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#### Broadly tunable, high-power terahertz radiation up to 73 K from a stand-alone Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub> mesa

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High-power, continuous, broadly tunable THz radiation from 0.29 to 1.06 THz, was obtained from the outer current-voltage characteristic (IVC) branch of a single stand-alone mesa of the hightransition temperature  $T_c$  superconductor Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+ $\delta$ </sub>. The particular metallic film structures placed both beneath and atop the mesas resulted in more efficient heat dissipation, higher allowed applied dc voltages, larger IVC loops, wider emission temperature ranges, and much broader emission frequency tunability than obtained previously. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4902336]

Since 2007, when intense and continuous THz radiation was first found to arise from a large mesa structure of the high transition temperature  $T_c$ superconductor  $Bi_2Sr_2CaCu_2O_{8+\delta}$  (Bi2212),<sup>1</sup> a great deal of interest in THz radiation from high- $T_c$  superconductors has focussed on this new compact source as a possible candidate to cover the THz gap.<sup>2</sup> From the accumulating results of intensive experimental and theoretical studies, the fundamental nature of this radiation has been revealed: For high-power radiation, at least two conditions upon the emission frequency f must be satisfied. First, f must satisfy the ac-Josephson relation  $f = f_{\rm J} =$ 2ev/h operating within the individual intrinsic Josephson junctions (IJJs) existing in the single crystal of Bi2212,<sup>3</sup> where e is the electronic charge, h is Planck's constant, and v = V/N, where V is the dc voltage across the N IJJs in the mesa. Second, the output power can be greatly enhanced if  $f = f_{m,n}^{c}$  also satisfies an electromagnetic (EM) cavity resonance condition associated with forming EM standing waves appropriate for the geometrical shape of the thin mesa structure.<sup>1,2,4,5</sup> For a thin rectangular cavity of length  $\ell$  and width w, the transverse magnetic (TM) mode resonance frequencies are  $f_{m,p}^{c} = c_0 \sqrt{(m/w)^2 + (p/\ell)^2/(2n)}$ , where *m* and *p* are positive integers,  $n \approx 4.2$  is the refractive index of Bi2212, and  $c_0$  is the speed of light in vacuum. Most often, the lowest  $f_{m,p}^{c}$  observed has been  $f_{1,0}^{c}$ , but lower  $f_{m,p}^{c}$  values were observed using either nearly square or stand-alone mesas,<sup>6</sup> the latter similar to those described in the following.

To obtain 1 THz emission amplified by resonance with the TM(1,0) mode, a rectangular mesa should have  $w \sim 35 \,\mu\text{m}$ . However, it has not yet been possible to generate frequencies above 1 THz by simply reducing the width of the mesa. This is due to the primary  $f = f_{\rm I}$  emission condition, whereby it is necessary for  $v \sim 2.07 \,\mathrm{mV}$ , or that V = 2.77 V for a Bi2212 mesa 2  $\mu$ m thick, corresponding to 1340 IJJs.<sup>7</sup> In previous studies, the strong Joule heating associated with larger current I and V values led to dramatic back-bending behavior in the current-voltage characteristics (IVCs), preempting such large V values.

Another possible route to attain higher radiation frequencies is to study the inner branches of the IVCs, because the reduced effective N values in the inner branches led to higher v values at fixed V.<sup>6</sup> Here, the "inner branch" means that when the mesa is current-biased not all the IJJs stacking along the *c*-axis are in general resistive, i.e., a part of IJJs is still in the resistive state, so that the IVC in such a incomplete resistive state lies inside as a "inner branch" of the full resistive state. The "outer branch" on the other hand, stands for the full resistive state, where all IJJs in the mesa are equally biased. However, for mesas simply cut from a Bi2212 single crystal, the very low thermal conductivity of the Bi2212 substrate always leads to large Joule heating effects, limiting V.

Here, we report a stand-alone mesa fabrication procedure that greatly improves the thermal contact of the Bi2212 mesa with the thermal bath.<sup>4,8,9</sup> With such stand-alone mesas, it is possible to dramatically increase the applied Vand the corresponding IVC loop region up to  $\sim 5 \text{ V}$ , three times the highest previously attained values,<sup>4,6</sup> and hence to produce powerful emission that was tunable from 0.29 to 1.06 THz ( $\pm 57\%$  from the midpoint), much larger than the previous record tunability range from 0.495 to 0.934 THz  $(\pm 31\%$  from the midpoint) attained from an acute isosceles triangular Bi2212 mesa.<sup>10</sup> This tunability was attained by varying the bath temperature T over the largest emission range yet reported,  $10 \text{ K} \le T \le 73 \text{ K}$ .

A thin plate-like large single crystal of Bi2212 is cleaved from a boule grown by the traveling solvent floating zone method<sup>11</sup> and is glued onto a 0.5 mm thick sapphire substrate with Ag paste. Rectangular mesas are fabricated with standard metal masks and Ar ion milling.<sup>4,12</sup> The mesas are then removed from the substrate with acetone and an ultrasonic device. Immediately after repasting them upside down with Ag onto the substrate and cleaving them with scotch tape, 90 nm Ag and 10 nm Au are successively evaporated onto the surfaces. After again removing the mesas with acetone and ultrasound, the sapphire substrate is successively coated with 10–20 nm Ti, 90 nm Ag, and 10 nm Au. The mesa Au surface is pressed and fixed to the top Au sapphire substrate surface. Finally, 90 nm Ag and 10 nm Au are successively deposited on the mesa top, and a Au wire electrode is attached thereupon by Ag paste.

We confirmed the reproducibility of the IVCs and radiation characteristics of the two nearly identical mesas we fabricated. Their dimensions measured by atomic force microscopy are top and bottom widths  $w_t = 74 \,\mu\text{m}$  and  $w_b = 84 \,\mu\text{m}$ , length  $l = 280 \,\mu\text{m}$ , and thickness  $t = 1.9 \,\mu\text{m}$  for sample #1; and top and bottom widths  $w_t = 75 \,\mu\text{m}$  and  $w_b = 88 \,\mu\text{m}$ , length  $l = 290 \,\mu\text{m}$ , and thickness  $t = 2.0 \,\mu\text{m}$  for sample #2. An optical micrograph of sample #1 is shown in the inset of Fig. 1(a).

The temperature T dependence of the c-axis resistance  $R_{c}(T)$  of sample #1 exhibiting its superconducting transition at  $T_c = 78$  K is shown in Fig. 1(a). This is typical for slightly underdoped Bi2212. Figure 1(b) displays the IVCs obtained at 10 K for sample #1. In contrast to previous studies,<sup>1,2,4–6,9</sup> the IVCs exhibit the remarkably high V region exceeding 5 V and very little back-bending in the higher I region, except for the characteristic jump from 14.8 to 10.5 mA that is often associated with the formation of a hot spot.<sup>13–15</sup> The anomalously large V > 5 V exerted across the relatively thin stand-alone mesa with  $t \approx 2 \,\mu m$  (~1340 junctions) gives rise to the very large  $v \sim 3.73 \text{ mV}$  across each IJJ. This value is several times larger than those obtained in all previous mesas. Since  $f = f_{\rm I} = 2ev/h$ , this higher v applied across the stand-alone mesa IJJs can potentially generate higher f EMwaves. This is indeed observed with sample #1 at 10K as shown in Fig. 1(c), where the emission peaks at 1.04 THz and 1.06 THz are obtained with 2.725 V and 2.808 V, respectively. Such high f emission was not attained in any previous study except in a single case observed previously.<sup>4</sup> The T dependence of the emission f of sample #1 was measured from 10 K to 73 K in 5 K increments. The highest f = 1.06 THz of this monochromatic radiation was observed at 10 K, and f



FIG. 1. (a)  $R_c(T)$  of sample #1 with  $T_c = 78$  K. Inset: an optical micrograph of sample #1. (b) A typical IVC of sample #1 at 10 K. The arrows indicate the cycle directions. (c) Spectral emission intensities measured by the FT-IR spectrometer at 10 K. The highest frequencies of 1.06 and 1.04 THz are detected at V = 2.808 and 2.725 V, respectively.



FIG. 2. Outer return branch of the IVCs of sample #1 measured from 10 K to 70 K in 5 K increments. The intensity of the THz radiation is proportional to the Si-bolometer output *V* as indicated in the right color scale.

decreases gradually as T is increased [see Fig. 4(b)]. This tendency agrees well with previous results.<sup>4</sup>

The overall T dependence from 10 K to 70 K of the outer return branch of the IVCs of sample #1 is shown in Fig. 2. The radiation intensity detected by the Si-bolometer in the region enclosed by the fishtail-shaped dotted blue curve is indicated on each IVC curve by the right color-coded output voltage. THz radiation occurs in Fig. 2 only below 17 mA for all T values studied. Both the jumps in the outer branch IVCs seen in the high V bias region between 4.2 and 5.5 V corresponding to T values between 10K and 45K and the jump from 4.8 V to 5.5 V in Fig. 1(b) are due to the disappearance of the hot spot.<sup>13–15</sup> The magnitude of the jump decreases systematically with increasing T up to 45 K, and completely disappears above 50 K. The hot spot, defined as the local  $T(\mathbf{r}) > T_c$  due to excessive Joule heating, has recently been an important issue, since it was claimed to be important for the mechanism of the synchronization of the THz radiation.<sup>13,16–21</sup> However, the results shown here as well as those presented previously<sup>14,15,22</sup> clearly show that the intensity of the THz emission is strongest in the higher T regions above 50 K for which there is no evidence for either IVC back-bending or a hot spot, in agreement with previous experiments, two of which directly measured the mesa  $T(\mathbf{r})$ maps.<sup>14,15,22</sup> Those authors noted that the hot-spot formation is unlikely to be related to the origin of the THz radiation.

In Fig. 2, there is a remarkable step structure that is enclosed by the dotted red ellipse for  $55 \text{ K} \le T \le 65 \text{ K}$ . Although it is sometimes unstable, the step structure is always observed in the outer IVC branch region where the highest emission power occurs.

Such IVC step behavior is also seen in the inner branch region of these stand-alone mesas, as shown in Figs. 3(a) and 3(b), where the step region is indicated more clearly by the red arrows in a further expanded scale at T = 65 K. Figure 3(c) displays the Si-bolometer output voltages as a function of the dc V applied across the mesa corresponding to Fig. 3(b). Note that in this high T regime, there is no evidence for hot spot formation. The radiation power P gradually decreases with the number N of IJJs as the IVC goes from right to left, as indicated by the yellow curved arrows in Figs. 3(a) and 3(b), even if there are pronounced step structures in the IVCs. This diminishing behavior of P in the inner



FIG. 3. Radiation characteristics of the inner IVC branches measured at 65 K, where the step structures are pronounced. (a) The well-separated inner branches are seen clearly. The output power is shown on the data points in color as indicated in the color code at the right, in which the radiation intensity near or above 0.1 V in output voltage of the Si-bolometer is shown in red, while no radiation is shown in black. In the inset, the overall multi-branching behavior of the IVCs at 65 K is presented. (b) The IVC curves in a further expanded scale and (c) the radiation intensities corresponding to the IVC curves shown in (b). The outer-most branch in (a) is shown by the black dots. In (b) and (c), the contact resistance is subtracted to estimate the power.

branches is simply explained by  $P \propto N^2$ , consistent with the first study.<sup>1</sup> Not only N but also the step structures are strongly related to the radiation P, because it seems that these step structures are more pronounced in the sample with stronger radiation intensity. This may be natural since the excess dc power fed into the mesa is converted into THz radiation power, so that extra power has to be supplied to the system. In estimating the radiation power, we assume that in the absence of THz emission, the IVC curve should decrease smoothly without the step anomaly as V is reduced.

The radiation power can then be calculated from the excess I and V appearing in the step anomaly of the IVC



FIG. 4. Radiation intensity from a single stand-alone mesa as functions of (a) T and (b) f. The increment in T between 45 K and 73 K is 1 K.

curve, assuming all the dc power is converted to THz radiation. The excess *I* is thus estimated to be  $\Delta I \sim 0.4$  mA at *V* of the emission peak  $V_p \sim 1.25$  V. Therefore, the emission power *P* can be roughly estimated as  $P \sim \Delta I \cdot V_p \sim 0.5$  mW. However, the measured radiation power is estimated in the following to be  $\approx 10 \ \mu$ W, which is at least an order of magnitude smaller than the above estimate. We also note that in spite of having a clear step in the IVC curve, sometimes reduced or even vanishing radiation power is observed. Therefore, although this step anomaly can be considered to be an indicator of THz emission, the emission power extracted from the mesa may not fully reflect the total energy fed into the system measured by the IVC curve. This phenomenon should be included in future studies of the fundamental aspects of the THz radiation mechanism.

Figure 4(a) summarizes the T dependence of the maximum output power from 10K to 73K, as detected by the Si-bolometer. As T increases from 10K, the outer-branch emission power gradually increases, jumps suddenly at 54 K, has a peak at 56 K, and decreases sharply above 70 K, until the radiation is no longer observed at 74 K. This behavior is consistent with recent results.<sup>22</sup> Those results were obtained from both high and low I bias regions, while our results are obtained only from the low I bias region. In order to estimate the integrated output power in the same manner as Sekimoto et al. did using the Si-bolometer.<sup>22</sup> The averaged intensity is  $V_{det} = 78 \text{ mV}$  at the detecting solid angle of  $6.43 \times 10^{-3} \text{ sr}$ . This corresponds to the incident power  $P_{det} = 2\sqrt{2}V_{det}/\alpha = 0.020 \,\mu\text{W}$ , where the system optical responsivity  $\alpha = 11 \text{ mV/nW}$ , assuming that the attenuation factor inside the Si-bolometer is 0.25. Integrating the output voltage measured by 5°, and taking account of the radiation pattern and the attenuation at the windows of the cryostat and the Si-bolometer, the total output power is estimated to be  $\sim 10 \,\mu$ W. Since the strongest radiation is observed at 56 K, which is two times as high as that at 65 K, the maximum output power of this sample is therefore estimated to be about 20  $\mu$ W. We emphasize that the THz radiation power has a higher intensity in the higher T region, shows a broad maximum between 55 K and 65 K, and decreases sharply as T approaches a few K below  $T_c = 78$  K. The details of such

THz radiation intensity behavior appear difficult to understand and remain a subject for future study.

The radiation frequencies measured by the FT-IR spectrometer at various T values from 10 K to 73 K are summarized in Fig. 4(b). The frequency resolution of the FT-IR spectrometer is 7.5 GHz. The vertical axis in Fig. 4(b) indicates the emission intensity proportional to the output radiation power on a logarithmic scale. We note that the sensitivity of the spectrometer to the emission from the slightly underdoped Bi2212 depends upon the measured f, as for other detectors.

While the lowest f measured is 0.29 THz detected at 73 K, the highest f is 1.06 THz obtained at 10 K. The f values measured at the peak of the radiation power ranging between 0.47 THz and 0.49 THz excellently agree with the fundamental TM(1,0) mode frequency 0.483 THz calculated from the top width of the mesa. The maximum power between 0.47 THz and 0.49 THz is estimated to be  $\sim 20 \,\mu\text{W}$ . This sharp intensity peak shown in Fig. 4(b) most likely represents the cavity resonance effect, which seems to work efficiently in this case. It is interesting to note here that the measured f spectrum of the THz radiation, although not continuous probably due to experimental constraints, is the most widely tunable range yet measured in Bi2212 mesas, spanning the range of 0.29–1.06 THz generated by only a single stand-alone mesa structure. Furthermore, we point out that the rather weak radiation found around 1.05 THz corresponds closely to that expected for the fundamental TM(2,0)mode at  $f_{2,0}^c$ , judging from the double junction voltage v from that of the TM(1,0) mode.<sup>24</sup>

In summary, we investigated the THz radiation from stand-alone mesas made with metallic surfaces both on their tops and bottoms from a high- $T_c$  superconducting Bi2212 single crystal and pressed onto a trimetallic film atop sapphire substrates. The metal films on both mesa surfaces result in significantly improved thermal contact of the mesa to the thermal bath. As a result, the I-V characteristics exhibit larger hysteresis and larger voltages per intrinsic Josephson junction, leading to higher frequency radiation up to 1.06 THz. This is the highest frequency ever reported to date from this type of emitter. The characteristic features of the THz radiation spectra are the widest range of emission temperatures, between 10 and 73 K, and of frequency tunability, between 0.29 and 1.06 THz, which is  $\pm 57\%$  from the midpoint value. Although the radiation frequency decreases as the temperature increases as reported previously,4,17,23 the radiation intensity increases with increasing temperature, where hot spots are not evident. These remarkable improvements in the THz radiation properties using this type of stand-alone mesa should be very useful for the operation of the compact THz source, and suggest that operation temperatures up to 77 K using liquid nitrogen might be attainable for practical applications.

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