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Symmetry breaking and nonlinear electrodynamics in the ceramic superconductor $YBa_2Cu_3O_7$

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For bulk samples of the ceramic superconductor $YBa_2Cu_3O_7$ in a radio-frequency magnetic field and in a dc magnetic field H_0, we observe novel nonlinear behavior including very-high-order odd-harmonic generation if $H_0=0$. If $H_0 \neq 0$, there is additional even-harmonic generation. The even-harmonic power displays a very sharp dip at $H_0 \sim 0$, of width $\sim 100\ \mu G$. This and related behavior can be semiquantitatively understood by modeling the system as a suitably averaged collection of flux-quantized supercurrent loops containing Josephson junctions.

There is considerable evidence that the granular microstructure of polycrystalline samples of the ceramic superconductor $YBa_2Cu_3O_7$ (Y-Ba-Cu-O) leads to properties similar to those of spin glasses, owing to the inclusion of Josephson junctions or other weak links between superconducting "grains." For example, models[1] of loop-coupled superconducting clusters have been used to interpret data from susceptibility measurements,[2] and low-field nonresonant microwave absorption.[3] The decoupling of these grains by a small external field is believed to account for the observed sharp drop of the critical current density in transport measurements.[4] We report here a new class of experiments on this material, revealing novel nonlinear behavior when driven by a radio-frequency magnetic field $H_1 \sin(\omega t)$: extensive generation of all odd harmonics $n\omega$ up to at least $n=23$; and symmetry breaking by a small dc field H_0, leading to even harmonics at $n=2,4,6,\ldots$. The even harmonic power shows a sharp decrease at $H_0 \to 0$ in a surprisingly narrow range $\Delta H_0 \sim 100\ \mu G$. After describing the experimental results we present a specific model which gives a semiquantitative account of these phenomena.

Apparatus. The experiments are performed on cylindrical samples ($2\times 10\ mm^2$) of Y-Ba-Cu-O, placed in a solenoid, and subject to three uniform coaxial magnetic fields: dc field H_0; ac field $H_1 \sin(2\pi f_0 t)$, $10^2 < f_0 < 10^5$ Hz; and a repetitive scan field of frequency $\sim 10^{-2}$ Hz. By a balancing arrangement we obtain a signal voltage V_s arising solely from the electrodynamics in the sample, and proportional to the time derivative of the net circulating currents induced in the sample by the driving field H_1. The signal is processed by an analog spectrum analyzer, allowing the power spectral components $P(nf_0)$, $n=1,2,3,\ldots$, to be studied as a function of H_0, H_1, and the sample temperature T. The signal V_s is simultaneously processed by a phase-sensitive coherent detector with output voltage V_c.

Data. The principal features of the nonlinear behavior reported here were found in a variety of samples of ceramic pellets and powdered Y-Ba-Cu-O; we believe the features to be generic. All the data, Figs. 1–5, were taken with a powdered sample;[5] the sample was sealed in a quartz tube, and cooled by immersion in liquid N_2 in "zero" magnetic field ($H_0 \lesssim 1$ mG). The power spectrum, Fig. 1(a), shows surprisingly extensive generation of all odd harmonics up to at least $n=23$. When a dc field $H_0=1.0$ G is applied, even harmonics also appear [Fig. 1(b)]. As $H_0 \to 10$ G the even and the odd harmonics attain roughly equal intensities. A key property of the even harmonics, not detectable from the power spectra, was discovered by examining the phase-coherent signal V_c at $2f_0$ as the dc field was scanned through zero [Fig. 2(a)]: V_c changes sign as it crosses through zero field, i.e., the

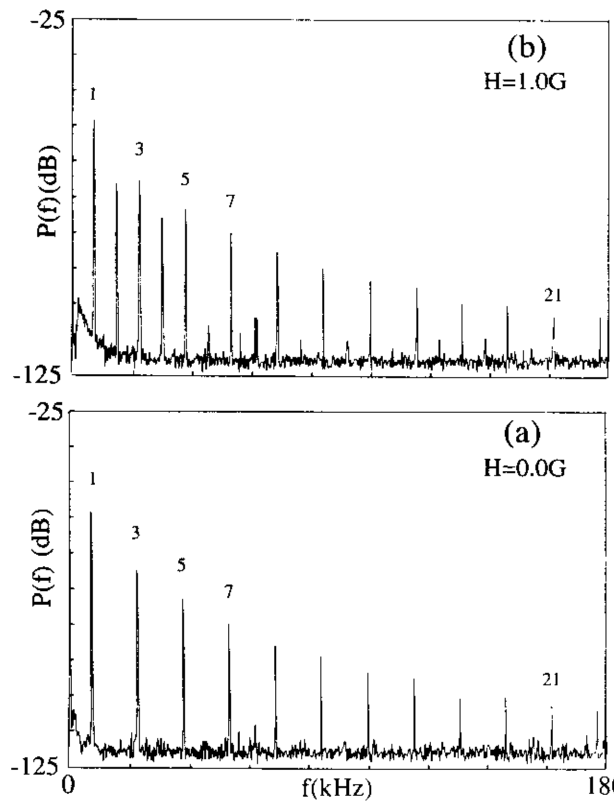

FIG. 1. Power spectra for powdered $YBa_2Cu_3O_7$ at $T=77$ K, driven by an ac field at $f_0=7.7$ kHz, of amplitude $H_1 \approx 14$ G. (a) dc field $H_0=0.0$ G. Odd harmonics $n=3,5,\ldots$ are generated. (b) In a parallel dc field $H_0=1.0$ G, even harmonics $n=2,4,\ldots$ also appear, owing to symmetry breaking by the dc field. The unit of the vertical scale is $P(\text{db})=20\log_{10}V_s+13$, where V_s is the rms signal voltage.

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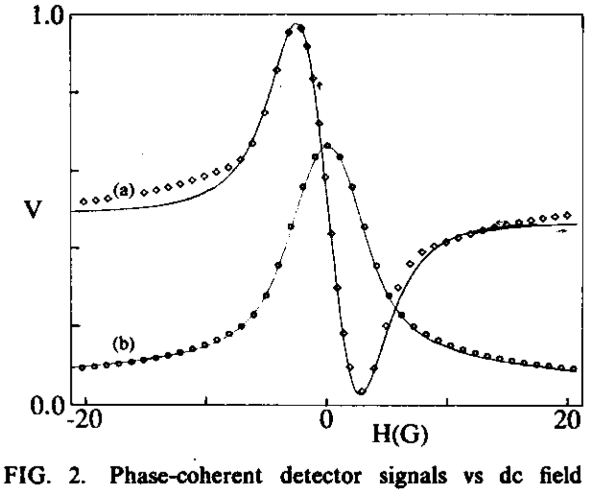

FIG. 2. Phase-coherent detector signals vs dc field for powdered Y-Ba-Cu-O at $T=77$ K, driven at $f_0=52.5$ kHz, amplitude $H_1=2.3$ G. (a) Solid line, observed component at $n=2$; diamonds, model prediction, see text. (b) Solid line, observed component at $n=1$; circles, model prediction, see text.

circulating currents of all even harmonics are odd functions of H_0.

For a more graphic display of this phenomena the detector coil was resonated by an external capacitor at $f_c=341$ kHz, and the envelope of V_s plotted [Fig. 3(a)] as the drive frequency f_0 was swept from 0 to 200 kHz. In the low-frequency wings of the coil response there appear peaks at f_c/n for $n=3,5,\ldots,$ corresponding to the odd harmonic peaks in Fig. 1(a). When $H_0=0.5$ G, additional peaks appear at even harmonics $n=2,4,\ldots$ [Fig. 3(b)] which were observed to reverse sign as H_0 was reversed in direction. The addition of a dc magnetic field, however small, can be viewed as a symmetry-breaking operation, or in the language of nonlinear dynamics, as a symmetry-breaking bifurcation by a control parameter H_0.

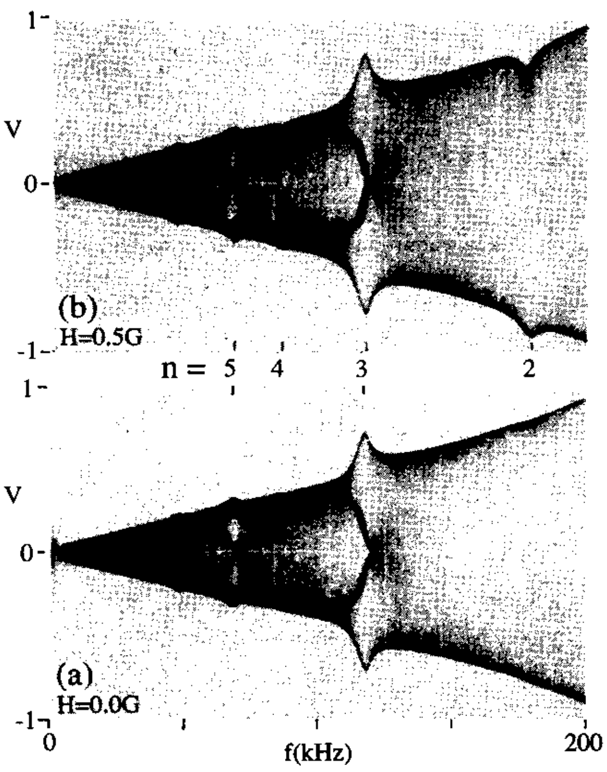

FIG. 3. Subharmonic response voltage V_s of $YBa_2Cu_3O_7$ at 77 K in a coil resonant at $f_c=341$ kHz vs the driving frequency. (a) dc field $H_0=0.0$ G; odd subharmonic peaks are observed. (b) $H_0=0.5$ G; even subharmonic peaks at $n=2,4,\ldots$ are also observed; the polarity of these peaks depends on the sense of H_0 relative to H_1.

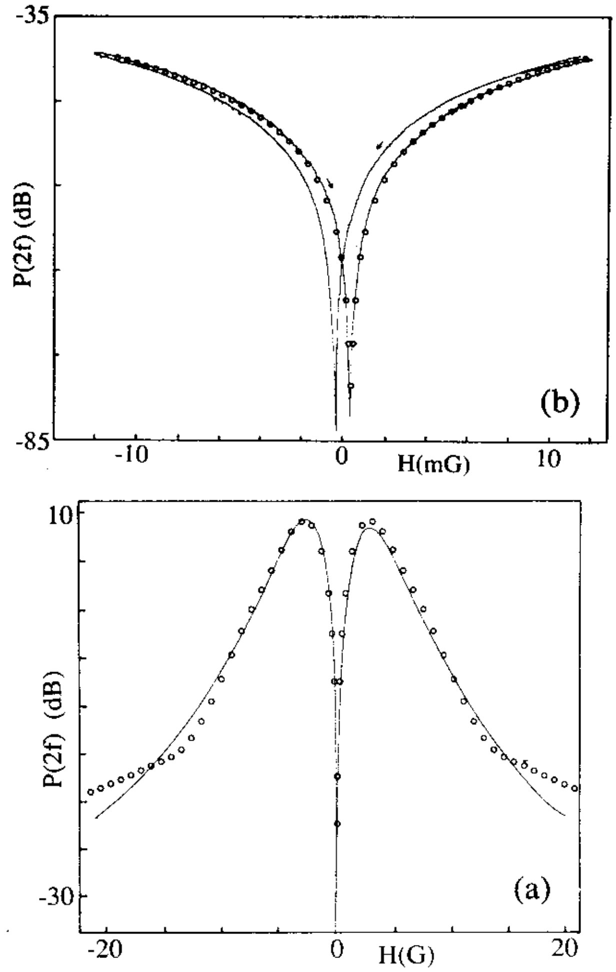

FIG. 4. (a) Solid line: second harmonic intensity $P(2f_0)$ vs dc field H_0 for Y-Ba-Cu-O at 77 K, $f_0=52.5$ kHz, $H_1=2.3$ G. Circles: model prediction, Eq. (4). (b) Solid line: $P(2f_0)$ vs H_0, expanded by factor 2000; the dip has a width of $\Delta H_0 \sim 200$ μG and a small hysteresis. Circles: model prediction, Eq. (4).

As H_0 is scanned the power $P(2f_0)$ of the second harmonic reveals two features in Fig. 4(a): a *broad smooth peak* of width ~ 5.3 G with a deep *narrow dip* at $H_0=0$. By expanding the scan resolution by 2000× [Fig. 4(b)] the width is seen to be less than 200 μG at its 3-dB points. To observe this remarkably sharp dip in the bulk material it was necessary to cool in zero field to reduce the remanent local magnetic fields due to pinned fluxoids. The hysteresis shown, ~ 600 μG, is somewhat dependent on scan rate and range. Plots of $P(nf_0)$ show behavior similar to Fig. 4(a) at other even harmonics, whereas odd harmonics show only a broad peak, without the central dip.

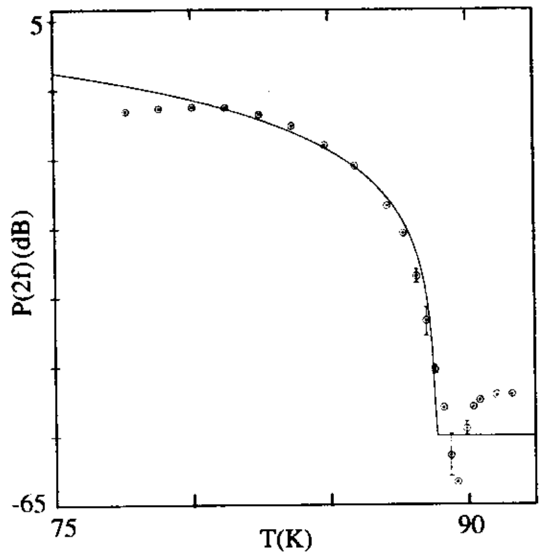

FIG. 5. Circles: observed harmonic intensity $P(2f_0)$ vs temperature T for Y-Ba-Cu-O at $T=77$ K, $f_0=52.5$ kHz, $H_0=2.7$ G, $H_1=2.3$ G. Solid line: computed expression from BCS theory, Eq. (3), for behavior of a Josephson junction assuming $T_c=88.5$ K.

Model. The phenomena in Figs. 1–4 have not been previously reported in Y-Ba-Cu-O or in any other granular superconductor. We present a simple zero-order model that is in semiquantitative agreement with much of these data. We assume, with Refs. 1–4, that Josephson junctions lie at the heart of the matter, and suppose that the ac field induces current loops within and between the superconducting "grains," presumed to be in contact through Josephson tunnel junctions or other weak links. The current loop within a grain will not be influenced by fields much less than $H_{c1}\sim 10^3$ G, and will not be considered in this paper. Although a typical intergranular current loop may be of tortuous shape and intersected by many weak links, we retain in the prototype only two essential features:[6] a single junction of area s in series with a ring-shaped loop of area S, normal to the applied fields. The bulk material will then be represented by suitable averaging over a collection of prototype loop junctions with a distribution of areas. Flux quantization in the loop specifies the loop current $I(t)=I_c\sin(2\pi\Phi/\Phi_0)$, where I_c is the junction critical current and Φ is the total flux through the loop due to all fields, which, by neglecting the loop inductance are just the applied dc and ac fields. In units of the flux quantum $\Phi_0=hc/2e$, these are $\alpha\equiv 2\pi SH_0/\Phi_0$ and $\beta\equiv 2\pi SH_1/\Phi_0$, so that

$$I(t)=I_c\sin[\alpha+\beta\sin(\omega t)] \ . \tag{1}$$

Expansion in a Fourier series yields the current components in terms of Bessel functions $J_n(\beta)$:

$$I_0=I_cJ_0(\beta)\sin\alpha \ , \tag{2a}$$

$$I_1=I_cJ_1(\beta)\cos\alpha\sin(\omega t) \ , \tag{2b}$$

$$I_2=I_cJ_2(\beta)\sin\alpha\cos(2\omega t) \ , \tag{2c}$$

$$I_3=I_cJ_3(\beta)\cos\alpha\sin(3\omega t),\dots \ . \tag{2d}$$

The junction critical current can be written as[7]

$$I_c=\left[\frac{\pi\Delta(T)}{2eR_n}\tanh\left(\frac{\Delta T}{2kT}\right)\right]\left(\frac{\sin x}{x}\right) \tag{3}$$

with $\Delta(T)$ the gap parameter, R_n the normal resistance; the $(\sin x)/x$ factor represents diffractionlike effects arising within the junction, where $x\equiv\pi sH_0/\Phi_0$. Since I_c is an even function of H_0 we see from Eq. (2) that even harmonic currents will be odd functions of H_0 and vice versa; this is in agreement with our data on bulk materials. We use Eqs. (2) and (3) to predict, with suitable averaging, the dependence on H_0, H_1, and T of our measured quantities.

From Eq. (3), for all harmonics the expected temperature dependence of harmonic power is $P(nf_0,T)\sim[F(T)]^2$, where $F(T)$ is the temperature dependence of I_c. The data for $n=2$ (Fig. 5) are consistent with this expression in its high-temperature limit using, from the Bardeen-Cooper-Schrieffer (BCS) theory,[7] $\Delta(T)/\Delta(0)=1.74(1-T/T_c)^{1/2}$, with $T_c=88.5$ K. Data for $n=3$, 4, and 5 are similar, but all show fluctuations at $T_c\pm 2$ K. We believe, however, that the temperature dependence of the phenomena reported in this paper at $T=77$ K is essentially that of the critical current of a Josephson junction.

We assume all loop and junction currents are time coherent, being driven by the same H_1 field. Focusing on the prediction of $P(nf_0,H_0)$ for even harmonics we note $I_n(H_0)\sim[(\sin x)/x]\sin\alpha$, where x and α both depend on H_0 but by different factors: junction area s and loop area S, respectively. We found that the data can be reasonably fit by the expression

$$P(nf_0,H_0)=\left[\left\langle\frac{\sin(Ax)}{Ax}\right\rangle_A\right]^2\langle\sin^2(A\alpha)\rangle_A \ , \tag{4}$$

where a Gaussian distribution of areas s and S is assumed, with a probability distribution function $P(A)=\exp[-(A-1)^2/2\sigma^2]$, $A=0$ to 2, where σ is the standard deviation. Equation (4) is plotted in Fig. 4(a) for $\sigma=0.5$, average junction areas $\bar{s}=2.0\times10^{-8}$ cm^2 and loop area $\bar{S}=0.6\bar{s}=1.2\times10^{-8}$ cm^2, all selected to fit the data in the region $H_0=1$ to 10 G. The overall agreement between model and experiment is reasonable, particularly for the sharp dip, expanded in Fig. 4(b), which is well fit over four orders of magnitude by Eq. (4) with the same parameters as in Fig. 4(a). It is clear that the broad peak shape arises from the junction characteristics and the sharp dip from the loop, and that both are required in the prototype. Other samples of Y-Ba-Cu-O require different values of $\bar{s}$ and $\bar{S}$, which must depend on the microstructure. Our values for $\bar{s}$ and $\bar{S}$ are consistent with those of Ref. 3.

To predict the coherent signal V_c for $n=2$ we compute the expression

$$V_c(H_0)\sim\langle I_2(H_0)\rangle_A=\langle[\sin(Ax)/Ax]\sin(A\alpha)\rangle_A,$$

plotted in Fig. 2(a) for $\sigma=0.33$, $\bar{s}=3.5\times10^{-8}$ cm^2, and $\bar{S}=0.6\bar{s}$. For $n=1$, the data are best fit by the rms average $V_c(H_0)\sim\{\langle[\sin(Ax)/Ax]^2\rangle_A\}^{1/2}$, plotted in Fig. 2(b) for $\sigma=0.33$, $\bar{s}=3.2\times10^{-8}$ cm^2.

Although the prediction of the zero-order model for the harmonic power as a function of H_1, $P(nf_0,H_1) \sim \langle [J_n(A\beta)]^2 \rangle_A$, is a reasonable fit to the data for $n=2$, it does not fit so well for $n=3$ and 4; more particularly, it does not explain the relatively large intensities observed at high order, $n \geq 9$, in Fig. 1(a). However the zero-order model neglects the loop inductance. If this is included one obtains a nonlinear differential equation for $I(t)$, which by numerical iteration, predicts significant high-order harmonic intensity, comparable to that of Fig. 1(a).

To summarize, the novel nonlinear electrodynamics of bulk Y-Ba-Cu-O reported in this paper appears to be generic to the granular microstructure and, in particular, to flux quantization of intergranular current loops containing weak links. The key features, including odd and even harmonic generation and very sharp dips in even harmonic power, can be understood from a simple zero-order model using averaging procedures. Higher-order predictions, requiring numerical iterations of coupled nonlinear differential equations, will be reported separately.[8]

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