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Harmonic generation and flux quantization in granular superconductors

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Simple dynamical models of granular superconductors are used to compute the generation of harmonic power in ac and dc magnetic fields. In zero order, the model is a single superconducting loop, with or without a weak link. The sample-average power is predicted by averaging over suitable distribution functions for loop areas and orientations in a dc magnetic field. In a first-order model, inductance and resistance are also included. In all models the power at high harmonics shows strikingly sharp dips periodic in the dc field, revealing flux quantization in the prototype loops.

Shortly after the discovery of the superconducting copper oxides, Müller, Takashige, and Bednorz and Razavi, Koffyberg, and Mitrovic[1] showed that they were granular and had some properties similar to spin glasses.[2] Other unusual properties were soon reported: strong nonresonant microwave absorption in very low fields ($H \sim 10$ Oe);[3] strong dependence of ac susceptibility on the ac field;[4] and a sharp drop in transport critical current in low fields.[5] Recent reviews[6,7] have considered in detail some theoretical aspects of the physical properties of high-T_c granular superconductors. In a previous paper[8] we reported extensive harmonic generation by ceramic samples of $YBa_2Cu_3O_7$ (Y-Ba-Cu-O) driven by an ac magnetic field $H_1 \sin(\omega t)$. For all even harmonics the power $P(n\omega)$ vanishes if the dc field $H_0=0$. These and other properties were explained by an elementary model. In this Communication we extend and generalize the model and show that it further predicts novel behavior: for high harmonics, very sharp periodic dips in $P(n\omega)$ as a function of H_0. We find, in effect, that observation of high harmonics should recover quantitative aspects of flux quantization in the microstructure of the sample. Although spin-glass models of granular superconducting media have been well developed we take here a simpler approach, and consider the dynamical behavior of microscopic currents, which is intrinsically nonlinear, and must be reckoned with directly to model harmonic generation.

Zero-order model. We consider a superconducting sample subject to parallel and uniform dc and ac fields, the total applied field being $H=H_0+H_1\sin(\omega t)$. The sample is assumed to have no electrical contacts but is surrounded by a solenoid; all measurements are made from the voltage induced into this "receiver" coil. The sample is imagined to be composed of superconducting "grains" in contact through weak links, e.g., Josephson tunnel junctions, point contacts, or proximity effects. For low fields $H < H_{c1}$ of the grains, the situation will be modeled by an ensemble of superconducting paths intersected by weak links, the specific prototype being a thin ring-shaped loop of area S_0 in series with a junction of area s_0, such that the flux due to the applied field is $S_0 H\cos(\theta)$ and $s_0 H \cos(\phi)$, respectively. We note that this geometry is similar to that used to model rf superconducting quantum interference devices (SQUID's).[9] In the zero-order model we neglect the flux due to the loop current itself, but reconsider it below. The electromagnetic properties of the sample are then predicted by taking suitable averages over a distribution of areas S_0, s_0 and orientations θ, ϕ. We define the dimensionless quantities

$$\alpha \equiv 2\pi S_0 H_0 \cos\theta/\Phi_0, \quad \beta \equiv 2\pi S_0 H_1 \cos\theta/\Phi_0, \quad \eta \equiv \pi s_0 H \cos\phi/\Phi_0, \tag{1}$$

where Φ_0 is the flux quantum and $\alpha/2\pi$ is just the number of flux quanta in the loop due to H_0, etc. The applied field induces current in the prototype loop, which, for a tunnel junction is given by the Josephson current-phase relation[10]

$$I(t) = I_c(\sin\eta/\eta)\sin\gamma, \tag{2}$$

where $\gamma \equiv \alpha + \beta\sin\omega t$, and I_c is the junction critical current.[11] We assume that the junction area s is sufficiently small that the diffraction term $[\sin(\eta)/\eta] \approx 1$, and consider only the Fourier components of $\sin\gamma$, arising from flux quantization of the loop:

$$I_{In} \propto J_n(\beta)\sin\alpha\cos(n\omega t), \quad n \text{ even}, \tag{3a}$$

$$I_{In} \propto J_n(\beta)\cos\alpha\sin(n\omega t), \quad n \text{ odd}, \tag{3b}$$

where $J_n(\beta)$ is the Bessel function of integer order.

We assume that each current loop induces a receiver coil signal voltage $v_n(t)$ proportional to $S_0 \cos\theta dI_I/dt$. If the sample were composed of only one loop the signal power $P(n\omega) \propto v_n^2$ for all harmonics would be periodic in H_0 due to flux quantization with period $\Delta\alpha=\pi$, i.e., $\Delta H_0 = \Phi_0/(2S_0\cos\theta)$ between dips, corresponding to the period of $\cos^2\alpha$ or $\sin^2\alpha$. We characterize the ensemble of current loops by a uniform distribution of orientation angles and an area distribution function $F(A)$, with $A \equiv S/S_0$. All the loops are assumed to be coherently driven, so that the total signal voltage $V_n(t)$ at some harmonic $n\omega$ can be represented by the algebraic sum of all $v_n(t)$. The sample-average signal amplitude $\langle V_n \rangle$ is computed by the expression, for odd n,

$$\langle V_n\rangle = n\omega \int_{A=0}^{\infty} dA \int_{\theta=0}^{\pi/2} A\cos\theta J_n(A\beta) \times \cos(A\alpha)F(A)\sin\theta\, d\theta/G; \quad (4)$$

for even n replace $\cos(A\alpha)$ by $\sin(A\alpha)$; G is the normalizing factor $\int\int F(A)\sin\theta\, dA\, d\theta$.

To examine the effects of averaging on $P(n\omega)$ vs α we take as an *example* a Gaussian distribution function for loop areas

$$F(A) = \exp[-(A-1)^2/2\sigma^2] \quad (5)$$

peaked at $S=S_0$, with standard deviation σ. First, to represent a single loop we take $\sigma=0$, and $\cos\theta=1$ in Eq. (4) and compute $P(n\omega)$, plotted in Fig. 1(a), which shows the expected periodicity $\Delta\alpha=\pi$; this plot is valid for all values of β and odd n. Next, for standard deviation $\sigma=2$, $\beta=5$, and $n=1$ we compute $P(\omega)$, plotted in Fig. 1(b). We see that the periodicity is "averaged out" for a distribution of areas and orientations. However, for large values of n the result is different. For $\sigma=2$, $\beta=5$, and $n=15$ we compute and plot $P(15\omega)$ in Fig. 1(c), finding deep and almost periodic dips, with an average dip spacing $\overline{\Delta\alpha}=1.03$. If we omit the averaging over θ in Eq. (4) the plot is essentially the same as Fig. 1(c), with $\overline{\Delta\alpha}$ smaller by 1.5%. For increased σ, the plots are very similar, with decreased $\overline{\Delta\alpha}$; the pattern converges for $\sigma \gtrsim 2$. Essentially, the same behavior is found for other values of n, with $\overline{\Delta\alpha} \propto n^{-1}$ for $n \gg 1$. If we include the $[\sin(\eta)/\eta]$ term in Eq. (2), the computed shapes of $P(n\omega)$ for small n are modified to an extent depending on the distributions of S and s. However, for large n, the shapes are not sensitive to the details of either S or s distributions, as long as they are monotonically decreasing at large values.

The principal result of this Communication is that this model of granular superconductors, even with a broad distribution of areas and grain orientations, predicts sharp and almost periodic dips in the harmonic power as the dc field is varied, giving evidence of flux quantization arising from the loop of the model. One would have naively expected the periodic flux quantization of a loop to be generally averaged out, this is not so for high harmonics. Other distribution functions $F(A)$ also yield sharp dips in $P(n\omega)$ vs α.

Loop model. We now explore the possibility that the current-flux relation of an individual current loop is not sinusoidal, as in Eq. (2), but still periodic with period Φ_0. There are several conceivable cases in which this occurs: (i) The current-phase relation of the weak links may deviate from Eq. (2), which was derived by Josephson for the case of a weakly coupled tunnel junction; (ii) the prototype loop has a *large* number of identical junctions, and the change in the loop current is then controlled by the change in phase-winding number of the loop rather than by the current-phase relation of individual junctions;[2] (iii) screening by the loop current effectively gives a skewed periodic current-*applied* flux relation, as in the case of rf SQUID's;[9] and (iv) there may be current loops which simply are superconducting without any junction or weak link in their paths. We now consider this special case, although the results should be applicable to the others. Fluxoid quantization in a loop requires that[12]

$$\int_S \mathbf{H}\cdot d\mathbf{S} + (m^*c/2e)\int_l v\cdot dl = n\Phi_0, \quad n=0,1,2,\ldots,$$

which, for a thin ring of radius R, yields the velocity v of the superconducting electrons and, hence, the current density $I_l \sim v = \hbar(n-\Phi/\Phi_0)/(m^*R)$, where $\Phi = H\pi R^2$ is the applied flux through the ring. The kinetic energy is proportional to $(n-\Phi/\Phi_0)^2$. As the flux Φ/Φ_0 is increased we allow n to switch from $n=0$ to 1, etc., maintaining the system in a minimum kinetic energy state. The current I_l

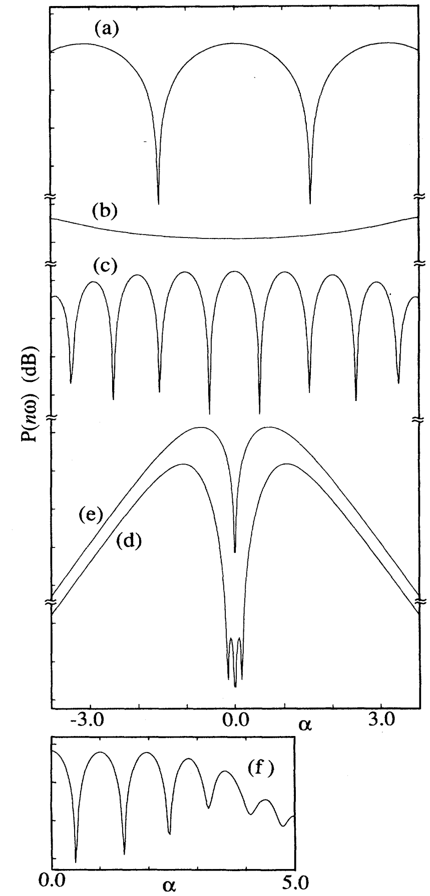

FIG. 1. (a) Harmonic power $P(n\omega)$ vs α, computed from Eqs. (4) and (5) with $\sigma=0$, corresponding to a single loop, with no averaging; figure is valid for all values of β and odd n, and shows periodicity $\Delta\alpha=\pi$ due to flux quantization of the loop. (b) Harmonic power $P(\omega)$ vs α computed from Eqs. (4) and (5) with $\sigma=2$, $\beta=5$, and $n=1$; sample averaging washes out the sharp dips of Fig. 1(a). (c) Harmonic power $P(15\omega)$ vs α, computed from Eqs. (4) and (5) for $\sigma=2$, $\beta=5$, and $n=15$; sample averaging does not wash out the sharp dips. (d) Harmonic power $P(2\omega)$ vs α, computed from Eq. (7) for $n=2$, $\beta=0.5$, and a monotonically decreasing distribution function $F(A)$. (e) Harmonic power $P(2\omega)$ vs α, computed from Eq. (4) for $n=2$, $\beta=0.5$, and same $F(A)$ as in (d). (f) Harmonic power $P(15\omega)$ vs α, computed for the first-order model, Eq. (8), with $\beta=5$, $\kappa=0.3$, and $L_0=0.35$, using $F(A)$ from Eq. (5) and $\sigma=2$.

is then a sawtooth function of (Φ/Φ_0) which we write as the Fourier series

$$I_l=\sum_{m=1}^{\infty}(-1)^{m+1}m^{-1}\sin(m2\pi\Phi/\Phi_0)\,, \tag{6}$$

with $2\pi\Phi/\Phi_0$ to be identified with $\alpha+\beta\sin(\omega t)$ in Eq. (1). Following the same procedure used to obtain Eq. (4), we use Eq. (6) to find, for odd n, the sample-average signal voltage components

$$\langle V_n\rangle=n\omega\sum_{m=1}^{\infty}(-1)^{m+1}/e^{m-1}\int_0^{\infty}dA\int_0^{\pi/2}A\cos\theta J_n(mA\beta)\cos(mA\alpha)F(A)\sin\theta\,d\theta/G\,, \tag{7}$$

where for convergence we have replaced $1/m$ in the summation in Eq. (7) by $\exp(-m+1)$ to round off the high harmonics of an otherwise infinitely sharp sawtooth. From Eq. (4), we see that the zero-order model is merely the first term of Eq. (7). Plots of $P(n\omega)$ vs α, computed from Eq. (7) are found to be quite similar to the zero-order model; however, at small values of β the loop model predicts additional structures. Shown in Fig. 1(d) is $P(2\omega)$ vs α computed from Eq. (7) for $\beta=0.5$. This is to be compared to $P(2\omega)$ for the zero-order model, Fig. 1(e), computed for $\beta=0.5$ and the same $F(A)$ as Fig. 1(d).

First-order model. So far we have made the assumption that the self-induced flux due to the current circulating in the loop could be neglected. We have also neglected the resistive current flowing in the loop. However, these assumptions ignore dissipation in the sample which can be caused by either the resistive current or bulk-pinning hysteresis.[13] It is equivalent to assuming that the sample magnetization $M(H)$ has the same functional form for both dc and ac magnetic fields. A result is that Eqs. (3) only give inductive components in the receiver coil signal. So we generalize the zero-order model by assigning a self-inductance to the loop and adding a resistance R in shunt with the junction. The loop current is then given by[14] $I(t)=I_c\sin\gamma+V/R$ where $V=(\hbar/2e)d\gamma/dt$ and $\gamma=\alpha+\beta\sin(\omega t)-2\pi LI/\Phi_0$. Combining these expressions one obtains

$$\omega^{-1}dI_1/dt=(\kappa L_0)^{-1}\{\sin[\alpha+\beta\sin(\omega t)-L_0I_1]-I_1\}+(\beta/L_0)\cos(\omega t)\,, \tag{8}$$

where $I_1\equiv I/I_c$, $\kappa\equiv\hbar\omega/2eRI_c$, $L_0\equiv2\pi LI_c/\Phi_0$. For given values of parameters α, β, κ, and L_0 and loop area $S=AS_0$, Eq. (8) is numerically iterated to yield dI_1/dt. This quantity is averaged over a Gaussian distribution of areas, Eq. (5), with L_0 assumed to vary as $A^{1/2}$, to obtain $\langle V(t)\rangle_A$, whose spectral components are computed using a fast Fourier transform algorithm, yielding real and imaginary components $V_{\mathrm{real}}(n\omega)$ and $V_{\mathrm{imag}}(n\omega)$. The corresponding power $P(n\omega)$ is plotted versus α in Fig. 1(f) for $n=15$, $\beta=5$. Although there is a clear correspondence with Fig. 1(c), one sees that now the inductive and dissipative terms have a different dependence on α so that the dips have a more complex pattern. We believe this model may serve as a *phenomenological* theory for the interaction of eddy-current and *bulk* hysteretic losses in granular superconductors.[13]

We have not considered some important aspects of the more general problem of high-T_c granular superconductors, such as the critical state and fluxon nucleation[15] which are beyond the scope of this Communication.

To summarize, in several simple dynamical models of granular superconducting samples in low magnetic fields we have computed the expected harmonic power $P(n\omega)$ generated by an ac driven superconducting loop, averaged over a wide distribution of areas and orientations in a dc magnetic field. For high harmonics, a strikingly sharp periodic behavior in magnetic field is predicted, revealing flux quantization of the loop of the model. The plots of Figs. 1(c) and 1(f) are reminiscent of those measured for the magnetic field behavior of the resistance of loop-coupled periodic arrays of Josephson junctions[16] and also the computed behavior of superconducting wire networks;[17] in these cases, the systems consist of arrays of essentially identical structures. We point out that in the case H_0 parallel to H_1, to lowest order in H_1 and in the low-frequency limit, measurements at the nth harmonic are equivalent to taking the nth derivative (d^nM/dH^n) of the nonlinear magnetization $M(H)$, and can recover averaged-out structure, even in bulk powder, that is unresolved in magnetization, susceptibility, magnetoresistance, and critical temperature measurements. Onset of additional structure is predicted for loops as the driving field becomes small. Dissipation is included in a higher-order version of the model. The main features of the model are experimentally observed, to be separately reported for $Bi_4Sr_3Ca_3Cu_4O_x$ (Ref. 18) and for $YBa_2Cu_3O_7$ (Ref. 19), an example of which is shown in Fig. 2. Experiments in which H_0 is perpendicular to H_1 are also under way.

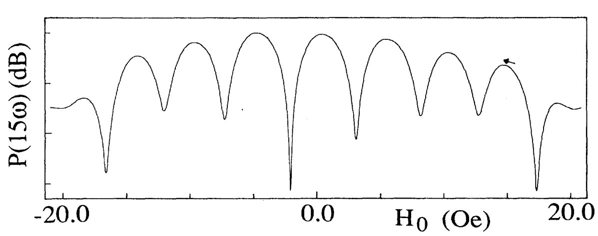

FIG. 2. Measured harmonic power $P(15\omega)$ vs dc field H_0 for $H_1=23$ Oe for the sample of powdered $YBa_2Cu_3O_7$ at $T=77$ K (from Ref. 19).

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