducting loops of large circumference ($S \gg \xi$) metastable states with fixed vorticity $L_n$ become unstable if the persistent current density is equal to the depairing current density $j_c$. In the present calculation we assume that when the current density in the loop (being a function of magnetic field $H$) reaches its maximum (at a fixed value of vorticity $L$) the loop switches to a new quantum state. The new value of the vorticity $L_m$ is then found from the condition that the pairing potential $\Delta$ (minimum depairing energy $\Gamma$) for a given magnetic field is maximal. In terms of a quasiclassical description this transition criterion corresponds to the supervelocity $v_s$ being equal to its critical value. In Fig. (1) the calculated dependencies of the superconducting gap, pairing potential $\Delta$ and superconducting current density are plotted as function of the applied magnetic field for a superconducting loop with parameters similar to the ones used in Ref. 4. Of particular interest is the fact that for small magnetic fields the period of oscillations is much larger than $\phi_0$ and this period decreases in higher fields. Both observations are qualitatively consistent with the recent experiments 2,3,4. However,
simulations systematically give larger vorticity jumps than the ones observed in the experiment although provide the same dependence on the loop size (compare Eq. (6) and Fig. (7) in Ref. 3). Reasonable explanation of the discrepancy is the presence of inevitable imperfections acting as weak links in real samples. Even at the lowest temperatures the system exhibits transitions from the 'frozen' metastable states at smaller values of the magnetic field. In Ref. 1 it was shown that an inhomogeneity in the loop can indeed strongly decrease the values of the actually observed vorticity changes. Close to the critical temperature thermal fluctuations disable the formation of metastable states, which results in the 'conventional' \( \phi_0 \) periodicity (\( \Delta L = \pm 1 \)).

It is interesting to compare the results of the present microscopic approach with calculations based on the Ginzburg-Landau model. For a rectangular loop with side \( w \) and thickness \( d (w \gg \xi \geq d) \) at \( T/T_c \ll 1 \) the transition criterion for a state with vorticity \( L = 0 \) corresponds to the condition \( \Gamma_c/\Delta_0 \sim 0.235 \) (at \( T \rightarrow 0 \)). Then the size of the vorticity jump is:

\[
\Delta L = \text{Nint} \left( \sqrt{\frac{2 \Gamma_c}{\Delta_0}} \frac{2w}{\pi \xi} \right) \approx \text{Nint} \left( \frac{0.68}{\pi} \frac{2w}{\pi \xi} \right) \tag{6}
\]

where \( \text{Nint}(x) \) returns the integer value of the real variable \( x \). The solution of the Ginzburg-Landau equations for the same transition gives:

\[
\Delta L^{GL} = \text{Nint} \left( \frac{1}{\sqrt{3}} \frac{2w}{\pi \xi} \right) \approx \text{Nint} \left( \frac{0.58}{\pi} \frac{2w}{\pi \xi} \right) \tag{7}
\]

The slight numerical discrepancy between Eqs. (6) and (7) is due to the different functional dependence of \( \Gamma_c \) in the Ginzburg-Landau model and the present one. The quantitative difference between Eqs. (6) and (7) is insignificant compared to experimental inaccuracies.

So far we have considered persistent currents in an isolated superconducting loop. Periodic modulation of these currents by an external magnetic field can be measured using magnetization or calorimetric methods. In the ultra-low temperature experiments of Ref. 4 an Al loop was used as the superconducting (S) electrode of a N-I-S junction being overlapped through a tunnel barrier (I) with a normal-metal (N) contact. The tunnel current \( I_{\text{tun}} \) was measured at fixed bias voltage (or the voltage across the barrier at a fixed tunnel current) as a function of the applied perpendicular magnetic field. We argue that the observed periodic variation of the tunnel current (voltage) with magnetic field is due to the modulation of the gap and the pairing potential by the sub-critical persistent current in the loop-shaped superconducting electrode of the N-I-S structure. To calculate the tunnel current we use the conventional 'semiconductor' model which gives for the tunnel current:

\[
I_{\text{tun}} = \alpha |T|^2 \times \int_{-\infty}^{\infty} \frac{N(E)}{N(0)} [f(E, T) - f(E + eV, T)] dE \tag{8}
\]
where \( f(E, T) \) is the Fermi distribution function, \( \alpha \) is the constant of proportionality, and \( T \) is the tunnelling matrix element. It can be shown that \( |\alpha|^2 = 1/eR_T \), where \( R_T \) is the tunnel resistance of the junction in the normal state and \( e \) is the electron charge. In our case of strong currents, to obtain the density of states in the superconductor \( N(E) \) we solved Eqs. (2-5) self-consistently.

The described approach provides a qualitative description of the main features observed in the experiment of Ref.\(^4\): multi-flux periodicity of the tunnel current and the decrease of the period of the tunnel current oscillation with increasing magnetic field. However, quantitative comparison is far from being perfect: the measured current of the voltage biased N-I-S junction in the limit \( eV < \Delta \) is systematically higher than the value calculated using Eq. (8) (see Fig. 2(a)). We believe that to obtain a better quantitative agreement one should consider higher-order processes describing tunnelling of pairs of electrons\(^13\), giving rise to the sub-gap current. To account for this process we use a combination of the results obtained in Ref.\(^13\) for pure superconductors (quasi-particle transport in the ballistic regime) and the simple 'semiconductor' model\(^15\). This leads to the semi-quantitative result for the sub-gap current:

\[
I_{\text{sub}} = \alpha |T|^2 \times \int_{-\infty}^{\infty} A(\Delta, E)[f(E, T) - f(E + eV, T)]dE
\]

where the probability of Andreev reflection \( A(\Delta, E) \) is taken from Table II of Ref.\(^13\):

\[
A(\Delta, E) = \begin{cases} \frac{\Delta^2}{E^2 + (\Delta^2 - E^2)(1 + 2Z^2)^2}, & E < \Delta, \\ \frac{(E^2 - \Delta^2)(\sqrt{E^2/(E^2 - \Delta^2)} + 2Z^2 + 1)^2}{\Delta^2}, & E > \Delta, \end{cases}
\]

with \( Z \) being the dimensionless barrier strength\(^13\), and \( \Delta \) is found from solutions of Eqs. (2-5). The tunnel current \( I_{\text{tun}} \) and the sub-gap current \( I_{\text{sub}} \) have different functional dependencies on \( \Delta \), and hence their variation in magnetic field is also different. While sweeping the magnetic field the magnitude of the oscillations of the tunnel current is much higher than the magnitude of oscillations of the sub-gap current. Strictly speaking, this over-simplified introduction of a sub-gap current in our model is not applicable for dirty limit electrodes studied in Ref. 4. However, it demonstrates that at energies \( \Delta << eV \) an account for the sub-gap current contribution gives qualitatively better agreement with experiment: Fig. 2 and Fig. 3. The described approach is not able to give quantitative exact values for the sub-gap current as other mechanisms (e.g. leakage current) might contribute to the total current measured in N-I-S structures. It is also known that the magnitude of the sub-gap current strongly depends on interference effects within the locus of the N-I-S junction, and hence is geometry-dependent\(^14\).

\[\text{FIG. 3: Magnetic field dependence of the total current through a N-I-S junction for different values of the bias voltage (a) and normalized magnitudes of the low field (B < 5 mT) current oscillations } \Delta I/I_{\text{max}} \text{ as function of the normalized bias } eV/\Delta(T) \]

(b). Parameters of the loop are the same as in Fig. 1.

In spite of the obvious simplifications when describing the sub-gap current, Eqs. (8-10) combined with calculations of the density of states (Eqs. (2-5)), is able to give good qualitative and reasonable quantitative agreement with experiment. As follows from Fig. 2, and in full agreement with the experimental findings\(^4\), the absolute value of the magnitude of the current oscillations as function of the magnetic field \( I(B, V = const) \) (Fig. 3(a)) and their normalized...
magnitude $\Delta I/I_{\text{max}}(B, V = \text{const})$ (Fig. 3(b)) depend on the bias voltage $V$. The only 'tuning' parameter used in fitting the calculations with experiment is the coefficient $Z$ (see Fig. 2(a)).

At high temperatures the current across a tunnel junction at $V \ll \Delta$ is mainly determined by the tunnel component Eq. (8) due to temperature smearing of the Fermi distribution function. While at very low temperatures the contribution of the tunnel current is negligible in the same limit, and the finite measured current is practically equal to the sub-gap term of Eqs. (9,10). For all temperatures and $V > \Delta$ the total current is determined by the tunnel component. In view of the relatively weak dependence of the sub-gap current (see Eqs. (9,10)) on magnetic field, this explains qualitatively the existence of a maximum in the dependence of $\Delta I/I_{\text{max}}$ versus voltage at low temperatures, and its absence at higher temperatures (Fig. 3(b)).

In order to improve our model one should include the injection of non-equilibrium quasiparticles from the normal electrode. Particularly at energies $eV \sim \Delta$ this effect results in a cooling of the normal-metal contact and a heating of the superconducting contact. The inevitable consequence is a spatially-inhomogeneous modification of the superconducting gap within the locus of the N-I-S junction. Manifestation of such an effect has been observed in S-I’S-I’S structures at temperatures comparable to the critical temperature of the S’ electrode. At $T \rightarrow 0$ there are very few equilibrium (thermally activated) excitations, and one might expect that even a small amount of extra quasiparticles injected from the normal electrode may affect the pairing potential $\Delta$ and, hence, influence the stability of the 'frozen' metastable states. We speculate that the mentioned non-equilibrium effect might explain also the variation of the period $\Delta B_I$ of the current oscillation on the bias voltage (Fig. 6 and Fig. 8(c) in Ref. 4). The larger the applied voltage, the higher the quasiparticle injection, and, hence, the stronger the deviation of the distribution function in the superconducting electrode from its equilibrium value. Therefore a metastable state with a given vorticity becomes unstable at smaller values of the depairing energy $\Gamma$, or, in other words, at smaller values of the applied magnetic field.

In conclusion, using a microscopic approach we analyzed the ultra-low temperature behavior of a mesoscopic-size superconducting ring in the presence of a magnetic field. The model was used to interpret recent experiments on N-I-S junctions with a loop-shaped superconducting electrode. The central result of the present work is the demonstration that the tunnel current oscillates in a magnetic field with a period, which scales with the loop circumference. For large loops flux changes are much larger than a single flux quantum. We found agreement with experiment. One should include the finite sub-gap current in order to improve the quantitative agreement between the present model and the measured total current across the junction. Using simple assumptions, we were able to obtain a reasonable quantitative agreement with the experimental results of Ref. 4.

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