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Quantum tunneling of the interfaces between normal-metal and superconducting regions of a type-I Pb superconductor

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Evidence of a non-thermal magnetic relaxation in the intermediate state of a type-I superconducor is presented. It is attributed to quantum tunneling of interfaces separating normal and superconducting regions. Tunneling barriers are estimated and temperature of the crossover from thermal to quantum regime is obtained from Caldeira-Leggett theory. Comparison between theory and experiment points to tunneling of interface segments of size comparable to the coherence length, by steps of order one nanometer.

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Quantum tunneling of relatively macroscopic solid-state objects like flux lines in type-II superconductors^{1,2} and domain walls in magnets³ have been subject of intensive research in the past. The corresponding energy barriers and spatial scales are non-trivially determined by statistical mechanics of the pinning potential^{1,2,4,5}. Interaction with environment makes this problem the one of macroscopic quantum tunneling with dissipation⁶. The latter is especially important for the tunneling of flux lines because of their predominantly dissipative dynamics^{7–11}. Type-I superconductors, when placed in the magnetic field, do not develop flux lines. Instead, they exhibit intermediate state in which the sample splits into normal and superconducting regions separated by planar interfaces of positive energy^{12–14}. Recently, there has been a renewed interest to the equilibrium structure, pinning, and dynamics of interfaces in type-I superconductors^{15–20}. In the presence of pinning centers the interfaces adjust to the pinning potential by developing curvature as is schematically shown in Fig. 1. Pinning by point or small-volume defects should result in a broad distribution of energy barriers. It is, therefore, plausible that at low temperature type-I superconductors continue to relax towards equilibrium via quantum diffusion of interfaces. This situation is similar to the diffusion of domain walls in disordered ferromagnets with one essential difference. Contrary to a ferromagnetic domain wall, the dynamics of the planar interface in a superconductor should be dominated by dissipation.

At low temperature the decay of metastable states created by pinning provides slow relaxation of magnets and superconductors towards thermal equilibrium. This relaxation is known as magnetic after-effect. At finite temperature it may occur via thermal activation with a probability proportional to $\exp(-U_B/T)$ where U_B is the energy barrier. As $T \to 0$ thermal processes die out and the only channel of escape from the metastable state becomes underbarrier quantum tunneling. Its probability is proportional to $\exp(-I_{eff}/\hbar)$ where I_{eff} is the effective action associated with tunneling. The pre-exponential factors in the two expressions are of lesser importance because the dependence of



FIG. 1: Schematic view of the interface between normal and superconducting regions of type-I superconductor in the random pinning potential.

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FIG. 2: Color online: Isothermal magnetization curves for sample A. (Data for sample B are similar.) The inset shows scaling with $m = M/B_c(T)$ and $h = H/B_c(T)$.

the probability on the parameters is dominated by the exponents. Equating the two exponents, one finds that the crossover from thermal activation to quantum tunneling occurs at $T_Q \approx \hbar U_B/I_{eff}$. Experimental evidence of such a crossover in type-II superconductors has been overwhelming²¹. There has also been some experimental evidence of quantum diffusion of domain walls in disordered ferromagnets²². However, to our knowledge, no literature exists on non-thermal magnetic relaxation in type-I superconducors. Experimental evidence of such a relaxation and its theoretical treatment are subjects of this paper.

Two samples (A and B) in the shape of an octagonal disk of thickness 0.2 mm and surface area 40 mm² were prepared by cold-rolling of short cylinders cut from a commercial Pb rod of purity 99.999%. They were annealed during one hour at 290°C and 280°C (melting temperature of lead is 327.5°C), respectively, in glycerol and nitrogen atmosphere to reduce the mechanical stress from defects that might have been introduced during preparation of the sample. Magnetic measurements were performed with the use of a commercial superconducting quantum interference device (SQUID) magnetometer in the field up to 1 kOe in the temperature range 1.8K - 8K. The magnetometer was equipped with a Continuous Low Temperature Control (CLTC) and Enhanced Thermometry Control (ETC) and showed thermal stability better than 0.01 K. Isothermal magnetization curves had the same shape for samples A and B, see Fig. 2. The fit of the data by $B_c(T) = B_c(0)[1 - (T/T_c)^2]$ produced identical values of $B_c(0) = 802 \pm 2$ Oe and $T_c = 7.23 \pm 0.02$ K for both samples, in accordance with the values of the critical field and transition temperature reported for lead. These values of the parameters, together with high purity of our samples, confirm that we are dealing with a conventional type-I superconductivity in lead. The observed magnetization curves are typical of a pure type-I Pb superconductor in a weakly pinned intermediate state (see, e.g., Refs. 15,18). In such a state the type-I superconductor has the magnetization curve that is qualitatively similar to M(H) of the type-II superconductor. This is because many of the physical processes involved in the formation of the intermediate state of a type-I superconductor are conceptually similar to the physical processes responsible for the formation of the mixed state of a type-II superconductor. The essential difference is that the magnetic field penetrates into a type-I superconductor in the form of normal domains vs quantized vortices in a type-II superconductor. The maxima in the virgin magnetization curves in Fig. 2 (not to be confused with H_{c1} -effect in a type-II superconductor) are due to the surface barriers for the nucleation of normal domains. These barriers and the pinning of interfaces separating normal and superconducting regions are responsible for the magnetic hysteresis. At some higher field (not to be confused with H_{c2} in a type-II superconductor) the magnetization goes to zero due to the complete expulsion of superconducting domains by normal domains.

Magnetic relaxation was measured by first applying the field $B > B_c(T)$, then subsequently switching the field off and recording (for more than one hour) isothermal temporal evolution of the remnant magnetization $M_{\rm rem}(T)$ in a zero field. Fig. 3 shows the time evolution of $M_{\rm rem}(t)/M_{\rm rem}(0)$ in sample A between 2 K and 6 K. Similar data with slightly different slopes were obtained for sample B. At all temperatures the observed slow relaxation followed very well the logarithmic time dependence, $M_{\rm rem}(t) = M_{\rm rem}(0)[1 - S(T) \ln t]$, where S(T) is the so-called magnetic viscosity. Temperature dependence of the viscosity for samples A and B is shown in Fig. 4. Remarkably it does not extrapolate to zero in the limit of $T \to 0$ but, instead, tends to a finite temperature-independent limit as the sample is cooled down.

Conceptually, the slow relaxation of interfaces separating normal and superconducting regions is similar to the magnetic after-effect due to relaxation of domain walls in bulk ferromagnets. In the latter case the logarithmic time-dependence of the relaxation is usually considered an indication of the broad distribution of energy barriers³. Same



FIG. 3: Color online: Magnetic relaxation of sample A at various temperatures. (Data for sample B are similar.) Logarithmic time dependence provides an accurate fit to the data.



FIG. 4: Color online: Temperature dependence of the magnetic viscosity for samples A (black) and B (red). S(T) tends to a non-zero value in the limit of $T \rightarrow 0$. Experimental error is less than the size of the points.

result can be obtained in a model with a single barrier if the height of the barrier is affected by the global relaxation²³. Regardless of the model, the finite value of S(0) points towards quantum mechanism of the escape from metastable states. By analogy with type-II superconductors where non-thermal magnetic relaxation is due to quantum tunneling of flux lines, and with ferromagnets where non-thermal relaxation implies tunneling of domain walls, it is reasonable to assume that in type-I superconductors the effect is due to quantum tunneling of interfaces separating normal and superconducting regions. The structure of the interface (see Fig. 5) is determined by two parameters: the coherence length ξ and the London length λ_L . Type-I superconductivity corresponds to $\kappa = \lambda_L/\xi < 1/\sqrt{2}$. Concentration of Cooper pairs $|\Psi|^2$ gradually goes to zero on a distance ξ as one moves through the interface from the superconducting to the normal region. When crossing the interface in the opposite direction one would see the magnetic field going down from its thermodynamic critical value B_c to zero on a distance $\delta = \sqrt{\lambda_L \xi} < \xi$.

The energy of the unit area of the interface is^{24}

$$\sigma = \xi B_c^2 / (3\sqrt{2}\pi) \,. \tag{1}$$

Pinning provides curvature of the interface, see Fig. 1. We shall describe such an interface by a singled-valued function $Z(x, y, \tau)$. The energy of the interface,

$$E = \sigma \int dx dy \left[\sqrt{1 + \left(\frac{dZ}{dx}\right)^2 + \left(\frac{dZ}{dy}\right)^2} + U(x, y, Z) \right], \qquad (2)$$



FIG. 5: Structure of the interface between normal and superconducting regions of type-I superconductor. The magnetic field decays on a scale $\delta = \sqrt{\lambda_L \xi}$, while the modulus of the Cooper-pair condensate wave function changes on a scale ξ .



FIG. 6: Flattening (or formation) of a bump via quantum tunneling of a pinned interface (I) separating normal (N) and superconducting (SC) regions.

consists of two parts: Elastic energy and energy due to pinning potential U(x, y, z). Metastable equilibrium is achieved through the balance of these two energies that corresponds to the minimum of Eq. (2). Magnetic relaxation occurs due to the decay (or formation) of the bumps in the interface shown in Fig. 1. We shall describe such a bump by the lateral size L and height a. For a particular bump these parameters are determined by the local pinning potential. Since the latter is unknown we shall test self-consistency of the approach based upon theory of tunneling with dissipation⁶ by extracting the average values of L and a from experiment.

Let us first estimate the energy barrier associated with the bump. It is easy to see that the change in the elastic energy of the interface due to formation of the bump (see Fig. 6) is independent of L and is generally of order $\sigma \pi a^2$. (This follows from the fact that the area of a spherical segment above any cross-section of a sphere differs from the area of that cross-section by πa^2). This energy must be balanced by the negative energy of the pinning to make the bump an equilibrium state of the interface. Consequently, $U_B \approx \pi \sigma a^2$ with the average value of a should represent the typical amplitude of the random pinning potential and, thus, the height of the energy barrier. Note that the transport current would tilt the pinning potential and lower the barriers. In this paper, however, we consider quantum relaxation towards equilibrium in the absence of the transport current (similar to the magnetic relaxation of a ferromagnet in a zero magnetic field), rather than quantum creep of the interfaces caused by the transport current.

We want to find the WKB exponent, I_{eff}/\hbar , for the tunneling of Z(x, y) between two configurations of the interface corresponding to the local energy minima (see Fig. 6). Same as for the flux lines⁷⁻¹¹ we shall assume that the tunneling probability is dominated by the dissipation part of the Caldeira-Leggett effective action⁶:

$$I_{eff} = \frac{\eta}{4\pi} \int_0^{\hbar/T} d\tau \int_{-\infty}^{\infty} d\tau' \int dx dy \, \frac{[Z(\tau) - Z(\tau')]^2}{(\tau - \tau')^2} \,, \tag{3}$$

where η is a viscous drag coefficient describing dissipative motion of the interface and $\tau = it$ is imaginary time. For a segment of the interface of size L, that tunnels by a distance a, the T = 0 value of the effective action in Eq. (3) can be estimated as

$$I_{eff} \approx \eta L^2 a^2 / (4\pi) \,. \tag{4}$$

The drag coefficient η can be obtained from the argument similar to that of Bardeen and Stephen for the flux lines²⁵. Let magnetic field be in the y-direction. In the presence of the current of density j in the x-direction, the

magnetic force experienced by the dxdy element of the interface in the z-direction is

$$dF = \frac{1}{c} \int dx dy dz \, jB \,. \tag{5}$$

Writing j via the electric field and normal-state resistivity ρ_n as $j = E/\rho_n$, and substituting here E = (V/c)B for the electric field produced inside the interface moving at a speed V in the z-direction, one has $j = (V/c)(B/\rho_n)$. This gives

$$\frac{dF}{dxdy} = \frac{V}{\rho_n c^2} \int dz B^2(z) \tag{6}$$

for the force per unit area of the interface. Substitution into this formula of $B \approx B_c \exp(-z/\delta)$ finally yields

$$\frac{dF}{dxdy} = \eta V, \qquad \eta = \frac{\sqrt{\lambda_L \xi} B_c^2}{2\rho_n c^2}.$$
(7)

As has been explained in the introduction, the crossover from thermal to quantum diffusion of the interface should occur around $T_Q = \hbar U_B / I_{eff}$. With the help of Eqs. (1), (4), and (7) one obtains

$$T_Q \approx \frac{4\pi^2 \hbar \sigma}{\eta L^2} = \frac{4\pi \sqrt{2} \hbar \rho_n c^2}{3\sqrt{\kappa} L^2}.$$
(8)

Notice that due to the dimensionality of the problem T_Q does not depend on the size of the tunneling step a. Recalling that $\lambda_L = [mc^2/(4\pi e^2 n)]^{1/2}$ in terms of the effective mass m and concentration n of the electrons and writing $\rho_n = (m\nu/e^2 n) = 4\pi\nu\lambda_L^2/c^2$ in terms of the normal electron collision frequency ν , the crossover temperature can be presented in the form

$$T_Q \approx \frac{16\pi^2 \sqrt{2}}{3} \kappa^{3/2} \left(\frac{\xi}{L}\right)^2 \hbar\nu \tag{9}$$

that shows its explicit dependence on the microscopic parameters of the material.

 T_Q can be estimated from experiment, based upon the following argument. At finite temperature the magnetic viscosity shown in Fig. 4 has contributions from both, thermal activation and quantum tunneling, $S = S_T + S_Q$, where $S_Q = S(0)$. The parameter T_Q is defined as temperature at which the two contributions are equal, that is, $S_T = S_Q$ and $S(T_Q) = 2S_Q$. This gives T_Q in the ballpark of 4 - 5K. The values of λ_L and ξ in lead are 37nm and 83nm, respectively, giving $\kappa = \lambda_L/\xi = 0.45$. For the energy of the unit area of the interface Eq. (1) with $B_c \approx 800$ G gives $\sigma \sim 0.4 \text{ erg/cm}^2$. Normal resistivity of lead at 4 K is of order²⁶ $5 \times 10^{-11} \ \Omega \cdot m \approx 5.6 \times 10^{-21}$ s. Eq. (7) then gives for the drag coefficient $\eta \approx 0.35 \text{ erg} \cdot \text{s/cm}^4$. We shall now check self-consistency of our model by computing the average size of the tunneling segment L and the tunneling step a. From Eq. (8) one obtains $L \approx 90 \text{ nm} \sim \xi$, which is rather plausible. Indeed, $L \sim \xi$ describes the segment of the interface inside which Cooper pairs are strongly correlated and, therefore, they can collectively participate in a coherent tunneling event. For the tunneling transition to occur in our experimental time window of one hour, I_{eff} cannot significantly exceed $25\hbar$. According to Eq. (4) this condition is satisfied by tunneling steps a below 1 nm, which is also quite plausible. The typical energy barrier, $U_B \approx \pi \sigma a^2$, must be then of order 100K in accordance with the fact that thermal activation dies out below 4 K.

In Conclusion, we have observed non-thermal magnetic relaxation in lead that we attribute to quantum tunneling of small segments of interfaces separating normal and superconducting regions. Theory of such a tunneling has been developed. Comparison between theory and experiment suggests macroscopic quantum tunneling of interface segments comparable in size to the coherence length, by steps of order one nanometer.

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