Harmonic susceptibilities of (Bi, Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_y$ bulk superconductors

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Abstract

Even harmonics of susceptibility have been measured as functions of dc magnetic fields, the ac field amplitude and frequency for (Bi, Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_y$ bulk superconductors with magnetic fields parallel to the slab sample. The experimental results are compared with theoretical susceptibility curves based on the numerical calculation of the flux diffusion equation. The theoretical results are in good agreement with the temperature-, field- and frequency-dependent features of the experimental data. The frequency independent critical state model breaks down in explaining all these results even though its generalization can generate even harmonics of susceptibility. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The measurements of the fundamental and harmonic ac susceptibilities $\chi_n = \chi'_n - iy_n$ ($n = 1, 2, \ldots$) have long been recognized as an important tool in the verification of physical models for flux pinning and motion in the mixed state of high temperature superconductors [1–9]. In comparison with resistivity measurements, the susceptibility technique may provide more information about the flux dynamics since the amplitude and frequency of ac magnetic field can be varied in addition to temperature and dc magnetic field. The harmonic susceptibilities were first interpreted by Bean’s critical state model, in which the critical current density $j_c$ was assumed to be independent of the local magnetic field [1]. However only odd harmonics were predicted by the original Bean model of critical state. It was found later [3,4] that even harmonics could be obtained in the framework of the critical state model by taking into account the field dependence of $j_c$, for example Kim–Anderson model. A more recent study showed that the global ac response was described by a single scaling parameter $\delta$, i.e. the effective penetration depth [10]. This implies that the measured curves of harmonic susceptibilities as a function of any experimental variables can be reduced to a universal curve that describes the harmonic susceptibilities versus the scaling parameter $\delta$.

Some of the experimental features of the temperature dependence of $\chi'_n$ and $\chi''_n$ have been

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successfully explained by the critical state model and its generalization. However, the observed frequency dependence of the fundamental and higher harmonics cannot be described within the framework of the critical state model. As a consequence, the simultaneous presence of hysteretic and dynamic losses has to be included in the model description. Recently, the magnetic relaxation effects have been considered in order to explain the frequency dependence of third harmonic susceptibility [11]. The numerical methods have also been applied to resolve the flux diffusion equation. Within such approach, the time evolution of flux profiles and magnetization curves have been calculated [12,13]. Qin et al. have reported the calculation results of ac susceptibility in the presence of a dc bias magnetic field [14–16]. In this work we investigate the effect of dc magnetic field $H_d$, ac field amplitude $H_{ac}$ and frequency $f$ on the harmonic ac susceptibilities of $(\text{Bi, Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (Bi2223) bulk superconductors. We find the experimental data are in good agreement with the theoretical behavior deduced from the numerical calculations of the flux diffusion equation. Since there have been many papers on odd harmonics [11,17], we focus our attention on the temperature dependence of even harmonics.

2. Experiments

The $(\text{Bi, Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ samples were prepared by a solid state reaction from $\text{Bi(NO}_3)_3$, $\text{Pb(NO}_3)_2$, $\text{CaCO}_3$, and $\text{CuO}$ powders. A nominal composition of $\text{Bi}_{1.8}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ was chosen as suggested by Takada et al. [18]. The mixture was ground and preheated in air at 700°C for 24 h. It was then cooled to room temperature in the furnace. It was reground, pressed into pellet, and subsequently sintered in air at 840°C for 80 h, and cooled in the furnace. We obtained the pure Bi2223 phase as confirmed by X-ray diffraction. The sintered pellet was cut into a rectangular slab with dimensions $6 \times 5 \times 1 \text{ mm}^3$. The electrical resistivity of the sample was zero below 107.9 K.

The complex ac susceptibility measurements were carried out in a home-built susceptometer with a three-coil mutual inductance bridge and a two-position background subtraction scheme at different frequencies and amplitudes of ac magnetic field $H(t) = H_{ac}\sin(\omega t)$. The applied magnetic field also had a dc component $H_d$. For all measurements, the magnetic fields were parallel to the slab. A function generator was used to provide the ac signal for the driving coil and the reference signal for the lock-in amplifier. The voltage outputs from the EG&G 5302 lock-in amplifier were proportional to the real part $\chi'_n$ and imaginary part $\chi''_n$ of the ac susceptibilities. A Lake Shore 340 temperature controller was used to monitor the temperature of the sample. At $T = 115$ K, the temperature stability was better than 50 mK. With our experimental setup we were not able to determine exactly the absolute value of the measured susceptibilities. We used the method suggested by Koziol [19] and normalized $\chi'_1$ to $-1$ at $T = 78$ K.

3. Numerical calculations

The harmonic susceptibilities are determined by the magnetic flux entering and leaving the sample. Therefore it is necessary to study the nonlinear diffusion equation that governs the spatial-temporal evolution of the local magnetic field. We consider an infinite-slab geometry with the sample to be located between the planes $x = -d$ and $x = d$, and the external field $H_E = H_{dc} + H_{ac}\sin(2\pi ft)$, applied parallel to the surface of the sample. Here $B_d$ is the applied dc bias magnetic field, $B_{ac}$ is the amplitude of ac field and $f$ is frequency of the ac field. In such a one-dimensional case the flux diffusion equation can be written as [14]

$$\frac{\partial B}{\partial t} = -\frac{\partial}{\partial x}(vB),$$

(1)

where $v \equiv v_0(j/j_c)\exp[-U(j)/T]$ is the mean velocity of vortices in the $x$ direction, $v_0 \equiv u\omega_m$, $u$ is the hopping distance, $\omega_m$ is the microscopic attempt frequency and the factor $j/j_c$ is introduced to provide a gradual crossover to the viscous flow regime $v \propto j$ at $T \gg U(j)$. We use the general form of the current dependence of the activation energy $U(j) = U_0/j^{\alpha}/((j/j_c)^{\alpha} - 1)$ given by the vortex glass/
collective creep models [20–23]. In such general case, the analytic solution of the nonlinear flux
cree ep equation constitutes intricate task even for
the linear Anderson–Kim dependence \( U(j) = U_0(1 - j/j_c) \). In the following, we present results of
numerical solutions of the creep equation. The
results are compared with our experimental data.

Due to the symmetry of the problem, only the
region \( x > 0 \) is considered. Then the boundary
conditions are \( B(x = d, t) = B_d + B_{ac} \sin(2\pi ft) \) and \( (\partial B/\partial x)(x = 0, t) = 0 \). The initial condition is
\( B(x, t = 0) = B_0 \). Note that the present calculation,
like those in the literature, is for a homogeneous
hard superconductor with \( H_{cl} = 0 \).

The numerical integration of Eq. (1) is carried
out by using a simple single step method as has
been discussed in detail by Qin and Yao [14]. In
order to obtain the ac susceptibility, the magnetization
\( M \) has to be calculated for the applied time
dependent field \( B(t) = B_d + B_{ac} \sin(2\pi ft) \). For
the geometry considered, the magnetization is given by
\[
\mu_0 M(t) = \frac{1}{d} \int_0^d B(x)dx - [B_d + B_{ac} \sin(2\pi ft)].
\]

The complex ac susceptibilities \( \chi' \) and \( \chi'' \) are
then calculated as
\[
\chi'_a = \frac{1}{\pi B_{ac}} \int_0^{2\pi} \mu_0 M(t) \sin(n \omega t) d(\omega t),
\]
\[
\chi''_a = \frac{1}{\pi B_{ac}} \int_0^{2\pi} \mu_0 M(t) \cos(n \omega t) d(\omega t),
\]
where \( \omega = 2\pi f \).

The critical current density \( j_c(B, T) \) and the
apparent activate energy \( U_0(B, T) \) as functions of
temperature and local magnetic field have to be
specified in order to account for the temperature
and field dependence of the ac susceptibilities. A
natural choice is to rely on pinning models
invoked in the literature for explaining experimental
data on irreversible magnetic properties. We choose the forms as [14]
\[
j_c(B, T) = j_c0\left[1 + (T/T_c)^2\right]^{-1/2}\left[1 - (T/T_c)^2\right]^{1/2} \frac{B_0}{B + B_0},
\]
\[
U_0(B, T) = U_{00}\left[1 - (T/T_c)^4\right] \frac{B}{B + B_0},
\]
where parameters \( j_c0, U_{00} \) and \( B_0 \), independent of \( T \)
and \( B \), serve as the scale of \( j_c, U_0 \) and \( B \), respectivly. Such temperature dependencies of \( j_c \) and \( U_0 \)
arise within the collective pinning model, where the
vortices are supposed to be pinned by a rando
mly distributed weak pinning centers, possibly
related to local variations of the electronic mean
free path [17]. For all calculations presented in this
work, the values of the parameters used in Eq. (1)
are \( \omega_\text{om} = 1 \text{ m/s}, U_{00}/k_B T_c = 10 \) and \( \mu = 0.6 \). A
more detailed description of the parameters has
been given in Ref. [16].

4. Results and discussion

In this section we compare measured even
harmonics of susceptibility with model calculations. Fig. 1(a) shows the calculated even
harmonics as a function of temperature at \( B_d = 20 \)
Gs, \( B_{ac} = 10 \text{ Gs} \) and \( f = 1111 \text{ Hz} \). Compared with
the real part \( \chi'_i \) and imaginary part \( \chi''_i \) of fundamental susceptibilities that are constrained to
negative and positive values, respectively, the
higher harmonics are more complicated. As
mentioned above, because of the symmetry of the
hysteresis loops Bean’s model does not predict
even harmonics. An asymmetry is introduced into
the hysteresis loops when a bias dc magnetic field
is present in extended critical state models, like
the Kim–Anderson model, which takes into account
the field dependence of the critical current density.
Then even harmonics are generated. It is expected
that the asymmetry is most prominent when dc
magnetic field is the same order as ac magnetic
field [4], which is the case in our calculations. In
Fig. 1(b) we plotted measured even susceptibilities
of Bi2223 versus temperature for an ac field
\( B_{ac} = 10 \text{ Gs} \) at \( f = 1111 \text{ Hz} \). There was a super
imposed dc magnetic field \( B_d = 20 \text{ Gs} \). It can be
seen that the experimental results are quite similar
to model predictions shown in Fig. 1(a). Quantitative agreements may be achieved by choosing
proper values of the parameters used in the model
calculations. In the present work the theoretical
and experimental results were not compared to
each other quantitatively since we could not obtain
the absolute values of the susceptibilities with our experimental setup.

In experiments we found the signs of all the even harmonics $\chi_n'$ and $\chi_n''$ $(n = 2, 4, 6)$ depended on the polarity of the superimposed dc field. The curves shown in Fig. 1(b) changed their sign for a negative dc bias field as observed by Ishida and Goldfarb [4]. Such dependence can be derived easily from Eqs. (2) and (3).

In order to show the effect of dc magnetic fields $B_d$, the ac field amplitude $B_{ac}$ and frequency $f$ on the harmonics clearly, in the following discussion we use the module of the harmonics $|\chi_n| = (|\chi_n'|^2 + |\chi_n''|^2)^{1/2}$ as a function of temperature at different $B_d$, $B_{ac}$ and $f$. In this work we only give results of the second harmonic since it is the strongest and the most easily measured of all the even harmonics. Fig. 2(a) shows the absolute value of the second harmonic susceptibility versus temperature for ac field $B_{ac} = 6$ Gs at the indicated dc magnetic fields. A pronounced peak could be observed in $|\chi_2|$ curve. As the dc magnetic field increased, the peak shifted to lower temperatures and became broader and broader while its height decreased monotonically. When $B_d$ was larger than 1000 Gs, the peak height of even harmonic approached zero, which implies that the asymmetry induced by dc fields tends to disappear. Fig. 2(b) shows the experimental data. We can see that with increasing dc fields the change of the peak position and peak height followed model calculations. At low temperatures, $|\chi_2|$ was larger for higher dc field, which
is not consistent with the theoretical results shown in Fig. 2(a). The deviation may be due to the inhomogeneity of our sample whereas in the model calculations we assumed the sample was homogeneous.

Shown in Fig. 3(a) is the temperature dependence of $|\chi_2|$ at various ac field amplitudes for $B_d = 12$ Gs, $f = 1111$ Hz. In contrary to the dc magnetic field, as the amplitude of the ac field increased the height of the peak increased gradually while it shifted to lower temperatures. The experimental curves were shown in Fig. 3(b), which are consistent with theoretical predictions. From Figs. 2 and 3, we see that the magnetic fields had strong effects on even harmonic susceptibilities. When the dc magnetic field $B_d$ gradually approached the amplitude $B_{ac}$ of the ac field, the peak height of the second harmonic always increased.

The calculated effect of the ac field frequency $f$ on the second harmonic was plotted in Fig. 4(a).
As the frequency decreased, the peak height of the second harmonic decreased while the peak shifted to lower temperatures. The experimental results were shown in Fig. 4(b). It has been found [16] that the fundamental susceptibility moves to the limit of the Bean critical state model when the frequency decreases. This means that the even harmonics will disappear gradually as the frequency approaches zero. From Fig. 4(b) we can see that our experimental data just followed theoretical curves even though the frequency changed in a relative narrow range. It is interesting to note that the peak height of the third harmonic increases as the frequency decreases [24], which is opposite to the behavior of the second harmonic. Bean’s model and its generalization cannot explain all these frequency-dependent features.

5. Conclusions

We have investigated the effects of dc magnetic fields, the ac field amplitude and frequency on even harmonics of susceptibility. The experimental results of (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_y$ bulk superconductors have been compared with theoretical susceptibility curves based on the numerical calculation of the flux diffusion equation. The theoretical curves are in good agreement with the temperature-, field- and frequency-dependent features of the experimental data. The frequency independent critical state model breaks down in explaining all these results even though its generalization can generate even harmonics of susceptibility.

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