A marked feature characterizing cuprate high temperature superconductors (HTSC) is its strong two dimensionality. In particular the bismuth-based HTSCs, for which this anisotropy is prominent, are best viewed as stacks of superconducting CuO$_2$ planes weakly linked through intrinsic Josephson junction (IJJ) type couplings. These built-in atomic scale links are tailor-made for technical applications difficult to achieve with artificial Josephson junctions (JJ), and its study has now developed into an active interdisciplinary field. We report below what is to our knowledge the first successful observation of the macroscopic quantum tunnelling (MQT) of the phase variable of the superconducting order parameter through the potential barrier of an IJJ, opening up an entirely new direction for HTSC applications. While the corresponding phenomena had been observed at around 300 mK in conventional JJs, we have confirmed MQT behavior at approximately 1 K apparently reflecting the characteristically high plasma frequency of IJJs. Our results are highly nontrivial in that they also demonstrate the feasibility of MQT in spite of the presence of dissipative low energy quasiparticles, which is the other hallmark of HTSCs.

Current-biased JJs offer an ideal stage for realizing a variety of macroscopic quantum phenomena, e.g. energy level quantization within the potential well and the associated MQT and macroscopic quantum coherence, all of which have come to be recognized as having immediate implications for qubit applications. In particular, a phase qubit utilizing MQT has been reported by Martinis and co-workers. Aside from external noises and disorder, a primary source which stands as an obstacle towards observation of MQT is the influence of non-superconducting quasiparticle excitations. In conventional s-wave superconductors, all quasiparticle states are separated from the superconducting ground state by a finite energy gap and thus become essentially inaccessible upon going to sufficiently low temperatures; hence the observability of MQT. The situation is drastically altered when we turn to HTSCs, which are d-wave superconductors. The latter are characterized by four nodes in the order parameter at which the energy gap vanishes. HTSCs therefore necessarily sustain a finite population of dissipative quasiparticles down to the lowest temperatures, which would lead one to render MQT unattainable. While this accounts for the absence of previous phase-MQT reports in HTSCs, several theoretical works have recently put this naive view into question by inferring that dissipation due to nodal quasiparticles is not strong enough to totally destroy the coherence required for MQT. In addition to the moderately weak influence of quasiparticles, it is also crucial from the experimental point of view to have JJs bearing a high degree of underramping and hysteresis, and a measurement system sufficiently detached from the environment. Devising samples and apparatus fulfilling this prerequisite constituted an integral portion of our project of observing MQT in the IJJs of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212).

We now turn to the description of our samples and measurement set-up. In Bi-2212, it is well known that the stacking crystal structure along the c axis consists of an alternating array of the superconducting (S) CuO$_2$ layers and the insulating (I) BiO and SrO layers. The excellent JJ properties exhibited by IJJs in Bi-2212 is by now well established; being ideal atomic sized junctions, they are in particular free of defects and surface roughness of the insulating oxide layers which degrade the quality of artificially made JJs, such as in Al and Nb. To probe the characteristics of the built-in IJJs, it is necessary to construct a pass for the interlayer current flow as illustrated in Fig. 1(a) employing the combination of the double side etching and 3-D focused ion-beam etching techniques. The junction thus fabricated will generally consist of several layers of IJJs coupled inductively and via charging effects –the number of which is technically difficult to control owing to its minute (Angstrom) scale. Therefore, the IJJs form a 1D-
array of S-I-S type JJ. Close to zero bias, the behavior is nevertheless well captured in terms of a single junction model [23], as evidenced from our experiments detailed below. The IJJs were mounted on the copper-mixing chamber of a $^3$He/$^4$He Oxford dilution refrigerator with an excellent thermal contact and cooled down from 4.2 K and 40 mK. All cable connections at room temperature were guided through high impedance cupronickel semi-rigid coaxial cables, which work as a low-pass filter above 1 GHz, to avoid propagating thermal flows and external noises into the sample. Moreover, by using LC low-pass filters along these cables we achieved a strong attenuation of the external noises above 15 MHz. The dilution refrigerator along with the entire analog-measurement system was placed in a shielding room.

The dynamics of a Josephson junction [24, 25] is conveniently described as the motion of a fictitious particle in a washboard potential $U(\phi) = -U_0(x\phi + \cos \phi)$, where the phase difference $\phi$ is to be viewed as the particle coordinate. Here $U_0 = hI_C/2e$ is the Josephson coupling energy and $x = I/I_C$ (I: bias current, $I_C$: critical current). The particle at the bottom of the potential well oscillates at the plasma frequency defined as $\omega_p = \omega_{p0}(1 - x^2)^{1/4}$, where $\omega_{p0} = (2eI_C/hC)^{1/2}$ is the zero-bias plasma frequency and $C$ is the junction capacitance. In the presence of a bias-current, $U(\phi)$ consists of a periodic potential structure separated by the energy barrier $E(x) = U_0\{-\pi x + 2[x\sin^{-1} x + (1-x^2)]^{1/2}\}$. Thermal fluctuations at sufficiently high temperatures assist a thermally-activated escape of the particle across the barrier whose rate is given by

$$\tau_{TA}^{-1} = \frac{\omega_p}{2\pi} \exp\left(-\frac{E(x)}{k_BT}\right),$$

where $k_B$ is the Boltzmann constant and $T$ is the temperature. As $T$ is lowered, thermal activation is suppressed exponentially and the escape process eventually gets dominated by MQT events. This crossover takes place at the temperature $T^* \sim \hbar\omega_p/2\pi k_B$ [12, 27], and the MQT rate is expressed as

$$\tau_{MQT}^{-1} = 12\omega_p \left(\frac{3E(x)}{2\pi \hbar \omega_p}\right)^{1/2} \exp\left(-\frac{36E(x)}{5\hbar \omega_p}\right).$$

The current-voltage ($I$-$V$) characteristic of the IJJs measured by biasing along the $c$ axis (see Fig. 1(b)) at 4.2 K is displayed in Fig. 1(c). The typical $I$-$V$ characteristic of IJJs exhibiting a multi-branch structure was observed. The total number of branches corresponds to the number of elemental junctions in the IJJs as one can confirm by applying a larger bias. When biased, a switching to a finite voltage state takes place at a random value of the switching current $I_{SW}$ and the $I$-$V$ curve shows a hysteresis. The zero-bias Josephson current is slightly larger than the first branch of IJJs, which indicates that the noise level of our system is quite low. In addition, the McCumber parameter $\beta_C = 2eI_CCR_q^2/h$ and the damping parameter $Q^{-1} = (\omega_{p0}CR_q)^{-1}$ were $\sim 10^5$ and $\sim 10^{-2}$, respectively. $R$ is the effective shunt resistance, and $R_q$ is the quasiparticle resistance which is estimated from the slope of the first branch of an $I$-$V$ curve at zero bias. This high degree of hysteresis and the underdamping are crucial for detecting the MQT.

The physical nature of the escape process in JJs can be elucidated by analyzing the statistics of the stochastic switching [4, 12, 24] from a zero-voltage state to a finite voltage state. To this end measurement of $I_{SW}$ was repeated for 2000 times at each temperature with a finite voltage state. To this end measurement of $I_{SW}$ was repeated for 2000 times at each temperature with a constant bias speed $dI/dt$. Histograms of $P(I_{SW})$ were calculated against the bias current and the bin width corresponds to the current resolution $\Delta I$ of the measurement setup. As $T$ is lowered, $P(I_{SW})$ narrows and shifts to higher currents. The $P(I_{SW})$ measured at 4.2 K are fitted with the thermal activation theory proposed by Kramers [28, 29], and the zero-noise critical current obtained from the fitting is $I_C = 48.54 \pm 0.02$ $\mu$A. This value gives a practical estimate of $I_C$ at 0 K. The results exhibit good agreements with the Kramers theory at 4.2 $\sim$ 1.1 K, but deviates substantially below 1K. Note also that below 1K, the amount of shift of $P(I_{SW})$ along the current axis reduces...
considerably. These findings strongly infer that quantum tunneling takes over and the escape process is essentially $T$-independent in this regime as verified below. We also expect that the good fitting of $P(I_{SW})$ in the thermally activated regime to conventional theories is an indicator that the weakest link in our IJJs effectively behaves like a single Josephson junction. Figure 2(b) shows $\tau^{-1}(I_{SW})$ calculated from the experimental data in Fig. 2(a) [12, 27]. As $T$ is lowered, the slope of $ln \tau^{-1}(I_{SW})$ increases and $\tau^{-1}(I_{SW})$ shifts to higher $I_{SW}$ similar to the behavior of $P(I_{SW})$. One observes from a comparison of the two figures that below 1 K, where the amount of shift of $P(I_{SW})$ gets smaller and saturates, the rates $\tau^{-1}(I_{SW})$ overlap with each other. This implies that $\tau^{-1}(I_{SW})$ is also independent of $T$ in this regime. Using the value of $I_C$ obtained from the fitting of $P(I_{SW})$, the tunneling rates are estimated from Eqs. (1) and (2). The fitting of $\tau^{-1}(I_{SW})$ agrees with the experimental data.

The standard deviation $\sigma(T)$ of $P(I_{SW})$ is plotted against $T$ on a double logarithmic scale for two samples in Fig. 3. In the high $T$ region ($1 \sim 4.2$ K), $\sigma(T)$ is proportional to $T^{-2/3}$ as expected, and decreases with decreasing $T$ owing to the suppression of thermal activation. But below 1 K it is saturated and independent of $T$ in accordance with Fig. 2. $T_{esc}$ also exhibits a $T$-independent behavior. (see the inset of Fig. 3) Actually, we have also observed this kind of saturating behavior of $\sigma(T)$ in the IJJs fabricated in Bi-2212 whisker single crystals. These phenomena agree with the tendencies which have been reported for low-$T_C$ JJs [3, 6]. To check that the observed saturation is not due to external noise, we confirmed that connecting $LC$ low-pass filters in series does not effect the value of $\sigma(T)$. Moreover, an intermittent ramp bias was applied to the sample in order to avoid heating. We are thus lead to conclude that the experimental results presented here are direct consequences of MQT of the macroscopic junction phase of IJJs fabricated in HTSC Bi-2212. The effect of MQT appears below the crossover temperature $T^*$, which we estimate from the data to be 0.75 K for $I_C=48.54 \mu A$ and 0.95 K for $I_C=84.92 \mu A$, which are shown in Fig. 3 by the open and filled arrows, respectively. Meanwhile, based on the uncertainties of $I_C$ and $C$, and the bias dependence of $T^*$ as obtained from the formula $T^* \sim h\omega_p/2\pi k_B$ [12, 27] is 0.75$\sim0.80$ K for $I_C=48.54 \mu A$ and 0.85$\sim1.0$ K for 84.92 $\mu A$. These results agree well with the experimental values of $T^*$.

Having established the occurrence of MQT, let us briefly account on its physical implications. We begin by mentioning two factors essential to our findings, i.e. the large magnitude $\omega_p$ of the overall energy scale involved, and the relatively weak form of dissipation with which the quasiparticle transport affects the system. The former aspect is apparently reflected in the thermal-to-quantum crossover temperature, which to the best of our knowledge is the highest reported to date. Though the temperature itself is not the main emphasis here, it does expose a potential advantage enjoyed by HTSC materials, which deserves to be explored further. Meanwhile the latter factor concerns the much more subtle issue of “taming” the nodal quasiparticles mentioned earlier, which brings us back to the $I$-$V$ characteristics displayed in Fig. 1(c). In general, the quasiparticle dynamics in a JJ is manifested in the return curve (the low-voltage subgap portion of the curve following the jump to the first branch, obtained upon switching off the bias current), a regime where Cooper pairs cannot participate in the charge transport. In JJ stacks artificially fabricated using conventional superconductors, this curve exhibits an abrupt drop to nearly zero current at low temperatures [20], signalling a strong suppression of quasiparticle excitations. Meanwhile, the resistively-shunted-
shaded crossover region calculated theoretically. The inset of the measured samples are around 1 K, falling within the crossover temperature $T^*$ the crossover temperature behavior termed the law-like tail seen in our data falls into a novel class of resistance becomes a relevant perturbation. The power-treatments of dissipation in JJs [2] can lead to the dominating processes) of quasiparticle tunnelling between $(e.g.$ transverse-momentum conserving and nonconserving processes) of quasiparticle tunnelling between $d$-wave superconductors, several of which may account for superohmic dissipation. Another related problem—with direct implications to optimizing device geometry—is to resolve how the differences between generic $d$-wave JJ's and the interlayer JJ's would influence MQT experiments. For instance, the absence of zero-energy bound states in [10] and the presence of a bilayer tunnelling matrix element suppressing nodal quasiparticle tunnelling in [31] in the latter can both be considered advantages peculiar to the JJ's

Our study bores out a number of fundamental questions, which reside on the intersection of two major areas of current research: HTSC physics and future quantum technology. For example it provides a fresh perspective on the issue of differentiating between very distinct forms (e.g. transverse-momentum conserving and nonconserving processes) of quasiparticle tunnelling between $d$-wave superconductors, several of which may account for superohmic dissipation. Another related problem—with direct implications to optimizing device geometry—is to resolve how the differences between generic $d$-wave JJ's and the interlayer JJ's would influence MQT experiments. For instance, the absence of zero-energy bound states and the presence of a bilayer tunnelling matrix element suppressing nodal quasiparticle tunnelling in the latter can both be considered advantages peculiar to the JJ's.

In summary we have successfully observed phase MQT in the JJ's of HTSCs, inviting further investigation on coherence-dissipation competition in these systems. Though yet an initial step towards applications, we anticipate that efforts along this line may lead to a rich cross-fertilization between the fields of HTSC science and quantum information.

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Note added- After completion of this work, we learned that Bauch et al. have also observed the MQT at around 40 mK in an HTSC YBa$_2$Cu$_3$O$_{7-\delta}$ grain boundary Josephson junction [32].

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