We measure the local harmonic generation from an YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) bicrystal grain boundary to examine the local Josephson nonlinearities without the influence of edges. Spatially resolved images of second and third harmonic signals generated by the grain boundary are shown. The harmonic generation and the vortex dynamics along the grain boundary are modeled with the extended resistively shunted Josephson array model, which shows reasonable quantitative agreement with the experimental data. The model also gives qualitative insight into the vortex dynamics induced in the junction by the probing current distribution. A characteristic nonlinearity scaling current density $J_{NL} \sim 1.5 \times 10^5$ A/cm$^2$ for the Josephson nonlinearity is also extracted.

DOI: 10.1103/PhysRevB.72.024527  PACS number(s): 74.50.+r, 74.25.Nf, 74.81.Fa, 74.78.Bz

INTRODUCTION

The nonlinear behavior of high-$T_c$ superconductors has been of great concern because it can potentially reveal the underlying physics of these materials. While the microscopic origins of nonlinear response still remain uncertain, all superconductors have an intrinsic time-reversal symmetric nonlinearity associated with the nonlinear Meissner effect (NLME). Calculations based on BCS theory and Ginzburg-Landau theory have been proposed to describe the harmonic generation (or intermodulation) response of the NLME. Many experiments have been conducted to study third order harmonic generation or intermodulation signals, which may arise from this intrinsic nonlinearity.

On the other hand, extrinsic sources of nonlinearities dominate the nonlinear response of superconductors in most cases. They include grain boundaries, enhanced edge currents, and weakly coupled grains. A number of experiments have been conducted to understand and characterize the nonlinear properties of one of these sources by measuring artificially-prepared features, e.g., bicrystal grain boundaries (GBs). Most of these experiments are done with resonant techniques, which by their nature study the averaged nonlinear response from the whole sample rather than locally. Such techniques usually have difficulty in avoiding edge effects, which give undesired vortex entry due to the enhanced currents and defects along the etched edges, and do not reveal the local intrinsic nonlinear properties of superconductors.

In our previous work, we have shown that by measuring the local nonlinear response in second and third harmonic generation, we can locally identify the superconducting bicrystal GB without the edge currents involved. However, the sample in Ref. 27 was not protected from vortex entry by external magnetic fields. In this work, we present additional data on the same sample with very effective magnetic shielding and with much more comprehensive analysis and simulations. Although there is no significant difference between data taken with and without magnetic shielding, employing the shielding assembly helps to eliminate potential ambiguity in identifying the origin of the observed nonlinear signals. This permits the study of the intrinsic Josephson vortex dynamics in a long Josephson high-$T_c$ bicrystal grain boundary.
microwave signal at frequency $f=6.5$ GHz, which is low-pass filtered to guarantee the purity of the spectrum, to the sample via the coupling between a loop probe and the sample. The power of the microwave signal output from the generator is $+12$ dBm. After calibrating the system to take the attenuation along the transmission line into account, we find that the microwave signals suffer an attenuation of 2 dB before reaching the probe. The microwave power mentioned later in the text will be the actual power reaching the probe, which is $+10$ dBm.

The loop probe is made of a nonmagnetic coaxial cable with its inner conductor (200 $\mu$m diameter) forming a $\sim500$ $\mu$m outer-diameter semicircular loop shorted with the outer conductor. When this loop probe is close to a conducting surface, it couples to the surface magnetically, and induces microwave currents flowing on the surface with a geometry defined by the loop size, shape, and orientation. If there are any local nonlinear mechanisms present in the range of the induced microwave currents, additional microwave currents at multiples of $f$ will be generated.

We detect the lowest-order harmonic signals at $2f$ and $3f$ by measuring the microwave signal coupled back from the sample surface through the probe with the spectrum analyzer. Since the harmonic signals are extremely weak, they are amplified by $\sim60$ dB before entering the spectrum analyzer. High-pass filters are used before amplification to prevent amplifying the fundamental signal and getting a strong signal at frequency $f$ into the spectrum analyzer. We measure the amplified second and third harmonic signals simultaneously with the fundamental signal. Since the harmonic signals are extremely weak, they are amplified by $\sim60$ dB before entering the spectrum analyzer. High-pass filters are used before amplification to prevent amplifying the fundamental signal and getting a strong signal at frequency $f$ into the spectrum analyzer. We measure the amplified second and third harmonic signals simultaneously with the spectrum analyzer as a function of temperature and position over the sample.

The sample is a 500-Å-thick YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) thin film deposited by pulsed laser deposition on a 30° misoriented bicrystal SrTiO$_3$ substrate at nearly optimal doping level. The spatially averaged $T_c$ is 88.9 K, as measured by macroscopic ac susceptibility. The loop probe is placed 12.5 $\mu$m above the sample, fixed by a Teflon™ sheet. The sample is kept in a high vacuum cryostat cooled with continuously flowing liquid helium, and its temperature can be controlled between $\sim3.5$ K and room temperature to within $\pm10$ mK. The probe can move in the $x$, $y$, and $z$ directions inside the cryostat to perform spatially resolved measurements. The sample is surrounded by two layers of mumetal shielding, and two layers of cryoperm shielding. They are supported by an ultralow-carbon steel base to provide a low background magnetic field environment ($<1$ $\mu$G) for the measurements. Thus all the measurements presented here are carried out in nominally zero dc magnetic field.

Measurements of temperature-dependent third-order harmonic power ($P_{3f}$) are first performed at locations above the grain boundary (GB) and away from the boundary (non-GB) as shown in the inset of Fig. 2. A strong peak in $P_{3f}(T)$ is observed around $T_c$ at all locations on the sample, and these peaks show similar magnitudes as shown in Figs. 2 and 3. In addition, these peaks are identical to those taken without magnetic shielding. This $P_{3f}$ peak near $T_c$ is predicted by all models of intrinsic nonlinearities of superconductors. For example, the Ginzburg-Landau (GL) theory gives a reasonable fit to this peak, which is also shown in Fig. 2 as a solid line. This fit employs Eq. (4) below with a GL expression for $J_{NL}(T)$ (introduced below), and known sample properties such as the thickness and the magnetic penetration depth. The most important fitting parameters used are the zero-temperature depairing critical current density $J_c=10^{12}$ A/m$^2$, $T_c=88.5$ K for non-GB and $T_c=88.0$ K for GB, and finite transition width $\Delta T$ (0.2 K for non-GB and 0.3 K for GB). Other parameters are the screening length cutoff $(1.1$ $\mu$m) and a cutoff current density scale $(3 \times 10^{10}$ A/m$^2)$.
perhaps associated with fluctuation-induced nonlinearity above $T_c$. A small shift in temperature $\sim 0.5$ K and broadening $\sim 0.1$ K of this peak with location is noted, which is most likely due to inhomogeneity at the grain boundary.

A significant difference in both the second and third harmonic response with position is observed for temperatures $T > 0.9T_c$, as shown in Fig. 3, where the GB region shows significant enhancement ($P_{2f} \sim 70$ dBm and $P_{3f} \sim 55$ dBm) in both $P_{2f}$ and $P_{3f}$, but non-GB regions show essentially only noise-level signals. The power dependence of both $P_{2f}$ and $P_{3f}$ signals were shown in Ref. 27 without magnetic shielding. They show noise-level nonlinear response in the non-GB region, and strong and saturating nonlinear response over the GB. Results with magnetic shielding are similar, and show the same saturation of harmonic response at moderate input power, suggesting that the enhanced $P_{2f}$ and $P_{3f}$ at the GB are due to the nonlinearity of the Josephson tunneling effect across the boundary. The microscope is capable of detecting and quantifying this local nonlinear mechanism, as discussed below.

**DATA**

**Spatially resolved 1D and 2D images**

To demonstrate that our microwave microscope is also able to spatially resolve the nonlinearity of the grain boundary, a measurement of $P_{2f}$ and $P_{3f}$ along a line crossing the grain boundary is performed at an intermediate temperature $T = 60$ K as shown in Fig. 4. A clear peak in both $P_{2f}$ and $P_{3f}$ is observed above the GB, with a full-width half maximum of about 700 $\mu$m for $P_{2f}$ and 500 $\mu$m for $P_{3f}$. The spatial widths of the $P_{2f}$ and $P_{3f}$ signals are determined by two factors. One is the local current distribution produced by the loop probe, which is on the order of the size of the loop; the other is the local quality of the film. If the local quality of the sample is degraded and more nonlinear, even a very small current will induce nonlinear signals. By comparing the one-dimensional (1D) plots presented in this work and those in Ref. 27, slightly broader $P_{2f}$ and $P_{3f}$ patterns are seen here, indicating the minor degradation of the local film quality near the GB, which makes the Josephson junction more non-

![FIG. 4. (Color online) $P_{2f}$ and $P_{3f}$ as a function of position across the bicrystal grain boundary, taken at $T = 60$ K, $f = 6.5$ GHz, and input power $= +10$ dBm. Enhancement of both $P_{2f}$ and $P_{3f}$ signals near the boundary is observed, with a width $\sim 700$ $\mu$m for $P_{2f}$ and $\sim 500$ $\mu$m for $P_{3f}$. These widths are on the order of the spatial distribution of the current density, determined by the probe geometry. The inset is the plot of $P_{2f}(x)$ and $P_{3f}(x)$ in linear scale, which more clearly shows the width of the $P_{2f}$ and $P_{3f}$ signals.](image1)

![FIG. 5. (Color online) Two-dimensional images of $P_{2f}$ [(a) in log scale and (c) in linear scale] and $P_{3f}$ [(b) in log scale and (d) in linear scale] response on a $3 \times 4$ mm$^2$ area containing the YBCO bicrystal grain boundary taken at $T = 60$ K, $f = 6.5$ GHz, and input power $+10$ dBm. The rf currents are primarily sent in a direction perpendicular to the grain boundary. The GB is noted by the vertical black line, and is clearly identified by the enhancement of $P_{2f}$ and $P_{3f}$ signals above the background level.](image2)
linear. This measurement is well interpreted and reproduced by the extended resistively shunted Josephson (ERSJ) junction model discussed below.

To show that our microwave microscope is also capable of imaging nonuniformity of the bicrystal grain boundary, we made a two-dimensional scanned image across the grain boundary with a loop probe at an input microwave power of +10 dBm. The loop probe is oriented so that the currents are flowing across the grain boundary with a maximum current density \( \sim 5 \times 10^5 \) A/cm\(^2\). The scanning steps are 50 \( \mu \)m across the boundary and 500 \( \mu \)m along the boundary. As shown in Fig. 5, the bicrystal grain boundary is identified in both the \( P_{27} \) and \( P_{3f} \) images. Panels (a) and (b) show the second and third harmonic images on a log scale, while (c) and (d) show them on a linear scale for comparison. It is clear that the harmonic response due to the nonlinearities of the GB varies along the length of the grain boundary, and that the \( P_{3f} \) image is more uniform than the \( P_{27} \) image. Also note that the probe was at least 3 mm away from all edges while taking this image, hence the contribution from the edge effect is avoided.

**Extraction of the characteristic nonlinearity current scales \( J_{NL} \) and \( J_{NL'} \)**

There are many different microscopic models predicting various nonlinearities in superconductors. Most known models of nonlinear response can be expressed in a general form for the penetration depth \( \lambda \) (or superfluid density) assuming that inductive nonlinearities dominate and time-reversal symmetry is preserved

\[
\frac{\lambda(T,f)^2}{\lambda(T,0)^2} = 1 + \left( \frac{J}{J_{NL}(T)} \right)^2 + \cdots , \tag{1}
\]

where \( J \) is the applied current density and \( J_{NL}(T) \) is a temperature-dependent, geometry-independent scaling current density, which can vary by orders of magnitude depending on the precise nonlinear mechanism and temperature [here we assume \( J \approx J_{NL}(T) \)]. For example, the Ginzburg-Landau theory of the nonlinear Meissner effect yields \( J_{NL} \approx 10^8-10^9 \) A/cm\(^2\) for cuprates, and a long ideal 1D Josephson junction array expects an upper limit for \( J_{NL} \) to be on the order of \( 10^7 \) A/cm\(^2\) in the intermediate temperature region \( T/T_c \sim 0.5 \).\(^{33} \) To evaluate the ability of our microwave microscope to detect intrinsic superconducting nonlinearities due to different mechanisms, we extract the \( J_{NL} \) from our data with the following algorithm.\(^{29,34} \)

When a single-tone microwave signal at frequency \( f \) is sent to a superconducting sample via the probe, microwave currents are induced in the sample due to the Meissner screening. The sample thickness is less than the magnetic penetration depth, and we assume that the supercurrents flow uniformly over the thickness to screen out the magnetic field from penetrating through the sample. Since in this case most of the energy is carried by the supercurrents, it is assumed that the kinetic inductance \( L_{KL} \) of the superconductor dominates its nonlinear ac response. The kinetic inductance can be derived as\(^{29,35} \)

\[
L_{KL} \equiv \mu_0 \int \int_{\text{cross section}} \lambda^2(T,f) J^2 ds = \mu_0 \int \int_{\text{cross section}} \lambda^2(T,f) \left( \frac{\bar{J}}{J_{NL}} \right)^2 \bar{J} \cdot d\bar{s}, \tag{2}
\]

where \( \lambda \) is the magnetic penetration depth, \( \bar{J} = \bar{J}(x,y) \) is the spatial distribution of the current density on the surface, and the numerator represents the energy stored in the supercurrents. The denominator involves the total current flowing through a cross section perpendicular to the \( y \) axis in the integral, as illustrated in Fig. 6. Using Eq. (1), Eq. (2) can be rewritten as

\[
L_{KL} \equiv \mu_0 \lambda(T,0)^2 \left( \frac{\int \int_{\text{cross section}} J^2 ds + \int \int_{\text{cross section}} J_{NL}^2(T) \bar{J}^2 ds}{\int \int_{\text{cross section}} \bar{J} \cdot d\bar{s}} \right) dy = L_0 + \Delta \ell J^4, \tag{3}
\]

where \( L_0 \) is the linear kinetic inductance resulting from integrating the \( \bar{J}^2 \) term, \( I \) is the total current in the film \( I = \max(\int \int_{\text{cross section}} \bar{J} \cdot d\bar{s}) \) and \( \Delta \ell \) is the coefficient of \( I^2 \) in the nonlinear kinetic inductance, associated with integrating the \( J^4 \) term. The coefficient \( \Delta \ell \) has units of \( H/A^2 \).

Since the superconducting sample is driven by a microwave current, it is equivalent to an ac circuit with a driving ac current source \( I_0 \sin(\omega t) \) and a lumped nonlinear inductor,
and develops a voltage drop across the inductor, given by
\[ V(t) \equiv L_0 \frac{dI(t)}{dt} + \Delta \ell \left( I(t)^2 \right) \frac{dI(t)}{dt}, \]
where \( \omega \) is the driving frequency. By solving these equations, we can extract the harmonic content from the voltage solution, and the expected third harmonic power becomes\(^3\)
\[ P_{3f} = \frac{|V_{3f}|^2}{2Z_0} = \left( \frac{\omega \Delta \ell I_0^3}{4Z_0} \right)^2, \]
where \( Z_0 \) is the characteristic impedance of the coaxial transmission line and
\[ \Delta \ell = \frac{\mu_0 \lambda^2(T,0)}{I_0^2 J_{NL}(T)^2} \int \frac{\int \text{cross section} J^4(x,y) ds}{\left( \int \text{cross section} J^2 ds \right)^2} dy, \]
from Eq. (3).
Since our sample is in the limit that its thickness \( t \) is much less than the magnetic penetration depth, \( t < \lambda \), the current density can be replaced by the surface current density, \( \vec{J} = \vec{K}/t \), and the volume integral can be reduced to surface integral of \( \vec{K} = \vec{K}(x,y) \),
\[ \Delta \ell = \frac{\mu_0 \lambda^2(T,0)^2}{I_0^2 J_{NL}(T)^2} \int \frac{\int K^4 dx}{K_x} \frac{dy}{\text{cross section}} \]
so that
\[ P_{3f} = \frac{1}{2Z_0} \left( \frac{\omega \mu_0 \lambda^2(T,0)^2}{4I_0^2 J_{NL}(T)^2} \right)^2 \left( I_0 \int \frac{\int K^4 dx}{K_x} \frac{dy}{\text{cross section}} \right)^2. \]

We use numerical solutions of Maxwell’s equations [obtained using the high-frequency structure simulator (HFSS) software] to simulate the microwave currents induced on the sample surface by a loop probe, and perform a surface integral of \( K^4 \) and a line integral in the \( x \) direction in Fig. 6 to obtain the (power-dependent) figure of merit of the probe:
\[ \Gamma \equiv I_0 \int \frac{\int K^4 dx}{\int K_x} \frac{dy}{\text{cross section}}. \]
With an estimation of \( \lambda(60 \text{K},0) = 2440 \text{ Å} \) and \( \Gamma = 31.2 \text{ A}^2 \text{m}^2/\text{cm}^2 + 10 \text{ dBm} \) input power for our probe geometry, we can convert the measured \( P_{3f} \) to \( J_{NL} \) using Eq. (4). Recall that \( J_{NL} \) is a geometry-independent quantity which can reveal the mechanism responsible for the observed nonlinear responses.

To interpret the second harmonic data, which implies the presence of a time-reversal symmetry-breaking (TRSB) nonlinearity, we empirically propose a linear term in Eq. (1). This linear term is characterized by an empirical TRSB characteristic nonlinear scaling current density \( J_{NL} \) (Refs. 29 and 31)
\[ \frac{\lambda(T,J)^2}{\lambda(0,T)^2} = 1 + \left( \frac{J}{J_{NL}(T)} \right)^2 \left( \frac{J}{J_{NL}(T)} \right)^2 + \cdots. \]
In this case, we do not propose that the intrinsic superfluid response of the superconductor breaks time-reversal symmetry. Rather, we expect that the total inductive response does break time-reversal symmetry, and simply propose Eq. (5) as a naive parametrization that captures the basic phenomenon.

Following the same algorithm, but now using the \( P_{3f} \) data, we can develop a similar formula for extracting \( J_{NL} \) from \( P_{3f} \) data. Starting with Eq. (2) and the current dependence in Eq. (5), a linear current dependence is introduced to the nonlinear kinetic inductance \( L_{Kl} \equiv L_0 + \Delta \ell + \Delta \ell \ell^2 \), where \( \Delta \ell \) comes from the integral containing the second term on the right-hand side of Eq. (5). Fourier analysis of an ac circuit with this nonlinear inductance shows that the \( V_{3f} \) term is due solely to the \( \Delta \ell \) term, and \( P_{3f} \) can be written as
\[ P_{3f} = \frac{|V_{3f}|^2}{2Z_0} = \left( \frac{\omega \Delta \ell I_0^2}{2Z_0} \right)^2 \int \frac{\text{cross section} K^4 dx}{\left( \int \text{cross section} K_x dx \right)^2} dy. \]
Once again a figure of merit (for the second harmonic response) can be defined and calculated, and Eq. (6) can be used to find \( J_{NL}(T) \) given the measured \( P_{3f} \) and a few sample and measurement parameters \([t, \lambda(T), \omega]\). Further details of this algorithm can be found elsewhere.\(^{29,31}\)
Figures 7 and 8 show the spatially resolved \( J_{NL} \) and \( J_{NL} \) across the bicrystal grain boundary, obtained from the \( P_{3f} \) and \( P_{2f} \) data in Fig. 4, and Eqs. (4) and (6). The \( J_{NL} \) near the
grain boundary is about $1.5 \times 10^5$ A/cm$^2$, whereas the measured $J_c$ on a similar bicrystal junction is $\sim 10^4$ A/cm$^2$ at 60 K. We expect these two current densities to be similar, but not necessarily identical. The onset of third-harmonic nonlinearity from an extended Josephson junction is a complicated matter (as discussed below) and is probably not simply characterized by the Josephson critical current $J_c$ in place of $J_{NL}$ in Eq. (1). The assumption of a purely inductive origin of the nonlinearity may also partly influence the value of $J_{NL}$.

The $J_{NL}$ value near the grain boundary (Fig. 8) is on the order of $10^7$ A/cm$^2$, which is significantly larger than $J_{NL}$. The physical interpretation of $J_{NL}$ is not clear for the case of a bicrystal grain boundary, as discussed above. The microscopic origins of $P_{3f}$ are discussed in detail below. The noise level in the measurements limits the sensitivity of this setup to $J_{NL} < 2.1 \times 10^8$ A/cm$^2$ and $J_{NL} < 4.7 \times 10^8$ A/cm$^2$.

To enhance the ability to study weaker nonlinearities due to smaller structural defects (rather than an artificially made GB), better spatial resolution and sensitivity to larger $J_{NL}$ and $J_{NL}'$ are desired. According to the algorithm sketched above, better sensitivity can be achieved by measuring thinner films (reducing $t$) and/or increasing the probe figure of merit $\Gamma$, while the spatial resolution can be improved by reducing the size of the loop probe and bringing it closer to the sample. We use both HFSS and an analytical model (described below and elsewhere$^{29}$) to calculate the probe figure of merit $\Gamma$ for different sizes of probes, and find that the smaller the loop probe and the closer it gets to the sample, the larger the figure of merit $\Gamma$ (see Fig. 9).

The analytical model consists of a filamentary circular loop with its identical image loop. The loop is located approximately at the center of the physical wire that forms the loop probe. Taking the geometry of the loop probe into account, the diameter of this ideal loop is on the order of 500 $\mu$m and can be varied for fitting purposes. The distance of two loops are restricted by the Teflon sheet and finite wire diameter, hence the closest distance between the loop edges is 225 $\mu$m, and is not a variable in the model. We use this model to calculate the current distribution in the plane midway between the loops. The loop diameter is varied to find the current distribution that has the same width as that from the HFSS calculation, and we find this diameter to be 540 $\mu$m.

Using the current distribution calculated by the analytical model with these parameters, we can calculate the figure of merit $\Gamma$ for different sizes of probes, which appears to be consistent with the HFSS calculation, as shown in Fig. 9. This figure shows that better spatial resolution and higher sensitivity to $J_{NL}$ and $J_{NL}'$ can be achieved simultaneously by using smaller loop probes.

**MODEL: EXTENDED RESISTIVELY SHUNTED JOSEPHSON JUNCTION (ERSJ)**

The purpose of this section is to develop a quantitative and microscopic understanding of the data presented in Figs. 4 and 5. It is well known that a purely singletone ac current-biased single Josephson junction generates harmonics at all odd integer multiples of the driving frequency,$^{11}$ which is measured in our data by $P_{3f}$. However, to obtain a more realistic and comprehensive understanding of a long weak-link junction, such as the YBCO bicrystal grain boundary, the extended resistively shunted Josephson (ERSJ) junction array model is introduced.

**Spatial distribution of harmonic data**

The ERSJ model we adopt was first used by Oates et al.$^{18,20}$ to describe the nonlinear behavior of a YBCO bicrystal GB in their edge-dominated stripline superconducting microwave resonator.

In the ERSJ model, the long weak-link Josephson junction in the sample is modeled as a 1D array of 2001 single
resistively shunted Josephson junctions combined in parallel, coupling with each other through lateral inductors (see Fig. 10). These inductances are assumed to be identical and estimated to be \( l = 3 \times 10^{-11} H \) using the formula \( l = \mu_0 d_m / 2 t \), where \( d_m \) is the magnetic thickness of the junction defined as

\[
d_m = 2 \lambda \coth(\lambda / \lambda_c),
\]

\( \lambda \) is the penetration depth, and \( t \) the thickness of the film. We assume \( \lambda(0) = 180 \) nm, and use mean-field BCS theory to estimate \( \lambda \) at other temperatures.

Since the measurement takes place with a less than 1 mm wide current distribution around the center of a 10 mm \( \times \) 10 mm film, we have an essentially infinite 2D superconducting plane with no edge effect involved. To simulate this, the array is terminated by much larger lateral coupling inductors with \( l = 10^{-8} H \) before the last junctions, and the simulation is not sensitive to the precise value of the termination inductance.

The spacing between junctions is determined by the Josephson penetration depth \( \lambda_j = \sqrt{\Phi_0 / [2 \pi \mu_0 J_c(T) d_m(T) \lambda_c(T)]} \), where \( \Phi_0 \) is the flux quantum. From measurements on the junctions deposited with the same procedure by the same group, we find the critical current density \( J_c \sim 10^8 A/m^2 \) and specific normal resistance \( R_c \sim 0.02 \mu \Omega \) cm\(^2\) at 60 K. Therefore, we estimate each junction in the ERSJ model to have a Josephson penetration depth \( \lambda_j \sim 1 \) \( \mu \)m, a critical current of \( I_c = 6 \) \( \mu \)A, and a shunt resistance \( R \) of 50\( \Omega \). We assume that all the junctions, resistors, and inductors are identical, and the junctions are equally spaced. We use a program called WRSPICE\textsuperscript{TM}, obtained from Whiteley Research, Inc., to carry out the simulations of this model. The physical quantities discussed above are parameters that can be varied in the WRSPICE\textsuperscript{TM} simulation.

It is assumed that the biasing microwave current with a frequency of 6.5 GHz is applied to each junction \( (I_n) \) in the model and varies according to the surface current distribution on the film induced by the loop probe. The current distribution is estimated from two calculations. First is a simplified analytical model of an ideal circular loop in a vertical plane, with an effective radius 270 \( \mu \)m, coupling to a perfectly conducting horizontal plane 382.5 \( \mu \)m away from the center of the loop.\textsuperscript{20} The magnitude of the current density is determined by a much more sophisticated microwave simulation software HFFS by Ansoft\textsuperscript{TM}. The full geometry of the loop probe is used in this calculation, and it also produces a similar current distribution. The maximum current density on the sample is \( \sim 5 \times 10^4 A/cm^2 \) for +10 dBm input power. A cross section through the peak current distribution is shown in Fig. 10 on a linear scale.

The calculated two-dimensional current distribution is applied to each junction in the WRSPICE\textsuperscript{TM} program. To reproduce the spatial distribution of the measured \( P_{2f} \) and \( P_{3f} \) in Fig. 11 from the model, we take a one-dimensional slice from the two-dimensional current distribution at 101 locations along the scanning direction and apply it to the junctions in the ERSJ model. We sum up the nonlinear potential differences across all junctions, calculated by WRSPICE\textsuperscript{TM}. The simulations are done in transient analysis, which simulates the evolution of the system in time. The potential differences are found to be periodic a few periods after starting the numerical analysis. We average the potential differences between the fifth period and 65th period to reduce the numerical error from the calculation, and extract the higher harmonics from this collective nonlinear potential difference via Fourier transformation. The calculated \( P_{2f} \) and \( P_{3f} \) show good quantitative agreement with experimental results in both magnitude and spatial resolution as shown in Fig. 11.

Since a single pure-tone ac current-biased Josephson junction only generates \( P_{3f} \) signal (as well as other odd harmonics), the \( P_{2f} \) signals are attributed to the presence and motion of Josephson vortices generated along the long grain boundary. The voltages across the junctions become time-irreversible because of the motion of the vortices, and therefore contain second harmonic content. This interpretation is confirmed by simulating the ERSJ model with no lateral coupling inductors (uncoupled ERSJ), with each junction responding to its biasing current independently. The calculated \( P_{3f} \) of the uncoupled ERSJ model has larger magnitude and narrower spatial distribution due to the lack of lateral coupling, and no \( P_{2f} \) is generated in the uncoupled case.\textsuperscript{27} The strong presence of \( P_{2f} \) in the experimental data is proof that the collective behavior of the Josephson system is crucial to our understanding.
The simulated $P_{2f}$ and $P_{3f}$ by the ERSJ model are shown in Fig. 11 with the experimental results. It appears that the model describes well the magnitudes and spatial distribution of the $P_{2f}$ and $P_{3f}$ data. However, the details of the data are not well described by the model because the nonuniformity in the sample (as seen in Fig. 5) is not present in the model. The broader distribution of $P_{2f}$ and $P_{3f}$ data in Fig. 11 may be due to inhomogeneity and pinning sites in the grain boundary, which are not included in the model. The wiggles observed in the calculated solid and dashed lines in Fig. 11 are due to the numerical nature of the calculation. They are reduced tremendously by averaging over more periods, at the expense of a significant increase of computation time and required computation resources. Because of these constraints, the results presented in this paper are averaged over only 60 periods.

It is also noted that by applying strong enough microwave power, the Josephson array system becomes chaotic. This phenomenon is also seen in our simulations at much higher input powers (>30 dBm), but does not play a role at the powers used in the experiments (~10 dBm).

**Vortex dynamics**

To further our fundamental understanding of the physics governing the local nonlinearities, especially the $P_{2f}$ responses, we use the ERSJ model to evaluate the nucleation and motion of Josephson vortices in the middle of a driven infinite superconducting GB.

A long Josephson junction can be described by the sine-Gordon equation \[ \lambda_j \frac{\partial^2 \Delta \gamma(x,t)}{\partial x^2} = \sin \Delta \gamma(x,t) + \frac{L_j}{R_j} \frac{\partial \Delta \gamma(x,t)}{\partial t} + C_j \frac{\partial^2 \Delta \gamma(x,t)}{\partial t^2}, \]
where $\Delta \gamma(x,t)$ is the gauge-invariant phase difference across the junction at position $x$ and time $t$, $\lambda_j$ is the Josephson penetration depth, $L_j = \Phi_0 / 2 \pi J_c$, $R_j = \rho d \rho$, and $d$ are the junction resistivity and thickness, and $C_j = e / d$. The parameters $L_j$ and $R_j$ are inductance and resistance times area and $C_j$ is the capacitance per area of the junction. Since our ERSJ model is equivalent to solving this equation on a discrete one-dimensional lattice, we calculate the key quantity $\Delta \gamma(n,t)$, where $n$ indicates the $n$th junction, to extract other physical quantities, such as the current, magnetic field, and flux at each junction.

The magnetic field along the grain boundary is given by $B(x) = (\Phi_0 / 2 \pi d_n) \left( \frac{\partial \Delta \gamma(x,t)}{\partial x} \right)$. Since the distance between the junctions is $\lambda_j$ in the model, the flux between adjacent junctions is determined by

\[ \Phi(n) = B(n) \times (d_m \cdot \lambda_j) = \Phi_0 \lambda_j \frac{\partial \Delta \gamma(x,t)}{\partial x} \bigg|_{x=x_n \times \lambda_j}. \]

From the **WS** simulation, we obtain $\Delta \gamma$, the current flowing through each junction, and the voltage across each junction as functions of space and time. Therefore, by identifying the cores of vortices, we can map out the trajectories of vortices in a space-time plot. The cores of vortices are identified as points where $\Delta \gamma$ is an odd multiple of $\pi$.
creased, the vortices are expelled further and further away from the creation event, and eventually additional VAV pairs are generated in the first half of a rf cycle (Fig. 13).

We interpret the discrete motion of vortices [seen in Fig. 13(a)] as a result of the simultaneous breakdown of many neighboring junctions. Considering the current distribution that we apply to the junctions in the ERSJ model as shown in Fig. 10, many junctions can experience currents exceeding their critical current and break down together. This is because the length scale of the current distribution variation is set by the loop size, which is $\sim 10 \mu \text{m}$. Since the shunted junctions are experiencing ac currents at GHz frequencies, they may not break down spontaneously when they experience currents exceeding $I_c$ (Josephson critical current). The dashed ovals in Fig. 12 mark the times and junctions which experience $I > I_c$. These simulations indicate that junctions break down only when they experience $I > I_c$ over a significant portion of time in each period.

As the currents reverse direction in the second half of a rf cycle, another VAV pair with opposite polarity to the pair in the first half of a rf cycle is nucleated at the center of the junction (as pointed out by arrow 1 in Figs. 12 and 13). These new vortices rapidly move out to the nearly stationary locations of the previous pair and annihilate them. As the power is increased, the VAV pairs in the second half of a rf cycle can even be nucleated near stationary vortex locations (as pointed out by arrow 2 in Fig. 13) and annihilate the vortex at the next stationary location.

The flux profile along the junction at $t=T/2$ is also shown with the trajectories in Fig. 13(b). The spikes in the flux profile correspond to the stationary locations of the vortices. However, the locations of the spikes are fixed throughout the whole rf cycle, and do not move with the vortex trajectories. It is noted that only when a vortex moves to one of the locations does the corresponding spike contain one integer flux quantum. Otherwise, the total flux beneath the spike is less than one flux quantum. Also note that the net flux in the junction remains zero at all times.

In summary, the simulation shows that nucleation of VAV pairs takes place in the first half of the rf cycle and annihilation in the second half of the rf cycle. The discrete motion of the VAV pairs is likely due to the simultaneous breakdown of many junctions which experience $I > I_c$ over a significant portion of an rf period. The VAV pairs are nominally nucleated at the center of the junction (directly beneath the probe). As the input microwave power to the junction increases, more VAV pairs are nucleated and pushed away from the center in the first half of rf cycle. In this case, VAV pairs are annihilated by another set of VAV pairs nucleated in the second half of the rf cycle at locations next to each vortex and antivortex. From the time-reversal asymmetries of the vortex trajectories, we find that this nucleation and annihilation process (vortex motion) is responsible for the observed second harmonic generation.

**CONCLUSION**

We have demonstrated local measurement of Josephson nonlinearities on an isolated YBCO bicrystal grain boundary. Spatially resolved harmonic generation images of this boundary are shown, and imply the ability of our microscope to measure nonuniformity along the boundary. Harmonic generation from the grain boundary is simulated by the ERSJ model, which agrees reasonably well with experimental data in the aspects of the magnitude and spatial distribution of both second and third harmonics. Vortex dynamics is also evaluated through this model. We also extract the nonlinear scaling current density $J_{NL}$ for the Josephson nonlinearity in our sample, and find $J_{NL} \approx 1.5 \times 10^5 \text{ A/cm}^2$ at 60 K.

**ACKNOWLEDGMENTS**

We gratefully acknowledge the technical support of Dr. Steve Whiteley in performing simulations using WRSPICE™, assistance from Gregory Ruchti and Marc Pollak in HFSS simulations, help from Dragos I. Micrea in taking harmonic generation data, and discussion with Dr. C. J. Lobb. We also acknowledge the support from Grant Nos. NSF/GOALI DMR-0201261, and the UMD/Rutgers Grant No. NSF-MRSEC DMR-00-80008 through the Microwave Microscope Shared Experimental Facility.

---

*Present address: P.O. Box 118440, Department of Physics, University of Florida, Gainesville, FL 32611-8440, USA.

30 Amuneal Manufacturing Corp., 4737 Darrah Street Philadelphia, PA 19124, USA.
32 The fact that the transition occurs near the $T_c$ determined by ac susceptibility implies that the microwave probe does not increase the temperature of the sample significantly. Other discussion ruling out systematic errors are presented in Refs. 27, 29, and 31.
33 M. Tinkham, *Introduction to Superconductivity*, 2nd ed. (McGraw-Hill, New York, 1996), p. 200. [Taking $I_cR_n=J_cR_s$, where $R_s$ is the specific normal resistance, and $2\Delta(0)/k_BT_c\approx 6$, $J_c$ is estimated to be on the order of $10^6$ A/cm$^2$ at 60 K.]