

Microwave-induced constant voltage steps in surface junctions of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals

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We have observed the zero-crossing steps in a surface junction of a mesa structure microfabricated on the surface of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal. With the application of microwave of frequencies 76 and 94 GHz, the current-voltage characteristics show clear voltage steps satisfying the ac Josephson relation. Increasing the microwave power, the heights of the steps show the Bessel-function behavior up to step number $n=4$. We confirm that the intrinsic surface junction meets the criterion for the observation of zero-crossing steps.

Irradiated with a microwave of frequency f , Josephson tunnel junctions can spontaneously exhibit quantized dc voltages of $V_n = nhf/2e$ in the absence of a bias current, where n is an integer and h is the Planck's constant.¹ In current-voltage (I - V) characteristics, this effect manifests itself as constant voltage steps crossing the zero-current axis.² The occurrence of these voltage steps is a direct consequence of the ac Josephson effect and the phase-coherent pair tunneling in response to an external electromagnetic excitation.³ Since no voltages other than the quantized values V_n are present for zero current bias, Josephson tunnel junctions are ideal as voltage standards which require constant voltage output independent of environmental parameters such as temperature or humidity.⁴ Thus, most Josephson voltage standards currently in use consist of several thousands of Nb/ AlO_x /Nb tunnel junctions connected in series, with each junction exhibiting highly hysteretic I - V characteristics.^{5,6}

The highly anisotropic high- T_c superconductors (HTSC's) with layered structures, such as $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) and $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Tl-2223), can be considered as series arrays of Josephson tunnel junctions along the c axis.⁷ For Bi-2212 single crystals the superconducting order parameter tends to be localized in $\sim 3\text{-}\text{\AA}$ -thick Cu-O bilayers and the transport along the c axis occurs mainly via Josephson tunneling between the neighboring Cu-O bilayers, which are $\sim 12\text{ \AA}$ apart from each other. The c -axis dc I - V characteristics of a Bi-2212 single crystal usually show multiple quasiparticle branches with very large hysteresis, where the number of branches corresponds to the number of intrinsic Josephson junctions (IJJ's) in the crystal.⁷⁻¹¹ In spite of the general consensus for dc I - V characteristics, the microwave response of a stack of IJJ's in HTSC's is unclear. Applied with a microwave, a stack of IJJ's has been observed to exhibit constant voltage steps. However, their values do not satisfy the Josephson frequency-voltage relation and strongly depend on the microwave power.¹² Thus, the authors of Ref.

12 have attributed their results to the microwave-induced phase-locked fluxon motion in a series stack of IJJ's, rather than the ac Josephson effect. Although Shapiro steps or zero-crossing steps are observed in the studies, the measured interval between the voltage steps, ΔV , shows a large difference from the expected value $Nhf/2e$ and depends sensitively on the frequency and power of the applied microwave.^{7,13} Here, N is the total number of IJJ's in a measuring stack. This discrepancy is ascribed to other coupling mechanisms which give rise to the phase locking of IJJ's in addition to the ac Josephson effect.¹⁴ The recent report on the microwave-induced current steps in IJJ's patterned on Bi-2212 single crystal has been interpreted as geometric resonance in the junction cavity.¹⁵

In this paper, we report an observation of clear zero-crossing voltage steps in I - V characteristics of stacks of IJJ's in Bi-2212 single crystals, irradiated with a microwave of frequency $f=76$ and 94 GHz. The voltage steps are believed to be from a single intrinsic Josephson junction formed on the top surface of the stack in contact with a metallic (Au) electrode. The voltage difference between the successive steps coincides with the expected value of $hf/2e$. The magnitude of the voltage steps follows the Bessel-function dependence on the applied microwave power up to the order of $n=4$. This implies that the observed voltage steps are genuine zero-crossing voltage steps. The critical current of the surface intrinsic Josephson junction is significantly suppressed due to the proximity contact to the normal-metal electrode. This severe reduction of the critical current allows one to isolate the microwave response of the "surface junction" from that of the rest of the "inner junctions" in a stack of Bi-2212 single crystals. From our experimental results we were able to confirm the necessary condition for observing zero-crossing steps in an intrinsic Josephson junction of highly anisotropic HTSC's.

Stacks of IJJ's were fabricated on the surface of a Bi-2212 single crystal using photolithography and Ar-ion etching.^{10,11}

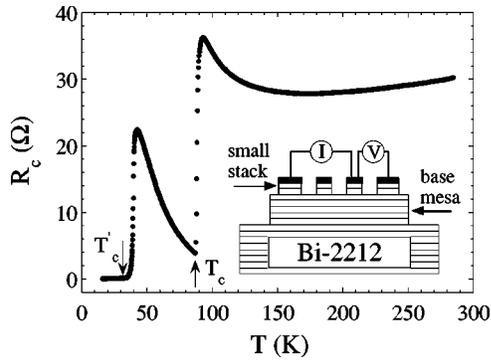


FIG. 1. The c -axis resistance $R_c(T)$ which was obtained using a three-terminal measurement method. The superconducting transition temperature of the inner junction was $T_c \sim 87$ K and the surface junction $T'_c \sim 31$ K layers. The contact resistance is not subtracted. Inset: a schematic configuration of the measurements.

The Bi-2212 single crystal, grown by standard solid-state-reaction method,¹⁶ was glued onto a MgO substrate using negative photoresist and was cleaved using pieces of Scotch tape until an optically smooth surface appeared. A thin (~ 50 nm) Au film was evaporated on top of the crystal immediately after cleaving to protect the crystal surface from any contamination during further fabrication processes. Then a large base mesa ($450 \times 20 \times 1.2 \mu\text{m}^3$) was formed by photolithography and ion-beam etching, using the beam voltage $V_{\text{beam}} = 300$ V and the beam current $I_{\text{beam}} = 0.8$ mA/cm². The ash of photoresist was removed by oxygen plasma etching. To prevent the regions of the specimen other than the top surface of the base mesa from being shorted to the contact electrodes, an insulation layer of photoresist was placed around the base mesa. Then a 400-nm-thick Au film was further evaporated and patterned afterwards by photolithography and ion-beam etching to form electrical extension pads and small stacks ($18 \times 20 \times 0.04 \mu\text{m}^3$) on top of the base mesa. The lateral dimensions of a small stack were determined by the narrow width of the base mesa and the breadth of the electrode, which also acted as a mask for the fabrication of the small stack. The thickness of the measuring stack, which corresponded to the number of junctions in it, was controlled by adjusting the ion-beam etching time. The fabrication procedure was completed by removing the remnant photoresist by oxygen plasma etching. The heat treatment of the specimen was limited to $T < 120$ °C during the entire microfabrication process.

The microwave response of the specimen was measured at $T = 4.2$ K. The microwave generated by a Gunn diode was transmitted through a waveguide and coupled inductively to the specimen placed at $\lambda/4$ distance from the end of the waveguide. The maximum available microwave power was 100 mW for $f = 76$ GHz and 50 mW for $f = 94$ GHz. The power coupled to the specimen was tuned by using a level set attenuator.

Transport measurements were carried out using a three-terminal measurement method (see the inset in Fig. 1). Shown in Fig. 1 is a typical c -axis resistance vs temperature, $R_c(T)$. The resistance shows a weak semiconducting behavior above $T_c \approx 87$ K, indicating that the crystal is in a slightly overdoped regime. One also notices that the resistance remains finite below T_c with a secondary peak appearing far

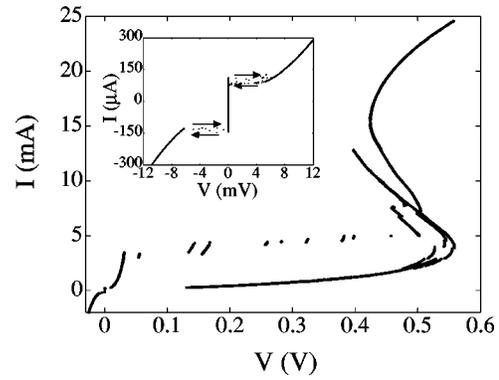


FIG. 2. The I - V characteristics of the specimen at $T = 15$ K without external microwave irradiation. Inset: magnified view of the low bias region, showing clear hysteresis of the surface junction.

below T_c . It is attributed to a weak intrinsic Josephson junction formed at the surface of a measuring stack in contact with a Au normal-metal electrode.^{10,11} The superconductivity of the topmost Cu-O bilayer of a stack is suppressed by the proximity contact to a normal-metal (Au) pad rather than by a degradation effect of the surface layer.¹⁰ Thus, the surface Cu-O bilayer has a superconducting transition temperature $T'_c \approx 31$ K far below the bulk T_c . In the temperature range of $T'_c < T < T_c$, the surface junction can be considered as a normal-metal/insulator/ $d_{x^2-y^2}$ -wave-superconductor (NID) junction¹⁷ consisting of the surface Cu-O bilayer in the normal state and the adjacent inner bilayer in the superconducting state. Thus, $R_c(T)$ corresponds to a quasiparticle tunneling resistance of the NID junction with a junction resistance¹⁰ $R'_n = R_c(T_c) = 3.9 \Omega$. As the surface junction becomes Josephson coupled below T'_c , $R_c(T)$, which is essentially the contact resistance between the Au pad and the topmost Cu-O bilayer, becomes less than 40 m Ω in our specimen.¹⁸

Figure 2 shows the I - V characteristics of IJJ's in a stack below T'_c in the absence of an external rf field. With increasing bias current just above the critical current of each intrinsic Josephson junction in a stack, periodic voltage jumps occur in units of $V_c \approx 23$ mV, and the I - V curves show highly hysteretic behavior. Although not apparent in the figure, the number of quasiparticle branches in the I - V characteristics indicates that 28 IJJ's are contained in the measuring stack. The average critical current I_c is about 4.5 mA and the normal-state junction resistance of the inner junctions, estimated from the linear portion of the I - V curves, is $R_n = 0.7 \Omega$. The inset of Fig. 2 shows the enlarged view of the I - V curves in the low bias region. One notices that the weak surface junction shows a much smaller critical current $I'_c \approx 130 \mu\text{A}$ with clear hysteresis. The reduced critical current of the surface junction, compared to the ones of the inner junctions, is due to the suppressed superconductivity of the surface layer. This result is consistent with the finite-resistance behavior of the $R_c(T)$ curve in Fig. 1.^{10,11}

Figure 3(a) shows the I - V characteristics of the specimen with the application of a microwave of frequency $f = 94$ GHz. Clearly seen are the two steps of height $\Delta I_1 = 120 \mu\text{A}$. The voltage difference between the two steps is about $\Delta V = V_1 - V_{-1} = 400 \pm 20 \mu\text{V}$ in agreement with the expected

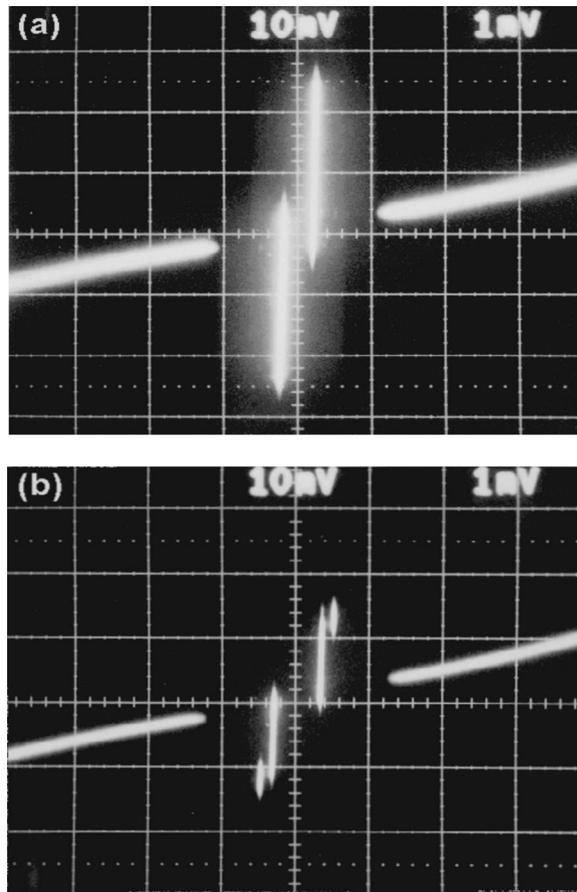


FIG. 3. The I - V characteristics showing zero-crossing voltage steps with application of a microwave of (a) $f=94$ GHz and (b) $f=76$ GHz at $T=4.2$ K. Each division in the vertical axis is $40 \mu\text{A}$ and in the horizontal axis is 1 mV .

value of $\Delta V=2hf/2e=389 \mu\text{V}$, implying that these steps are genuine zero-crossing steps corresponding to the step number $n=\pm 1$. Due to the weakness of the transmitted microwave power, we could not observe other steps of $n>3$ for $f=94$ GHz.

Shown in Fig. 3(b) are the I - V characteristics of the same stack at a microwave frequency $f=76$ GHz. Compared to the case of $f=94$ GHz, the height of the $n=1$ step becomes reduced while the steps of higher orders $n=\pm 2, \pm 3$ are seen more clearly. The steps of $n\geq 3$ do not cross the zero-current line, possibly due to a large leakage current. The remarkably large leakage current for the c -axis tunneling in Bi-2212 HTSC's is attributed to the existence of the gapless node for the $d_{x^2-y^2}$ -wave order parameter. By increasing the microwave power, we were able to identify other voltage steps up to the order of $n=4$. Further increase of the microwave power caused a noticeable slope in the voltage steps, possibly due to chaotic switching of the surface junction between a Josephson-tunneling state and a resistive one.¹⁹ Once any of the Josephson junctions in the stack becomes resistive, all the steps are bound to exhibit a finite resistive slope, making it difficult to identify the steps at high bias voltages.

The Josephson penetration depth in the surface junction is estimated¹⁴ to be about $2.3 \mu\text{m}$ in the thin-electrode limit ($0.5 \mu\text{m}$ in inner junctions). Since the lateral dimension of

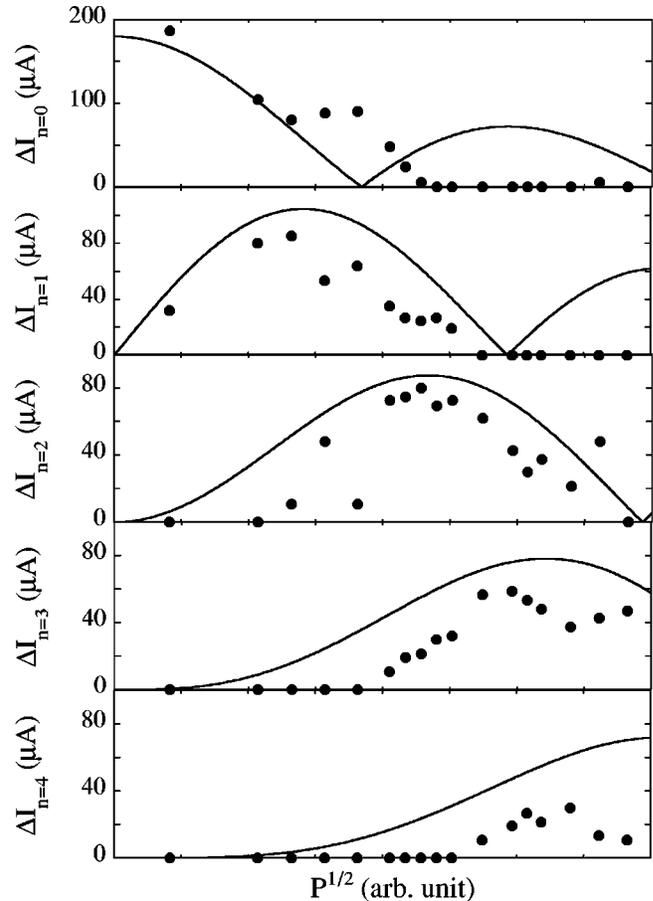


FIG. 4. Measured step heights, from the step number $n=0$ to 4, as a function of the square root of the microwave power for $f=76$ GHz. The solid lines are fits to $\Delta I_n = I'_c |J_n(I_{ac} f'_c / I'_c f)|$ with I'_c (4.2 K) = $180 \mu\text{A}$ as a fitting parameter.

our stack is about $20 \mu\text{m}$, the surface junction is in a long-junction limit. But we did not observe the microwave-induced dissipative flux-flow branches¹² in the current-voltage characteristics of our stack. The observed zero-crossing steps cannot be attributed to the collective resonance of Josephson fluxons excited by the microwave, because the position of voltage steps has no microwave power dependence. They are not caused by the geometric resonance of the junction cavity,¹⁵ either, because the spacing of voltage steps depends only on the microwave frequency in our experiment. Thus, we exclude the possibility of the formation of Josephson fluxons in the surface junction.

Figure 4 shows the measured step heights as a function of the square root of the applied microwave power, $P^{1/2}$. Varying the step order from $n=0$ to 4, the measured step heights are in qualitative agreement with the relation of $\Delta I_n = I'_c |J_n(I_{ac} f'_c / I'_c f)|$, where I'_c is the critical current of the surface junction at $T=4.2 \text{ K}$, J_n the n th order Bessel function, I_{ac} the applied rf current, and $f'_c = 2eI'_c R'_n / h$ the characteristic frequency of the surface junction.²⁰ We obtained the fitting parameter I'_c (4.2 K) to be $180 \mu\text{A}$, which corresponds to the critical current density of 50 A/cm^2 . This value is consistent with the ones observed in the surface junctions of other specimens at 4.2 K .^{10,11} To reveal the Bessel-function behavior, a Josephson junction is required to satisfy the condition¹⁹ of $\Omega^2 \beta = (f/f_p)^2 \gg 1$, where $\Omega = f/f_c$ is the

TABLE I. The junction parameters of the *surface* junction and the *inner* junctions for our specimen and those of Bi-2212 single crystals and Nb/Al₂O₃/Nb used in other works. Variables are defined in the text.

	J_c (A/cm ²)	f_p (GHz)	f (GHz)	f/f_p	$\Omega^2\beta$
Surface Junction	50	22	76	3.5	12.3
Inner Junction	1200	110	76	0.69	0.48
As-grown ^a	2200	150	12	0.08	0.006
Ar-annealed ^b	150	40	3.1	0.078	0.006
O ₂ -annealed ^b	1050	100	3.2	0.032	0.001
Air-annealed ^c	650	80	31	0.39	0.15
Nb/Al ₂ O ₃ /Nb ^d	33	21	70	3.3	10.9

^aReference 12.

^bReference 7.

^cReference 13.

^dReference 6.

frequency reduced with the characteristic frequency of a junction, $\beta = 2eI_c R_n^2 C / \hbar$ the hysteresis parameter,²⁰ and $f_p = \sqrt{eI_c / \pi \hbar C}$ the Josephson plasma frequency. C is the capacitance of the Josephson junction. This criterion is not satisfied with the *inner* IJJ's in the stack but is satisfied with the *surface* junction (see Table I). Thus we infer that the observed zero-crossing steps should originate from the weak surface junction. As shown in Table I, specimens used by other groups concerning the microwave-induced fluxon motion¹² or collective behavior of the IJJ's^{7,13} do not meet the above criterion. Nonetheless, one can notice that the typical parameters of Josephson junctions currently used for Nb-based voltage standards⁶ are similar to the ones of the surface junction. Although the observing condition¹⁹ for the zero-crossing steps was originally proposed for Josephson tunnel junctions made of conventional superconductors, our

results indicate that the IJJ's in HTSC's provide high potential for observing the same phenomenon.

The observing condition for the zero-crossing steps can be rewritten as $\Omega^2\beta = (\pi\hbar/e)(\epsilon f^2/dJ_c) \gg 1$, where ϵ is a dielectric constant of the blocking layer between adjacent conducting bilayers, d an interbilayer distance, and J_c a critical current density of the intrinsic Josephson junction. For our specimen in this study, the critical current density of the inner junctions is 24 times larger than that of the surface junction. An inner junction thus has the Josephson plasma frequency f_p about five times larger than that of the surface junction. IJJ's with larger critical current densities require higher microwave frequencies and higher power to produce stable zero-crossing steps. Reducing the tunneling critical current density is, therefore, required to obtain the stable voltage steps from the inner IJJ's in a stack. In addition, to prohibit any nonuniform rf-current flow into the junction, which becomes more probable at higher microwave frequencies, one needs to reduce the junction size. These requirements may be fulfilled with ultrasmall IJJ's in Bi-2212 single crystals²¹ or with IJJ's in Bi-2212 single crystals intercalated with guest molecules such as HgI₂ or HgBr₂.²²

In summary, we have studied the inverse ac Josephson effect from an intrinsic Josephson junction located in the surface of Bi-2212 single crystals irradiated with the external microwave of $f = 76$ and 94 GHz. The surface weak Josephson junction shows clear voltage steps satisfying the ac Josephson relation and the step heights follow the Bessel-function behavior with increasing microwave power, up to the step number $n = 4$. Our results indicate that the intrinsic Josephson junctions in highly anisotropic HTSC's with very low tunneling critical current density may be a promising candidate for the observation of zero-crossing steps.

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