Thermally Activated Phase Slippage in High- T_c Grain-Boundary Josephson Junctions

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The effect of thermally activated phase slippage (TAPS) in $YBa_2Cu_3O_7$ grain-boundary Josephson junctions has been studied. TAPS has been found to be responsible for the dc noise voltage superimposed on the dc Josephson current near the transition temperature. Because of the reduced Josephson coupling energy of the grain-boundary junctions, which is caused by a reduced superconducting order parameter at the grain-boundary interface, TAPS is present over a considerable temperature range. The implications of TAPS on the applicability of high- T_c Josephson junctions are outlined.

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At temperatures near the transition temperature thermal fluctuations can disrupt the coupling of the phases of the order parameter of two superconductors forming a Josephson junction. The phase difference between the two superconductors can thereby slip by 2π resulting in a nonzero time-averaged voltage proportional to the phase-slip rate. The effect of this thermally activated phase slippage (TAPS) on the dc Josephson effect of single overdamped Josephson junctions has been worked out in detail by Ambegaokar and Halperin. ' Because of the short coherence length of the high- T_c oxides the superconducting order parameter can be reduced at interfaces and surfaces.² This results in a reduction of the Josephson coupling energy of high- T_c grain-boundary Josephson junctions and TAPS is expected to be present over a wide temperature range below the critical temperature. For single crystals and epitaxial films the specific properties of high-temperature superconducting oxides, such as the large anisotropy, the short coherence length, the high transition temperature, and the low pinning energies, result in unusually large flux creep and flow effects in the magnetic and transport properties 3.4 and in interesting new features of the vortex behavior.⁵ Similarly, for high- T_c Josephson junctions TAPS will be important for their electromagnetic properties. Moreover, for Josephson-junction devices operating at 77 K the unwanted dissipation due to TAPS will set a lower limit for the maximum Josephson current and hence the device size. In this Letter we report on the experimental study of thermally activated phase slippage in $YBa₂Cu₃O₇$ grain-boundary Josephson junctions. Our experimental results are in good agreement with the Ambegaokar-Halperin (AH) model and demonstrate the importance of TAPS for high- T_c Josephson devices. Some general implications are pointed out.

Recently the AH model was used by Tinkham³ to describe the unusual broadening of the resistivity versus temperature curves of $YBa₂Cu₃O₇$ crystals in a magnetic field. In that work the resistance of the sample was treated as arising from the thermally activated slipping of fluxons over barriers between local minima in the pinning potential of the material. The passage of each vor-

tex thereby gives a phase slip of 2π . It was pointed out that the fluxon movement in the bulk material involves essentially the same physics as the thermally activated slippage of the phase in a single highly damped currentdriven Josephson junction treated by the AH model. Therefore, the AH model could be used as a plausible semiquantitative model for the broadening of the resistivity versus temperature curves of single crystals in a magnetic field. For the high- T_c grain-boundary Josephson junctions the AH model is directly applicable and a quantitative comparison between the experimental data and the model predictions can be made.

The equations of motion for a current-driven Josephson junction of maximum Josephson current $I_c(T)$, tunneling resistance R, and capacitance C_J are $d\theta/dt = 2eV/$ h and $C_J dV/dt = I - I_c(T) \sin \theta - V/R + L(t)$. Here θ is the phase difference of the order parameter, V is the potential difference between the two superconductors forming the Josephson junction, I is the bias current, and $L(t)$ is a fluctuating noise current induced by thermal noise. These equations of motion are applicable only for Josephson junctions smaller than the Josephson penetration depth so that in the absence of external magnetic fields the current is uniformly distributed over the junction area. The movement of the phase θ is entirely equal to the Brownian motion of a particle of mass $M = (h)$ $2e^{\frac{1}{2}C_J}$ in a potential $U = -E_J\{\cos\theta + [I/I_c(T)]\theta\}$, the so-called tilt washboard potential. Here $E_J = \hbar I_c(T)/2e$ is the Josephson coupling energy. For a small tilt angle $(I \rightarrow 0)$ the activation energy U_0 which must be overcome to allow a phase slip of 2π is

$$
U_0 = 2E_J = \hbar I_c(T)/e \tag{1}
$$

and a normalized barrier height for the thermally activated phase slip process can be defined by

$$
\gamma_0 = U_0 / k_B T = \hbar I_c(T) / e k_B T \,. \tag{2}
$$

In the limit of very small transport currents, i.e., very small tilt angles of the washboard potential, the AH model predicts a resistance R_P caused by TAPS of R_P/R $=\{I_0(\gamma_0/2)\}$ ⁻², where I_0 is the modified Bessel function. For the calculation of $R_P(T)/R$ the temperature depen-

FIG. 1. Resistance-vs-temperature curve of a $YBa_2Cu_3O_7$ grain-boundary Josephson junction. In (b), part of the transition curve is shown on an enlarged scale.

dence of I_c has to be known. It is important to note that $I_c(T)$ used in Eqs. (1) and (2) is the critical current without any reduction due to fluctuation effects. Therefore, the measured critical current $I_{cm}(T)$ cannot be used. As a consequence of TAPS, $I_{cm}(T)$ will be zero between T_c and a temperature T_c^{\times} , at which the TAPS causes an average dc voltage as large as the applied voltage limit in the critical-current measurement. Here T_c denotes the critical temperature of the superconductors forming the Josephson junction. Assuming a $(1 - T/T_c)^n$ temperature dependence of I_c , we have

$$
\gamma_0 = C \left(1 - T/T_c \right)^n, \tag{3}
$$

and hence

$$
R_P/R = \{I_0(\frac{1}{2}C(1-T/T_c)^n)\}^{-2}.
$$
 (4)

Figure ¹ shows the resistance-versus-temperature curve of a $YBa₂Cu₃O₇$ grain-boundary Josephson junction in the temperature range near T_c measured with a current of 1 μ A in zero magnetic field. The curve shows a very sharp drop of the resistance starting at $T = 94$ K, which comes from the transition of the grains forming the grain-boundary junction, and a broad foot structure caused by the transition of the grain-boundary Josephson junction. The broadened resistive transition of the grainboundary junction is shown on an enlarged scale in Fig. 1(b). In Fig. 2 the normalized resistive transition

FIG. 2. Experimental $R_P(T)/R$ dependence (diamonds) of a YBa₂Cu₃O₇ grain-boundary Josephson junction and curves computed from (4) for $T_c = 94.0$ K, $n = 2$, and different values of the parameter C.

 $R_P(T)/R$ of the grain-boundary junction (diamonds) is plotted on a logarithmic scale together with the $R_P(T)$ / R dependence calculated for $n=2$, $T_c = 94.0$ K, and for $C=1000$, 1350, and 1800. The experimental data in Fig. 2 have been obtained using a measuring current of 1 μ A for $R_P/R < 0.5$ and 0.2 μ A for $R_P/R > 0.5$. Figure $3(a)$ shows the *I-V* characteristics of the grain-boundary junction at temperatures near T_c . The rounding of the I-V characteristics and their finite slope at zero current due to TAPS are clearly visible. In Fig. $3(b)$ the $I-V$ characteristics calculated according to the AH model for $n=2$, $T_c = 94.0$ K, and $C=1350$ are shown for the same temperatures.

The experimental data shown in Figs. 1-3 have been obtained with a [001]-tilt thin film $YBa_2Cu_3O_7$ grainboundary junction. The misorientation angle between the principle in-plane directions was 24° . The YBaCuO film has been grown epitaxially on a $SrTiO₃$ bicrystal using a laser-ablation technique.^{6} Details of the properties and the fabrication technique of the grain-boundary samples have been reported recently. $7-11$ The zerotemperature critical current density of the sample was 3×10^{7} A/cm² for the grains and 1.6×10^{5} A/cm² for the grain boundary. The other parameters of the grainboundary Josephson junction were $I_c(4.2 \text{ K}) = 2.8 \text{ mA}$, $R=1.0 \Omega$, width $W=5 \mu m$, and thickness $d=0.35 \mu m$. Here I_c and R are directly obtained from the $I-V$ characteristics. The grain-boundary resistance R was constant between 4.2 K and T_c .

Figures 2 and 3 show that there is good agreement between the experimental data and the AH-model predictions both in the low- (Fig. 2) and higher-current regimes (Fig. 3). By fitting both the low- and highercurrent data by the AH model using a single set of parameters, the best fit has been obtained for $C = 1350$, $T_c = 94.0$ K, and $n=2$. No satisfactory fit was possible

FIG. 3. (a) Current-voltage characteristics of a $YBa₂Cu₃O₇$ grain-boundary Josephson junction at different temperatures near the critical temperature and (b) characteristics calculated according to the AH model for the same temperatures using the parameters $T_c = 94.0$ K, $n=2$, and $C=1350$.

for $n \lesssim 1.5$ or $n \gtrsim 2.5$ for varying values of T_c and C. The value of T_c obtained fitting the data corresponds to the onset temperature of the resistive transition in Fig. 1. In Fig. 2 a deviation between the experimental and the calculated data is observed above about $T/T_c \approx 0.98$. This deviation occurs because the grains start to become resistive above this reduced temperature resulting in an upward bending of the experimental R_p/R data. At lower reduced temperatures some deviation is observed approaching the voltage noise level of the experiment. Also in Fig. 3 the largest deviation between the experimental and the calculated I-V characteristics is observed for the lowest temperatures. The deviations between the experimental and the calculated data for $T/T_c \lesssim 0.93$ indicate that the critical current has a $(1 - T/T_c)^2$ temperature dependence only close to T_c and a dependence with an exponent $n < 2$ at lower temperatures.

Figures 2 and 3 show that the behavior of the grainboundary Josephson junctions near T_c is fitted by a thermally activated model with an activation energy $U_0 = (10.8 \text{ eV})(1 - T/T_c)^2$. Moreover, the good agreement of the grain-boundary I-V characteristics at higher currents with those calculated according to the AH model suggests that the AH treatment of the thermally activated phase slippage in a resistively shunted junction describes the behavior of the grain-boundary junction reasonably well. By analogy with the AH model a current parameter $I_{\text{AH}} = eU_0/\hbar = (2.63 \text{ mA})(1 - T/T_c)^2$ can be defined for the regime close to T_c . Interestingly, the value of 2.63 mA agrees well with the experimental value of the critical current of 2.8 mA at 4.2 K.

In the following, we briefly discuss the temperature dependence of the critical current expected for YBa₂- $Cu₃O₇$ grain-boundary junctions near T_c . The measured temperature and magnetic field dependence of the critical current and the observation of microwave-induced voltage steps in the current-voltage characteristics give clear evidence that the $YBa₂Cu₃O₇$ grain-boundary junctions are Josephson-type junctions.⁷⁻¹¹ The question whether these junctions are superconductor-insulatorsuperconductor (SIS) or superconductor-normal-superconductor (SNS) type Josephson junctions, however, has not been answered yet. Recently, we demonstrated that the subgap structures in the current-voltage characteristics of the grain-boundary Josephson junctions are caused by self-excited resonances of the grain boundary.¹⁰ This gives evidence that the grain-boundary junctions are SIS-type Josephson junctions, since the observed electronic excitations of the grain boundary should be strongly damped in a SNS-type junction. Additionally, the fact that the resistance of the grainboundary junction stays constant between 4.2 K and T_c favors a SIS-type junction. According to the Ambegaokar-Baratoff theory, $I_c(T) \sim 1 - T/T_c$ is expected for a SIS junction close to T_c , in contrast to a SNS-type junction, where $I_c(T) \sim (1 - T/T_c)^2$ near T_c .¹² However, because of the short coherence length of the high- T_c materials, the superconducting order parameter Δ can be reduced at a superconductor-insulator interface.² According to the picture of Deutscher and Müller the energy gap at the interface is $\Delta_i(T) \approx \Delta_0(T)$ $\times (1 - T/T_c)^{1/2} \xi(0) / a$, where a is the lattice constant and ξ is the coherence length. For [001]-tilt grain boundaries the relevant coherence length is the $a-b$ plane coherence length (-15 Å) and a is about 4 Å. Using the reduced energy gap in the Ambegaokar-Baratoff expression results in

$$
I_c(T) = [\pi \Delta_t^2(0) / 4eRk_B T_c](1 - T/T_c)^2
$$
 (5)

for T close to T_c . That is, the observed $(1 - T/T_c)^2$ temperature dependence of I_c is also expected for SIStype junctions.

Assuming the Ambegaokar-Baratoff dependence of I_c , the energy gap $\Delta_i(T)$ at the grain-boundary interface is obtained as $\Delta_l(T) = (5.1 \text{ meV})(1 - T/T_c)$. The value of 5. ¹ meV is comparable in magnitude to the directly measured $I_c(4.2 \text{ K})R$ product of 2.8 mV. The typical values of $I_c(4.2 \text{ K})R$ products of the grain-boundary junctions

fabricated by laser ablation range between 2 and 6 mV. Provided that the Ambegaokar-Baratoft theory is applicable, low $I_c R$ values indicate that the energy gap at the grain-boundary interface is considerably reduced for these junctions. The unusual temperature dependence and the small value of Δ_i is difficult to explain. As discussed above the model of Deutscher and Miiller can account for the reduced value and the $(1 - T/T_c)^2$ temperature dependence of Δ_i . However, this model can explain the observed small values of the energy gap only very near to T_c (T/T_c \gtrsim 0.98), since the relevant coherence length for the investigated grain boundaries is the longer a-b plane coherence length and not the very short c-axis coherence length. The deviations between the experimental and the calculated data at lower temperatures in Figs. 2 and 3 indicate that the $(1 - T/T_c)^2$ temperature dependence turns over to a dependence with $n < 2$ for $T/T_c \lesssim 0.93$. At present we do not know how structural disorder, chemical segregation, or deviations from the bulk stoichiometry at the grain boundary influence the value of the energy gap and the coherence length. Probably these phenomena are responsible for a considerable reduction of these quantities at the grainboundary interface, resulting in the observed behavior.

According to Eqs. (4) and (5) the width ΔT of the resistive transition of a high- T_c Josephson junction should scale as

$$
\Delta T/T_c \sim \sqrt{R} k_B T_c / 2\Delta_i(0) \tag{6}
$$

at any constant R_P/R level. For Josephson junctions fabricated of conventional metal-type superconductors, $\Delta T/T_c \sim Rk_B T_c / 2\Delta_0(0)$ is expected. This demonstrates that the larger width of the resistive transition observed for the high- T_c Josephson junctions is mainly caused by the lower Josephson coupling energy resulting from the reduction of the superconducting order parameter at the barrier interface. At the moment we do not have enough data on samples with widely varying resistance values to check the expected ΔT vs R dependence. It should be noted that the result (6) can be viewed as an expression for an "irreversibility line" for high- T_c Josephson junctions of different resistance R in analogy to the definition of the irreversibility line given in Refs. 3 and 4 for bulk material at different magnetic field values. The irreversibility line will certainly depend on the choice of the value R_P/R . Perhaps a reasonable choice might be that the phase-slip rate $d\theta/dt$ below this line shall be small enough to cause a noise voltage smaller than the experimental voltage resolution δV , that is, $(\hbar/2e) d\theta/dt \leq \delta V$.

Finally, we would like to comment on the practical application of high- T_c Josephson junctions. Generally, for most applications of Josephson junctions as SQUID's, digital devices, or microwave mixers the dissipative effects caused by TAPS are unwanted, since they introduce additional noise, which reduces the performance and stability of these devices. The operation temperature of high T_c Josephson devices will most probably be 77 K. Therefore, it is interesting to calculate the dependence of R_P/R on the resistance of the Josephson junction and the value of the reduced energy gap at the barrier interface for this temperature. Provided that the $I_c(T)$ dependence of high- T_c SIS junctions can be expressed by (5) and assuming $\gamma_0/2 > 10$, i.e., $R_P/R \lesssim 10^{-5}$, is a sufficient condition for most applications, the general requirement

$$
R \lesssim 5[\Delta_i(0)/k_B T_c]^2 \ \Omega \quad \text{for} \quad T/T_c = 0.83 \tag{7}
$$

is obtained. Equation (7) demonstrates that TAPS will set a lower limit for the area A of YBaCuO SIS Josephson junctions operated at 77 K depending on their $R \times A$ value. For example, taking $\Delta_i(0)/k_B T_c$ \sim 0.6 as found above and $R \times A \sim 10^{-6}$ Ω cm², which is a typical value for Josephson junctions fabricated of metal-type superconductors, the area of a YBaCuO junction should be larger than about 50 μ m² to have $R_P/R < 10^{-5}$ at 77 K. The dissipative effects due to TAPS at 77 K can be avoided by using materials with a critical temperature well above 77 K, as, e.g., the Tl compound. Using YBa-CuO, low-resistance junctions are necessary and the reduction of the energy gap at the barrier interface should be as small as possible. Grain-boundary junctions represent such low-resistance junctions, For the junction discussed above $(R \times A = 1.75 \times 10^{-3} \Omega \text{ cm}^2, \Delta_l/k_B T_c$ $=0.63$) the criterion (7) results in a minimum area of only 1 μ m².

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