Nonlinear electrodynamics and nonresonant microwave absorption in ceramic superconductors

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We work out some of the consequences of a simple model for nonlinear electrodynamics and nonresonant microwave absorption in the high-temperature superconductors. The model assumes that these materials contain loops of weakly coupled superconducting grains. As the magnetic field is changed, a loop jumps from one energy state to another, thus experiencing phase slips. This produces both electromagnetic absorption through the normal shunt resistance in the junctions, and induced currents at high-order multiples of the applied frequency. The calculated power spectrum from a single loop, with and without a dc applied magnetic field, is in qualitative agreement with the measurements of Jeffries et al. [Phys. Rev. B 37, 9840 (1988)].

Many experimental studies of the electrodynamics properties of the high-$T_c$ superconductors have been carried out in an effort to characterize these new compounds, and to understand the origin of superconductivity in these materials. The experiments can generally be divided into two classes. One is the measurement of a dc magnetic field ($H_0$) dependent nonresonant microwave absorption, in which most experimental groups have observed large low-field peaks in the superconducting phase. The other experiment is the measurement of the nonlinear radio-frequency response of some property, such as the time-dependent current $I(t)$, and its power spectrum. In this case, a novel nonlinear behavior has been observed, including the generation of very high-order odd harmonics for $H_0=0$, and additional generation of even harmonics for $H_0\neq 0$. A number of qualitative explanations have been advanced for both classes of behavior in a variety of materials. However, no quantitative theoretical treatment has yet been developed to interpret these novel phenomena in the new superconductors.

In this paper, we work out some of the consequences of a simple mechanism to describe both the nonlinear electrodynamics and the nonresonant microwave absorption of simple superconducting clusters. In the model, superconducting grains, each with small dimensions compared to a penetration depth, are weakly coupled into closed loops. These support screening supercurrents in response to an external magnetic field. The system can be described by the Hamiltonian

$$H = -\sum_{i,j} J_{ij} \cos(\phi_i - \phi_j - A_{ij}),$$

$$A_{ij} = \frac{2\pi}{\Phi_0} \int_I A \cdot dl,$$

where $\Phi_0=hc/2e$ is the flux quantum and the integral is taken along a line joining the centers of grains $i$ and $j$. In the presence of randomness and magnetic-field-induced frustration, a complex cluster, composed of many such loops, has numerous competing low-lying states of nearly equal energy. Here we consider, for simplicity, only a single loop without randomness. When such a superconducting loop is exposed to a dc magnetic field $H_0$ perpendicular to the plane of the loop, and an ac magnetic field $H_1\sin(\omega t)$ parallel to $H_0$, the system will experience phase slips by jumping from one energy state to another as the field is changed, or, more precisely, as the flux through the loop changes with time. This phase slip generates a voltage difference between neighboring grains and hence leads to energy absorption through the normal resistance in parallel with the Josephson coupling. Because of the abrupt changes in supercurrents with time, arising from these flux slips, the current will have many higher-order harmonics and hence an unusual power spectrum, which we calculate to be similar to that observed by Jeffries et al. Our model is similar to that proposed by these authors. However, we assume that the current in the loop follows the applied magnetic field, and, in particular, jumps from one state to another, undergoing a phase slip whenever the field crosses an energy cusp. These jumps seem to be responsible for the many higher harmonics seen in their experiments.

We consider a loop of $N$ identical superconducting grains, each with the same coupling energy $J$ and the same distance $a$ between the centers of neighboring grains. The Hamiltonian describing the system is given by Eq. (1). The supercurrent from grain $i$ to grain $j$ is

$$I_{ij} = I_c \sin(\phi_i - \phi_j - A_{ij}) = I_c \sin \left( \frac{2\pi}{N} \left[ I + f(t) \right] \right),$$

where $I_c=2eJ/h$ is the critical current of the junction, $I$ is an integer, and $f(t)=f_0 - f_1 \cos(\omega t)$ is the total flux through the loop, in units of the flux quantum $\Phi_0$. Evidently, $f_0=S\Phi_0/\Phi_0$, $f_1=S\dot{\Phi}_0/\Phi_0$, where $S$ is the loop area. The integer $I$ is determined by the requirement that the loop should be in its state of minimum energy for a given flux.

As the flux $\Phi$ through the loop varies, phase slips will occur when the flux crosses one of the cusps of the energy diagram, i.e., when $\Phi = (n+\frac{1}{2})\Phi_0$, or $f = n+\frac{1}{2}$, $n$ being an integer. At this field, the integer $I$ changes by unity. We also assume that the ac frequency is sufficiently low that this flux slip will occur in a small fraction of a cycle. Flux slips presumably occur on a time scale $\tau = L/R_N$, where $L$ is the self-inductance of the loop and $R_N$ is some characteristic resistance, probably on the order of shunt...
resistance of the junction. For loops of micron dimensions, and resistances of order a few ohms, \( r \) is presumably of the order of \( 10^{-12} \) or smaller, so that, for frequencies in the microwave range or lower, this condition is satisfied for most loops, except perhaps at very low temperatures.

To illustrate these effects, we assume that \( f_0 \) and \( f_1 \) are such that, at most, only the two cusps at \( f = \pm \frac{1}{2} \) and \( f = - \frac{1}{2} \) are traversed in a single cycle (cf. Fig. 1). In order to have phase jumps, we need either \( f_1 \geq f_0 + f_c \) or \( f_0 \geq |f_c - f_1| \), where \( f_c = 0.5 \), corresponding to the first energy cusp. The physics behind these conditions is that the amplitude of either the dc or the ac field must be large enough to produce a phase slip when the field varies with time. Thus these criteria give the thresholds for the external magnetic field to produce the observed nonresonant microwave absorption. In the rest of this paper, for simplicity, we restrict our attention to the weak-field case, i.e., \( f_0 + f_1 < 1 + f_c \), corresponding to the two phase slips at lowest applied flux, although the same mechanism can be applied to the whole range of external fields without any difficulty.\(^{11}\)

We first calculate the power spectrum for the magnetic moment of the superconducting loop. We consider “low temperature” in the sense that, at any given field, the loop is assumed always to fall into its state of lowest energy. From the analysis above, the time-dependent phase difference \( \phi(t) \) between two neighboring grains can be written as follows:

\[
\phi(t) = \frac{2\pi}{N} \left[ f_0 - f_1 \cos(\omega t) - 1 + \Theta(t - t_1) + \Theta(t - t_2) - \Theta(t - T + t_2) - \Theta(t - T + t_1) \right] \Theta(f_1 - f_0), \quad 0 \leq f_0 < f_1 - f_c; \tag{4}
\]

\[
\phi(t) = \frac{2\pi}{N} \left[ f_0 - f_1 \cos(\omega t) + \Theta(t - t_2) - \Theta(t - T + t_2) \right], \quad \min(f_c + f_1, f_c + 1 - f_1) > f_0 \geq |f_c - f_1|; \tag{5}
\]

where

\[
t_1 = \frac{1}{\omega} \cos^{-1} \left( \frac{f_0 + f_c}{f_1} \right), \quad t_2 = \frac{1}{\omega} \cos^{-1} \left( \frac{f_0 - f_c}{f_1} \right);
\]

\( T = 2\pi/\omega \) is the period of the ac field; \( \Theta(x) \) is the usual step function; and \( \min(a, b) \) means the smaller of the quantities \( a \) and \( b \). The magnetic moment \( \mu \) of the loop is

\[
\mu = \frac{1}{2c} \sum_{i,j} (x_{ij} \times I_{ij} x_{ij}), \tag{6}
\]

where \( I_{ij} \) is given by Eq. (3), \( x_{ij} = (x_i + x_j)/2 \) is the vector joining the origin to the midpoint between grains \( i \) and \( j \), and \( x_{ij} = x_i - x_j \). We have

\[
\mu(t) = \mu_0 \sin(\phi(t)), \tag{7}
\]

where \( \phi(t) \) is given by Eqs. (4) and (5), and

\[
\mu_0 = \frac{S I_c}{c} \frac{\sin(2\pi/N)}{2\pi/N}. \tag{8}
\]

Expanding \( \mu(t) \) in a Fourier series, assuming \( 2\pi/N \ll 1 \), and using the initial conditions gives

\[
\mu(t) = \mu_0 \left[ \frac{1}{2} a_0 + \sum_{n=1}^{\infty} a_n \cos(n \omega t) \right], \tag{9}
\]

with

\[
a_0 = \frac{4}{N} [a_1 + a_2 - \pi(f_0 + 1)], \tag{10}
\]

\[
a_n = \frac{4}{N} \left[ \sin(na_1) + \sin(na_2) + \frac{\pi f_1 \delta a_1}{2} \right], \tag{11}
\]

where \( a_1 = \omega t_1, \ a_2 = \omega t_2 \), and \( n = 1, 2, 3, \ldots \).

For the special case of zero dc field \( (f_0 = 0 \text{ or } H_0 = 0) \), we obtain \( a_1 = \cos^{-1} (f_c / f_1); \ a_2 = \cos^{-1} (f_c / f_1) = \pi - a_1 \). The magnetic moment of Eq. (9) is then non-zero only for the odd harmonics. Since \( \mu(t) \) is proportional to the current \( I(t) \) circulating in the loop, we can define a power

\[
P(t) \approx I^2(t) = \mu^2(t). \tag{12}
\]

The power spectra calculated from Eqs. (8), (11), and (12) are plotted in Fig. 2. Figure 2(a) shows the spectrum for zero dc field \( (H_0 = 0) \). Because of the symmetry of the system in this case, all even harmonics are zero, and only odd harmonics \((n = 1, 3, 5, \ldots)\) are generated. The spectrum shows a surprisingly extensive generation of odd harmonics up to a large number \( n \). The envelope of the spectrum is \( 1/(2n+1)^2 \), which falls off very slowly with increasing \( n \) (note that Fig. 2 is plotted on a logarithmic vertical scale). Besides this envelope, the spectrum shows some slight oscillations with \( n \), generated by the term \( \sin[(2n+1)a_1] \), so that, for example, the \( n = 19 \) harmonic is stronger than the \( n = 17 \). The spectrum for the case \( H_0 = 0 \) is shown in Fig. 2(b). Because of the symmetry breaking by the nonzero static field, even harmonics now
experiment. These oscillations persist in our calculations irrespective of our initial conditions, or the precise magnitudes of $H_0$ and $H_1$. They also persist when a suitable average is taken over loop orientations.

Next, we make a simple estimate of the nonresonant electromagnetic absorption in these loops, under the assumption that this absorption is determined by losses through the normal resistances in parallel with the Josephson or proximity links in the loops. We begin from the Josephson relation\(^{13}\)

$$V(t) = -\frac{h}{2e} \frac{d\phi}{dt},$$

(13)

describing the voltage across a junction in which the phase difference is $\phi$. Substituting (4) and (5) into (13) gives a voltage which involves a sum of $\delta$ functions in time, and hence leads to an infinite absorption. This result is expected from our simple model, because in the above discussion we have assumed that the system relaxes from one current carrying state to another in time $\tau = 0$ as one of the cusps in the energy diagram of Fig. 1 is crossed. In reality, one expects a finite relaxation time.

To include a finite relaxation time, we take $\tau$ as a parameter which we assume to satisfy $\tau \ll T$, $T$ being the period, and introduce the function

$$\rho_\tau(t) = \frac{1}{2\tau} \exp\left(- \frac{|t|}{\tau}\right),$$

(14)

which has the property $\lim_{\tau \to 0} \rho_\tau(t) = \delta(t)$. Next, we calculate the voltage from Eqs. (4), (5), and (13), but with the substitution $\delta(t) \to \rho_\tau(t)$. The absorption per unit time, denoted $\chi''$, is given by

$$\chi'' = \frac{1}{R_n} \langle V^2(t) \rangle,$$

(15)

where the angle brackets denote a time average over one period. The measured quantity is more closely related to the field derivative of the absorption, which becomes, on substituting (13) into (15),

$$\frac{d\chi''}{dH_0} \approx \frac{2V^2R_n^2}{\pi R_n \Phi_0} \left[ \frac{f_c - f_0}{(f_0^2 - (f_c - f_0)^2)^{1/2}} - \frac{f_c + f_0}{(f_0^2 - (f_c + f_0)^2)^{1/2}} \right] \Theta(f_1 - f_c)$$

(16)

for $0 \leq f_0 > f_1 - f_c$, and

$$\frac{d\chi''}{dH_0} \approx \frac{2V^2R_n^2}{\pi R_n \Phi_0} \left[ \frac{1}{(2\pi)^2 (\tau/T)^2} \left[ f_c - f_0 \right]^2 \right] \left[ f_0^2 - (f_0 - f_c)^2 \right]^1/2$$

(17)

for $\min(f_c + f_1, f_c + 1 - f_1) > f_0 \geq |f_1 - f_c|$. Note that in the limit $\tau \to 0$, this derivative, unlike the absorption itself, becomes independent of $\tau$.

In Fig. 3 we plot $d\chi''/dH_0$ as a function of dc field for fixed amplitude of the ac field and for a representative loop size. The shape of the curve, in this simple model, scales with the area of the loop. It also depends on the relaxation time $\tau$ and on the shunt resistance $R_n$, which in the usual model of a resistively shunted Josephson junction varies inversely with the critical current through the

FIG. 3. Derivative of the absorption $d\chi''/dH_0$ [plotted in units of $V^2\omega^2S/(\pi R_n \Phi_0)$] vs dc field for fixed amplitude of ac field. In this calculation, we have used the values $S = 10^{-6}$, $H_1 = 0.04$ G, and $\tau = 0.01$ T.
junction. The general shape of the absorption derivative of Fig. 3, and in particular the absence of absorption below a threshold dc field corresponding to one-half flux quantum per loop, seems consistent with a number of experiments on ceramic high-temperature superconductors.\textsuperscript{2-8,14} The results for the absorption derivative seem also to agree rather well with measurements on nominally single-crystal samples of YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7−δ},\textsuperscript{15} suggesting, as has also been proposed by Blazey \textit{et al.},\textsuperscript{15} that these materials contain weakly linked loops of characteristic, possibly geometrically determined, areas. In these experiments, the width of the field range over which \(dx''/dH_0\neq0\) is proportional to \(H_1\), the amplitude of the ac field, as is also predicted by the present, single-loop calculation.

The good agreement between the present model and experiments on the nonlinear electrodynamic response of these materials, as well as the consistency with microwave experiments, strongly suggests the existence of superconducting loops of various areas in the ceramic high-\(T_c\) oxides. Of course, a more detailed comparison with experiment would require averaging over a variety of loops of various areas, orientations, as well as the consideration of possibly more topologically complex loops, with flux slips occurring almost continuously as the dc field is varied, rather than only at discrete fields as predicted in the present calculations. Ideally, it would also be desirable to treat the dynamics microscopically rather than to include it only via a parameter \(\tau\). If more complex loops are included, the frequency dependence of the absorption at fixed dc field may differ from the single-loop picture, since the absorption rate depends on the number of flux slips per cycle, as well as the absorption in each slip. More detailed and realistic calculations of this nature will be presented in a subsequent publication.\textsuperscript{11}

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\textsuperscript{11}T. K. Xia and D. Stroud (unpublished).
\textsuperscript{14}Microwave absorption experiments are generally carried out with dc and microwave fields perpendicular. For a randomly oriented loop, however, there will be components of both dc and ac fields oriented parallel to the loop axis, so that the present mechanism still applies.