Evidence for a Dual-Source Mechanism of THz Radiation from Rectangular Mesas of Single Crystalline Bi$_2$Sr$_2$CaCu$_2$O$_{8+d}$ Intrinsic Josephson Junctions

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The THz radiation emitted from rectangular mesas of single-crystal Bi$_2$Sr$_2$CaCu$_2$O$_{8+d}$ was studied using angular distribution measurements and Fourier transform infrared spectroscopy. Unlike the recent theoretical predictions, the results provide strong evidence for a dual-source mechanism in which the uniform and non-uniform parts of the ac-Josephson current act as electric and magnetic current sources, respectively. The latter synchronizes with cavity modes of the mesa with integral harmonics of the fundamental radiation.

Keywords: THz radiation, THz emitters, intrinsic Josephson junctions, Josephson plasma, Bi$_2$Sr$_2$CaCu$_2$O$_{8+d}$ single crystals, ac-Josephson effect, high-$T_c$ superconductors, continuous THz sources

The recent discovery of coherent electromagnetic waves at terahertz frequencies (1 THz = 10$^{12}$ Hz) from the intrinsic Josephson junctions (IJJs) within a mesa fabricated from high-quality single crystals of the high transition temperature $T_c$ superconductor Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO), presently denoted as STAR (Stimulated Terahertz Amplified Radiation) emitter, has triggered a great deal of research interest. Besides fundamental studies of the physics and chemistry, there has been a great interest in the development of possible compact and all-solid-state THz radiation sources, which may be very useful for applications in science and engineering fields such as medicine, diagnostics, pharmaceutical development, biotechnology, ultrahigh-speed communication, environmental studies, security issues, and various types of nondestructive and noninvasive sensing and imaging, etc. Some basic features of the STAR emitter were previously reported, but the emission is stable, continuous and powerful ($\sim \mu$W), and its spectral width is less than the resolution limit of the Fourier transform infrared (FTIR) spectrometer, 7.5 GHz. By varying the applied dc voltage $V$ across the stack of $N$ IJJs, the frequency of the THz radiation appears to be locked by cavity resonance mechanism, which fixes it to that of one of the standing wave modes inside the mesa. However, the precise nature of the radiation mechanism as well as the fundamental features were not understood yet. Moreover, to develop the STAR emitter with greater power for useful applications, it is first important to understand the physical nature of the emission mechanism experimentally.

Theoretically, a unique radio engineering approach to the mechanism was made by Pedersen and Madsen, whereas models based on numerical simulations of the coupled sine-Gordon equations for stacked Josephson junctions were widely used by others. Among them some models assumed a magnetic current radiation source (or cavity mode) only without taking into account the electromagnetic fields at near-field, and also neglecting the substrate effect. Here we provide clear experimental evidence contradicting those model calculations.

We measured the angular dependence of the far-field radiation from various rectangular mesas. Our experimental results are inconsistent with the predictions of either an electric or a magnetic surface current sources acting alone. Instead, they strongly suggest a dual-source THz radiation mechanism: the uniform part of the ac-Josephson current acts as an electric surface current source, and the inhomogeneous part of the ac-Josephson current sets up a displacement current that excites a cavity resonance mode in the mesa, which locks the radiation frequency and acts as a magnetic surface current source. By adjusting the relative amplitude and phase of the two source currents and accounting for the substrate, excellent agreement with experiment is obtained. Furthermore, from FTIR analysis of the dc current-voltage ($I$-$V$) characteristic, the locking onto the resonant cavity mode appears to occur by the sequential synchronization of each of the IJJs.

High-quality single crystals of BSCCO were grown by the conventional traveling solvent floating zone method using a modified infrared image furnace. A piece of crystal was annealed overnight to an appropriate doping level at 600°C in Ar+0.1% O$_2$ atmosphere. The resulting crystals are underdoped with $T_c \sim 75 - 86$ K. Two types of rectangular mesas were prepared using either conventional photolithography or focused ion beam milling technique. Photographs of a cleaved piece of a pristine single crystal, an atomic force microscope (AFM) image of a rectangular mesa after processing and the final form of the mesa are shown in Figs. 1(a), 1(b) and 1(c), respectively. From the AFM measurements shown in Figs. 1(b) and 1(d), its length was $L \sim 400$ mm, width $w = 77.4 \pm 4.5$ mm and thickness $d = 1.2$ mm.

The temperature dependence of the normal state e-
I mesas with Lxz are shown in Fig. 3(a), where the I from several mesas. Similar results in the xy plane, or I from the same mesa, would vanish at \( \theta = 90^\circ \), as observed. However, for the appropriate value \( n = 4.2 \), the predicted \( I(90^\circ, 90^\circ) \) from the simple cavity resonance model is \( \sim 87\% \) of its maximum value,\(^{15}\) as shown in Fig. 2(a), unlike the experiments.

Third, radiation, with an order of magnitude weaker than the fundamental, at several integral higher harmonics of the fundamental frequency is generally observed, as the previous report,\(^{3}\) but subharmonics were never observed. Fourth, small but clear minor lobes are also observed at \( \theta \sim \pm 75^\circ \) as seen in Figs. 2(a) and 3(a), although their integrated intensity appears to be only a few \% or less of the total. These lobes may arise from higher harmonics, but the spectrum of the entire \( I(\theta, \phi) \) has not yet been measured.
These results can naturally be understood by combining both radiation sources, since there actually exist both the displacement current and the real $\nu$-Josephson relation, since there exist both $\nu$-Josephson and the real $ac$-Josephson current flowing across the mesa, which have both spatially uniform and non-uniform parts. Problems in fitting the data near to $\theta = \pm 90^\circ$ can be overcome by an appropriate model of the superconducting substrate.\textsuperscript{15)  

It was shown previously that the fundamental frequency $\nu$ satisfies the $ac$-Josephson relation,\textsuperscript{2, 3)  

$$\nu = \frac{2\pi \hbar}{c} V / N,$$  

where $\hbar$ is the Planck’s constant. Since $w$ is comparable to the wavelength of the plasma waves in the mesa, cavity resonances may occur as standing waves. Empirically, the wavelength $\lambda = 2w$ of the lowest energy state in the mesa, leading to the condition $\nu = c_0 / \alpha \lambda = c_0 / 2nw$, where $c_0$ is the speed of light in vacuum, $n = \sqrt{\epsilon}$, and $\epsilon$ is the relative permittivity of the junctions. This empirical relation appears to work very well in many samples with different widths and $L / w$ ratios, as shown in Fig. 4. From the slope of the line in Fig. 4, $n = 4.2$ is obtained fairly accurately, corresponding to the THz-frequency dielectric constant $\epsilon = 17.6$, which is about 50% larger than the value ($\epsilon = 12$) obtained by infrared spectroscopy.\textsuperscript{20)  

No anomaly in $\epsilon$ for frequencies up to 0.9 THz has been observed.

For $d < L, w$, the lowest mode frequencies satisfy $\nu_{0M} = \frac{c_0}{\alpha \lambda} \sqrt{(m/L)^2 + (p/w)^2}$, where $m$ and $p$ are integers.\textsuperscript{18)  

Hence, the lowest frequency mode for $L > w$ is expected to be the $(010)$ mode with $\nu_{010} = \frac{c_0}{\alpha \lambda \sqrt{100}}$. As seen in Fig. 4, the experimental results are definitely consistent with the $(001)$ mode, not the $(010)$ mode, contradicting the simple cavity resonance model. This may be related to an instability that could arise from the energy loss by the penetration of the magnetic field in the $z$-direction into the mesa, since $\lambda \sim \lambda_c$, where $\lambda_c$ is the superconducting penetration depth in the $c$-axis direction, resulting in higher inductive energy states for the $(0m0)$ modes. But whatever the actual reasons for the apparent non-resonant $(0m0)$ modes are, the angular dependence of the far-field radiation is calculated by combining the output from the two sources, one of which excites the $(001)$ mode taking into account the superconducting substrate.\textsuperscript{15)  

Least-squares fits to the data shown in Figs. 2(a) and 3(a) were performed using Model I, which averages the two source contributions symmetrically about $\theta = 0^\circ$, while preserving the Love magnetic equivalence principle boundary condition $\hat{n} \times \mathbf{H} = 0$ on the mesa edge,\textsuperscript{15, 18)  

and using Model II, which relaxes the boundary condition while preserving $\lambda$.\textsuperscript{15)  

The best fit with standard deviation $\sigma = 0.122$ was found for Model I with mixing parameter $\alpha_0(0) = 0.310$, corresponding to 24% of the overall intensity arising from the magnetic (cavity) source. The cross-sections of the best dual- and single-source fits are shown by the solid- and dashed-curves in Figs. 2(a) and 3(a), and the full radiation pattern resulting from the dual-source best fit is shown in Fig. 3(b). Preliminary but similar results were obtained from numerical simulations.\textsuperscript{11, 12, 14)  

Figure 5 presents the detailed $dc$ $I$-$V$ characteristics and the corresponding output of the $\mathrm{Si}$-bolometer detector as functions of $V$, when the scan is made very slowly (2.8 hours for a one-cycle measurement). In this case many radiation peaks are observed. Examining them in detail, one finds an interesting feature. When $I$ is reduced slowly on the return $I$-$V$ branch, the radiation energy starts to build up gradually until the growth is interrupted by a jump to a different $I$-$V$ branch. This is more clearly seen in the inset of Fig. 5, in which the sawtooth-like radiation peaks are seen over the range of $V$ from -0.67 to -0.57 V. From the detailed FTIR measurements performed at various points in the sawtooth region, it turns out that the observed frequency shifts to lower frequency as the power increases (i.e., $I$ decreases.

This can be interpreted as the gradual decrease of the $ac$-Josephson frequency as $I$ decreases, until the mesa eventually enters a cavity resonance state. Meanwhile,
the radiation energy increases in the mesa, and when the power reaches a threshold, $I$ jumps to a different $I$-$V$ branch. Therefore, the mechanism of the strong coherent continuous radiation appears to be due to gradual development of the synchronization in the individual or groups of Josephson junctions onto the cavity resonance mode, rather than a sudden catastrophic transition. This fact strongly suggests that the stronger output power may be achieved by preventing the $I$-$V$ curve from jumping.

The combination of two conditions necessary for the resonant emission may be written as $V_{dc}^{tot} = (c_0/2nu)(1/K_J)(2d/c)$, where $V_{dc}^{tot}$ is the required $dc$ voltage to meet the cavity resonance oscillation, the Josephson constant $K_J = 438.5979$ (GHz/mV), and $c \approx 30.65$ Å is the $c$-axis lattice constant of BSCCO. This reduces to $V_{dc}^{tot} = 48.2(d/w)$, which surprisingly depends only upon $d/w$.21) Since an emitting junction must be in its resistive state, its resistance $R_J$ and $dc$ current $I$ must satisfy $R_J I \geq V_{dc}^{tot}$. Because $R_J(T)$ has a steep negative temperature coefficient, and BSCCO each junction is subject to severe Joule heating, the whole mesa may thermally be stabilized. On the other hand, because of large reduction of $R_J$ the above condition for synchronized emission often may not be satisfied, especially for overdoped samples, due to their considerably smaller $R_J$ values. This may explain partially why we initially had many unsuccessful mesas.

In conclusion, we measured the angular dependence of the far-field THz emission from mesas of single-crystal Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. Our results provide strong evidence for a dual-source mechanism in which the uniform and inhomogeneous parts of the ac-Josephson current respectively act as an electric surface current source and set up a displacement current that excites a mesa cavity resonance mode which locks the radiation frequency, and acts as a magnetic surface current source. By adjusting the relative amplitude and phase of the two source currents and accounting for the substrate, excellent agreement with experimental results is obtained. From FTIR analysis of the $I$-$V$ characteristic, the radiation appears to build-up from gradual synchronization of the individual or groups of Josephson junctions to the cavity resonance mode.

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