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Observation of Energy Levels Quantization in Underdamped Josephson Junctions above the Classical-Quantum Regime Crossover Temperature

P. Silvestrini, ^{1,2,*} V. G. Palmieri, ³ B. Ruggiero, ^{1,2} and M. Russo ^{1,2}

¹Istituto di Cibernetica del CNR, I-80072 Arco Felice, Napoli, Italy

²Istituto Nazionale di Fisica Nucleare, Sezione di Napoli, I-80125 Napoli, Italy

³Laboratory for High Energy Physics, University of Berne, CH-3012 Bern, Switzerland (Received 19 May 1997)

We present a clear observation of the presence of energy levels quantization in high quality Nb-AlO_x-Nb underdamped Josephson junctions at temperatures above the quantum crossover temperature. This has been possible by extending the measurements of the escape rate out of the zero-voltage state at higher sweeping frequency (dI/dt up to 25 A/sec) in order to induce nonstationary conditions in the energy potential describing the junction dynamics. [S0031-9007(97)04275-0]

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Quantum mechanics at the macroscopic level is one of the most fascinating topics of fundamental physics. The possibility that a macroscopic physical system obeys the laws of quantum mechanics leads to the need of including in a quantum picture the effect of dissipation and of other thermodynamical variables, such as the temperature. The Josephson effect represents in this contest a very powerful tool to experimentally investigate a variety of interesting phenomena, including macroscopic quantum tunneling (MQT) [1], energy levels quantization (ELQ) [2], macroscopic quantum coherence (MQC) [1,3] of two states, and Coulomb blockade of Cooper pair tunneling [4].

Experiments performed both on current biased Josephson junction [5,6] and rf SQUID [7,8] support the idea of both MQT and ELQ. These experiments measure the escape rate Γ out of the metastable state as a function of the external energy biasing the system, namely, the bias current in the junction and the external magnetic flux in the SQUID. At a temperature below a certain "crossover temperature" T_0 ($T_0 \cong \hbar \omega_i/2\pi k$, where ω_i is the plasma frequency defined below), quantum effects become observable [1]. Previous experiments have shown a saturation of the measured Γ observed both in junctions [5.6] and in SQUIDs [7], as due to MQT, as well as an oscillating behavior of Γ versus the bias energy as due to ELQ, observed in current biased junction [6] in the presence of microwave irradiation, and in SQUIDs [8], in the presence of resonant macroscopic quantum tunneling (RMQT) [9]. Above the crossover temperature any eventual quantum effect is covered by the competitive thermal hopping, so that no clear evidence of a macroscopic quantum behavior at $T > T_0$ has been reported in literature so far. In this Letter, we present a clear observation of quantum effects (ELQ and MQT) in junctions at temperatures well above the crossover temperature. This has been possible by extending, for the first time, the measurements of the escape rate at higher sweeping frequency of the bias current (resulting in dI/dt up to 25 A/sec) in order to enhance nonstationary effects on the population of the levels in the energy potential describing the junction. Under these conditions, the escape process induces a fast reduction of the statistical population trapped in the metastable state and, as a consequence, the occupancy of the energy levels is far from being in equilibrium [10,11]. We wish to stress, however, that from the point of view of quantum mechanics, the potential energy describing the junction is changing adiabatically, since the highest sweep frequency ω used in the experiment is several orders of magnitude smaller than the plasma frequency ω_j , which determines the level spacing $(\omega/\omega_j \cong 10^{-6})$. In our experiment the escape rate is an oscillating function of the bias energy up to high temperatures (1 order of magnitude above the crossover temperature) due to the effect of ELQ [10].

The dynamics of a Josephson junction is described by a macroscopic quantum variable, namely, the phase difference ϕ of the order parameters of the superconductors on the two sides of the barrier. The dissipation of the system is typically described in terms of an effective resistance R of the junction (resistively-shunted-junction model, RSJ) [11]. Within this model the junction dynamics can be described in terms of a mechanical analog, namely, a particle performing its motion in a washboard potential $U(\phi) = -U_0(\alpha \phi + \cos \phi)$, with a friction coefficient $\eta = \hbar^2/\text{Re}^2$. $U_0 = \hbar I_c/2e$ is the Josephson coupling energy, and α is the bias current I normalized to the critical one, $\alpha = I/I_c$. The zero-bias (ω_{i0}) and the current dependent (ω_j) plasma frequencies are defined as $\omega_{j0} = (2eI_c/\hbar C)^{1/2}$, $\omega_j = \omega_{j0}(1 - \alpha^2)^{1/4}$, and C is the junction capacitance. For $\alpha < 1$, $U(\phi)$ shows a series of minima (potential wells) separated by an energy barrier $E_0 = U_0 \{ -\pi \alpha + 2 [\alpha \sin^{-1} \alpha + (1 - \alpha)] \}$ α^2)^{1/2}], which is decreasing with increasing the current. Switching from the V = 0 state to the $V \neq 0$ state in the junction is related to the escape from the well via either MQT or thermal activation. In a quantum picture we must consider the presence of energy levels in the potential well of the washboard potential associated to the Josephson junction. In the weak friction limit the energy levels E_i are sharp and well separated inside the potential well, and the dynamics of the escape process must be

described by the kinetic equation for the probabilities ρ_j of finding the particle at the *j*th energy level [2]:

$$\frac{\partial \rho_j}{\partial t} = \sum_i (w_{ji} \rho_i - w_{ij} \rho_{ji}) - \gamma_j \rho_j, \qquad (1)$$

where w_{ji} is the transition from the *i*th into the *j*th level due to the interaction with the thermal bath, and γ_i is the tunneling probability through the barrier which strongly depends on the energy level position E_i . In the experiments, the bias current is increased with the time at a certain rate dI/dt until a transition out of the V=0 state is observed at a value of I smaller than I_c . This switching current value is a random variable whose probability distribution P(I) is measured by repeating the observation many times. The process is described in statistical terms in Eq. (1). At the initial time the total probability $\rho = \sum_{i} \rho_{i}$ of finding the system in one of the energy levels inside the potential well is $\rho(t = 0) = 1$. However $\rho(t)$ is a decreasing function of time due to the escape process. The escape rate out of the metastable state is defined as $\Gamma(I) = -d \ln \rho / dt$. The presence of quantized energy levels can be observed by a fast sweep of the junction bias. The effect, discussed in some detail in Ref. [10], can be summarized here as follows: During the escape process the population of the upper levels shifts due to a fast tunneling rate, which is high for levels close to the barrier top. This process pushes the system out of the thermal equilibrium. At the same time a diffusion process from the bottom levels towards the top of the barrier refills the upper levels. No quantum effects can be observed for $T > T_0$, as long as the characteristic time of the escape process will be dominated by the thermal diffusion. Increasing the sweeping frequency, which in turn produces a fast reduction of the potential barrier, the escape events will occur at higher bias values with higher escape rate. If the sweep rate is so high that the refilling from the lowest levels by thermal diffusion has not enough time to take place, the upper levels will be depopulated and, once empty, can no longer contribute to the escape. So that until the next level approaches energy values very close to the top, with high tunneling rate, there is a rapid decrease of the decay rate. This occurs periodically as a level is emptied and leads to a distribution modulation. This is a clear manifestation of the presence of quantized energy levels. These oscillations are evident for $T > T_0$. Since the w_{i+1} elements are proportional to effective dissipation, which in turn depends exponentially on temperature, there is an upper limit for T to observe the oscillations due to discrete energy levels, depending on the damping level and on the sweep frequency. Defining the parameter $L = [R(T)C\omega]^{-1}$, where $\omega = (dI/dt)(1/I_c)$, we expect that, for $T > T_0$, a necessary condition to observe experimentally the oscillatory behavior due to discrete energy levels discreteness is L < 200 [10,12]. In our experiment, this condition holds for T < 2 K, while the classical quantum crossover temperature is $T_0 = \hbar \omega_{i0}/2\pi k \approx 200$ mK.

Measurements employed high quality Nb-AlO_x-Nb Josephson tunnel junctions, which exhibited a very low leakage current (Vm > 80 mV), and a quite uniform critical current density J_c . In order to have an extremely low intrinsic dissipation level, a junction with a low critical current density is chosen ($J_c = 53 \text{ A/cm}^2 \text{ at } T = 1.4 \text{ K}$) while the area is $A = 10 \times 10 \ \mu \text{m}^2$. The capacitance C, as measured from the Fiske step voltage and using the penetration depth of Nb film (obtained from the magnetic field diffraction pattern) [11] is $C = 5.5 \pm 0.5$ pF. The sample was mounted on a chip carrier especially designed for the experiment, which contained an integrated high frequency filtering stage and a SMD 87.3 k Ω limiting resistor, both located very close to the junction. All of the connections at room temperature went through coaxial cables with a strong attenuation above 150 MHz. It is of fundamental importance in our experiments to have a very low dissipation level. The quasiparticle resistance $R_{\rm qp}$ at zero bias (V < 5 μ V), as measured by the application of a small magnetic field to suppress the dc Josephson current, depends exponentially on the temperature. This leads to a very low intrinsic damping level at low temperatures ($R_{\rm qp}\cong 100~{\rm k}\Omega$ at $T=1.4~{\rm K}$; $R_{\rm qp}\cong 100~\Omega$ at $T=4.2~{\rm K}$). It is worth stressing that the effective resistance in this kind of experiment may be limited by any external shunting impedance. In our case we had the 87.3 k Ω shunt resistor located close to the junction, while great care has been devoted to avoid stray capacitance, which may reduce the real part of the complex impedance at the plasma frequency, which at zero bias is 170 GHz. The junction was biased through the 87.3 k Ω resistor with a triangular-shaped wave form at frequencies ranging from 100 Hz up to 100 kHz. The switching current distributions are measured by a flight time technique similar to the one described in Refs. [13]. The data as recorded by our multichannel system are histograms showing the number of counts P(K) in the channel K. We then associate a current I(K) to channel K by using the calibrated time interval per channel and the measured current sweep rate dI/dt (to determine the current interval per channel, denoted as ΔI), as well as the measured current corresponding to the first channel I(t=0). We denote by K_{max} the channel corresponding to the highest value of the switching current. From the measured histograms the escape rate Γ is computed according to the formula [14]

$$\Gamma(K) = \frac{dI}{dt} \frac{1}{\Delta I} \frac{P(K)}{\sum_{j=1}^{K_{\text{max}}} P(j) - \sum_{j=1}^{K} P(j)}.$$
 (2)

It is worth noting that P(K) is transformed into $\Gamma(K)$ without the need of any junction parameters, and therefore the two ways of showing the data contain the same information.

In Figs. 1 and 2, we report the experimental current switching histograms measured at (a) T = 4.2 K and (b) T = 1.4 K for two sweeping frequencies: In Fig. 1 = 25 mA/sec (low frequency distributions), while in

Fig. 2 dI/dt=25 A/sec (high frequency distributions). The classical quasistationary histograms of Fig. 1 are fitted with the Kramers theory [15], whose predictions are reported as solid lines. Using a well-established fitting procedure [13], we can obtain from these low frequency distributions the "zero-noise" critical current and the effective resistance values at the two considered temperatures, $I_c=47.0\pm0.3~\mu\mathrm{A}$ and $R\cong100~\Omega$ at $T=4.2~\mathrm{K}$, while $I_c=52.8\pm0.3~\mu\mathrm{A}$ and $R\cong20~\mathrm{k}\Omega$ at $T=1.4~\mathrm{K}$ [16]. The junction capacitance C is independently obtained from the Fiske step voltage, so we know all the relevant junction parameters to compare the high frequency measurements of Fig. 2 directly with the theory: Note that we have $L\cong4350$ at $T=4.2~\mathrm{K}$

(quasistationary limit) and $L \cong 20$ at T=1.4 K (nonstationary limit). In fact the 4.2 K distribution of Fig. 2(a) well fits the Kramers quasistationary theory. This shows that, when increasing the sweep frequency, no significant external noise is introduced. As expected, the histogram in Fig. 2(b), obtained at low temperature and high frequency, presents the typical oscillations due to discrete energy levels. The experimental modulation spacing $(\Delta I \cong 60 \text{ nA}, \text{ corresponding to energy differences between levels <math>\Delta E_j \cong 0.6 \text{ K})$ is in close agreement with the one expected from the macroscopic energy level quantization. This agreement is quite significant. In fact, from a theoretical point of view the levels position can be easily calculated with great precision by using the quasiclassical

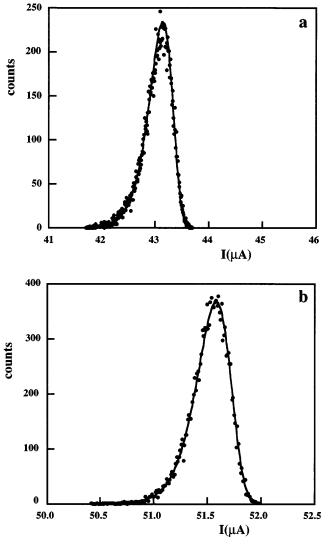


FIG. 1. Experimental histograms (dots) equivalent to the switching current distributions, taken at a sweeping frequency of dI/dt=25 mA/sec, for two different temperatures: (a) T=4.2 K and (b) T=1.4 K. The full lines are the theoretical predictions within the classical Kramers theory [15] with the following junction parameters: (a) $I_c=47~\mu\text{A}$, T=4.2 K, $R=85~\Omega$, and C=5.5 pF; (b) $I_c=52.8~\mu\text{A}$, T=1.4 K, T=

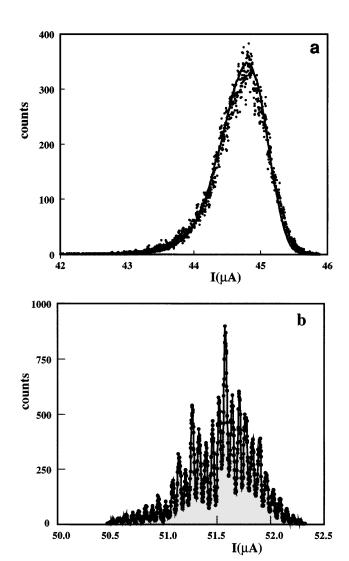


FIG. 2. Experimental histograms (dots) of the switching current at a sweeping frequency of dI/dt=25 A/sec, for two different temperatures: (a) T=4.2 K and (b) T=1.4 K. In (a), L=4350 so that a theoretical fitting within the quasistationary approximation is still possible. The full line is in fact the theoretical predictions within the classical Kramers theory with the following junction parameters: $I_c=47~\mu\text{A},~T=4.2$ K, $R=85~\Omega$, and C=5.5 pF. In (b), L=20 and the histogram presents the oscillations due to discrete energy levels.

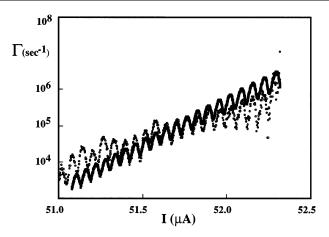


FIG. 3. Experimental data (dots) and theoretical predictions (solid line) within the quantum theory [Eq. (1)] for the escape rate out of the V=0 state, Γ vs I. Data are obtained from the nonstationary distribution shown in Fig. 2(b), by using Eq. (2). The theoretical curve is obtained with the following junction parameters: $I_c=52.8~\mu\text{A}, R=20~\text{k}\Omega$, and C=5.5~pF.

expressions [2]. The resulting modulation spacing of P(I)[10] depends only on I_c and C, mainly as $\sqrt{I_c/C}$, and this ratio is independently determined with a small experimental uncertainty (\sim 5%) [17]. Moreover a quantitative comparison between data and theory can be performed by solving the time dependent Eq. (1) [2,10]. The results are shown in Fig. 3, in terms of the escape rate $\Gamma(I)$. The theoretical curve corresponds to the effective resistance (and I_c) obtained by the fitting of the low frequency distribution reported in Fig. 1(b). The dissipation determines the oscillations amplitude in the escape rate; namely, the higher is the resistance the deeper is the oscillation. Although the oscillation amplitude of data is slightly larger than the expected one, the agreement, seen as a whole, is quite convincing since the absence of any free fitting parameter.

In conclusion, we have observed, in the proper experimental conditions ($L \ll 200$), oscillations in the switching current distributions of an extremely underdamped Josephson junction, whose spacing fits the one expected from the energy levels quantization [17]. As expected, the effect of discrete energy level monotonically decreases with decreasing the sweeping frequency, and at low temperature any oscillation in $\Gamma(I)$ disappears for dI/dt < 2 A/sec. The quantitative agreement between the measured escape rate and the theoretical predictions within the quantum picture of the junction, including MQT and transitions between levels, is also quite convincing. Therefore we can conclude that the experiment provides the first evidence of macroscopic quantum effects above the quantum crossover temperature. Moreover the new experimental idea, as well as the low dissipation level obtained, opens interesting possibilities towards investigations of thermodynamical effects in quantum mechanics at the macroscopic level.

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- *Electronic address: silvestrini@fisps.na.cnr.it
- A. D. Caldeira and A. J. Leggett, Phys. Rev. Lett. 46, 211 (1981).
- [2] A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Teor. Fiz. 91, 318 (1986) [Sov. Phys. JETP 64, 185 (1986)].
- [3] C. D. Tesche, Phys. Rev. Lett. **64**, 2358 (1990).
- [4] K. K. Likharev and A. B. Zorin, J. Low Temp. Phys. 59, 347 (1985).
- [5] S. Washburn, R.F. Voss, R.A. Webb, and S. Faris, Phys. Rev. Lett. 54, 2712 (1985).
- [6] J. M. Martinis, M. H. Devoret, and J. Clarke, Phys. Rev. B **35**, 4682 (1987), and references therein.
- [7] D. B. Schwartz, B. Sen, C. N. Archie, and J. E. Lukens, Phys. Rev. Lett. 55, 1547 (1985).
- [8] R. Rouse, Siyuan Han, and J. E. Lukens, Phys. Rev. Lett. 75, 1614 (1995).
- [9] J. M. Schmidt, A. N. Clealand, and J. Clarke, Phys. Rev. B 43, 229 (1991); P. Silvestrini, B. Ruggiero, and Yu. N. Ovchinnikov, Phys. Rev. B 54, 1246 (1996).
- [10] P. Silvestrini, Yu. N. Ovchinnikov, and R. Cristiano, Phys. Rev. B 41, 7341 (1990); P. Silvestrini, Phys. Lett. A 152, 306 (1991); P. Silvestrini, B. Ruggiero, and A. Esposito, Fiz. Nizk. Temp. 22, 195 (1996) [Sov. J. Low Temp. Phys. 22, 252 (1996)].
- [11] See, for instance, A. Barone and G. Paternò, *Physics and Applications of Josephson Effect* (Wiley, New York, 1982), Chap. 6.
- [12] A. Barone, R. Cristiano, and P. Silvestrini, J. Appl. Phys. 58, 3822 (1985).
- [13] P. Silvestrini, O. Liengme, and K. E. Gray, Phys. Rev. B
 37, 1525 (1988); P. Silvestrini *et al.*, Phys. Rev. Lett. 60, 844 (1988); B. Ruggiero, P. Silvestrini, C. Granata, V. G. Palmieri, A. Esposito, and M. Russo (to be published).
- [14] T. A. Fulton and L. N. Dunkleberger, Phys. Rev. B 9, 4760 (1974).
- [15] H. A. Kramers, Physica (Utrecht) 7, 284 (1940).
- [16] The error bars on I_c are due to systematic errors in the calibration procedure. The best fitting values of the resistance are obtained with large uncertainty.
- [17] The same significant agreement has been found in very recent measurements we have performed in several junctions with different level spacing.