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Non-linear AC magnetic susceptibility of the high-T_c superconductor $NdBa_2Cu_3O_7$

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Abstract. The non-linear AC magnetic susceptibility of *polycrystalline* $NdBa_2Cu_3O_7$ superconductor has been studied to examine the intergrain contribution to magnetic properties at low field. A superconducting sample is subjected to a DC magnetic field and a small AC magnetic field in parallel. The complicated dependence of the magnetic susceptibility on the applied field is attributed to the critical state of trapped magnetic flux in the intergrains. The non-linear magnetic susceptibility of higher harmonics with frequency up to $4f$ was measured as a function of varying applied fields. Numerical results of calculations using a modified critical state model are in good agreement with the measured AC susceptibility data. We find the relation $J_c \sim H^{-n}$ ($n = 2.5$) for the dependence of the intergrain critical current density J_c on the applied magnetic field H in conformity with the previous results of $YBa_2Cu_3O_7$ where a smaller value of $n = 1.8$–2.2 was obtained.

1. Introduction

High-T_c superconductors, mostly granular superconductors, consist of grains and weak links connecting the grains. Below the superconducting transition temperature, the grains are each superconducting, the weak links are of Josephson-junction type (Ekin *et al* 1988, Dimos *et al* 1990). The grains and the weak links are also referred to as intragrains and intergrains respectively.

Recent studies on critical current (Ekin *et al* 1988, Evetts and Glowacki 1988, Küpfer *et al* 1988, Peterson and Ekin 1988, 1989, Male *et al* 1989) and magnetic susceptibility (Müller *et al* 1988, 1989, Müller and Pauza 1989, Müller 1989, Ishida and Goldfarb 1990, Lam *et al* 1990, Kim *et al* 1991) of high-T_c superconductors under applied magnetic fields have been successful in separating out the intergrain (weak link) from the intragrain (superconducting grain) characteristics. At low fields the intergrain contribution predominates and at higher fields the intragrain characteristics dominate. The intergrain character in the critical-current measurements is represented by a rapid decrease of the critical current density J_c by several orders of magnitude with increasing magnetic field H. After the rapid decrease it remains almost field independent, which is considered an indication of the intragrain character.

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Magnetic AC susceptibility measurement in the low-field region has been reported for the complicated dependence on applied fields and it was ascribed to the intergrains. An attempt was made to explain the measurement by applying a modified critical state model to $YBa_2Cu_3O_7$ (Lam *et al* 1990, Kim *et al* 1991).

In this paper we want to report our experimental investigation of the field dependence of higher-order AC magnetic susceptibility $\chi_n = \chi'_n - i\chi''_n$ of the polycrystalline high-T_c superconductor $NdBa_2Cu_3O_7$ to find the general roles of the intergrain in the complicated non-linear structure of magnetic responses. We present the susceptibility data up to the fourth harmonic ($n = 4$) response in $NdBa_2Cu_3O_7$ and compare it with the results of theoretical calculations using the modified critical state model.

2. Experimental details

The granular superconductor $NdBa_2Cu_3O_7$ was prepared by the solid-state reaction method. Mixed powders of Nd_2O_3, $BaCO_3$, CuO were dry-milled and pressed to make a pellet. The pellet was calcinated at 910 °C for 12 h and sintered at 950 °C for 12 h. After sintering, the pellet was annealed at 500 °C for 4 h and slowly cooled to room temperature in a furnace. All processes were done under an oxygen atmosphere. We cut the pellet to get a sample of dimension $1.6 \times 3 \times 7\,\mathrm{mm}^3$. Measurements of resistivity and AC magnetic susceptibility showed that T_c of the sample was about 90 K.

The measuring system we used consists of a Hartshorn bridge, current sources for the magnetic fields (HP 8116A function generator and a voltage-controlled current source) and a signal detecting system (PAR 113 preamplifier, SR 530 lock-in amplifier and a frequency multiplier circuit). The Hartshorn bridge consists of two kinds of coils. The primary coil is a cylindrical solenoid of length 60 mm and diameter 15 mm. The coil, driven by the current source which is controlled by an applied voltage $V = V_{DC} + V_{AC}\cos(2\pi f t)$ of a function generator, generates the magnetic field $H_a = H_0 + H_1 \cos(2\pi f t)$. Inside the primary coil are a pair of coaxial secondary coils. One of the secondary coils encloses the sample and is connected in series with the identical secondary coil which is counterwound so that the signal from the secondary coils is almost zero when the sample is absent or non-magnetic. The sample within one of the secondary coils produces a voltage proportional to the time derivative of the magnetization of the sample. The induced voltage is amplified by a preamplifier and analysed by using a two-phase lock-in amplifier.

Higher-order susceptibility can be measured by applying a phase-stabilized reference signal of frequency nf to the lock-in amplifier where n is an integer and f is the driving frequency of the AC magnetic field. We used a frequency multiplier circuit for the reference signal and checked for the frequency by using a frequency counter. We also corrected the phase by checking the second-harmonic signal using two different methods: one using a $2f$ mode of the lock-in amplifier with reference frequency f, the other using an f mode of the lock-in amplifier with frequency $2f$ as reference. The sample and the Hartshorn bridge were cooled to liquid-nitrogen temperature during data acquisition. Experimental data were taken by slowly increasing the sweep magnetic field H_0 from 0 to approximately 14 Oe, and then decreasing H_0 to 0 and so on. During the experiments we adjusted the phase of the lock-in amplifier so that the imaginary part of the magnetic susceptibility at the fundamental frequency f would remain zero at very small AC field ($H_1 < 50$ mOe) and the phase was fixed for the measurement of higher-order susceptibility. The frequency of the AC field we used was 394 Hz.

3. Results and discussion

The measurements were made on a $NdBa_2Cu_3O_7$ sample of rectangular shape. The applied field H_a was parallel to the long axis of the sample. The magnitude of the applied AC field H_1 was varied from 0.6 Oe to 7.8 Oe.

Figure 1 shows the observed data ((*a*) and (*c*)) and the calculations ((*b*) and (*d*)) of AC susceptibility at the fundamental frequency f. The real or dispersive component of the first-order AC susceptibility $\chi_1'(H)$ is $-1/4\pi$ near $H_0=0$ for small H_1 but increases rapidly to zero as H_1 or H_0 increases. This is a typical characteristic of the intergrains or the weak links (Peterson and Ekin 1989, Lam *et al* 1990, Kim *et al* 1991). It suggests that coupling of the intergrains to the intragrains is easily broken by penetration of magnetic vortices. The vortex penetration also accounts for a peak of the imaginary (or 'loss') component χ_1'' at a certain value of H_0 for small H_1 as shown in figure 1(*c*). Since χ_1' corresponds to magnetic shielding while χ_1'' corresponds to AC loss, the minimum χ_1' (near to $-\frac{1}{4}\pi$) means the strongest diamagnetic state of least flux penetration and the maximum χ_1' (near to 0) corresponds to the normal state of largest flux penetration. The maximum in χ_1'' corresponds to the maximum phase difference between the rate of change in the total penetrated flux with respect to time and the AC driving field.

For $H_a \geqslant 1$ Oe it is energetically favorable for vortices to enter intergrain and the Lorentz force on the penetrated magnetic vortices in the intergrain region is in equilibrium with the pinning force due to a complex array of the weak links of Josephson junction type. Therefore the vortices are in a critical state as proposed by Bean (1964) for type II superconductors. The critical state in the applied field H_a is characterized by the local field $H(r)$ and a boundary condition $H(R) = H_a$ where r is the distance from the centre of the cylindrical sample and R is the radius of cylinder.

The critical-state equation for $H(r)$ is given as follows:

$$\mathrm{d}H(r)/\mathrm{d}r = \pm(4\pi/c)\, J_c(H) \tag{1}$$

where c is the speed of light, J_c is the modulus of the critical current (see equation (4) below), and (±) is determined by the sense of vortex motion. Equation (1) with a suitable assumption for $J_c(H)$ is referred to as a critical-state model (Kim *et al* 1963, Anderson and Kim 1964, Bean 1964). The harmonic-order AC magnetic susceptibility $\chi_n = \chi_n' - \mathrm{i}\chi_n''$ is defined as follows (Ishida *et al* 1990)

$$4\pi\chi_n' \propto (2f/H_1) \int_0^{1/f} B(t)\cos(2\pi n f t)\,\mathrm{d}t \tag{2}$$

$$4\pi\chi_n'' \propto (2f/H_1) \int_0^{1/f} B(t)\sin(2\pi n f t)\,\mathrm{d}t \tag{3}$$

where $B(t)$ is a spatial average of the local field $H(r,t)$ for a time-dependent applied field $H_a(t)$.

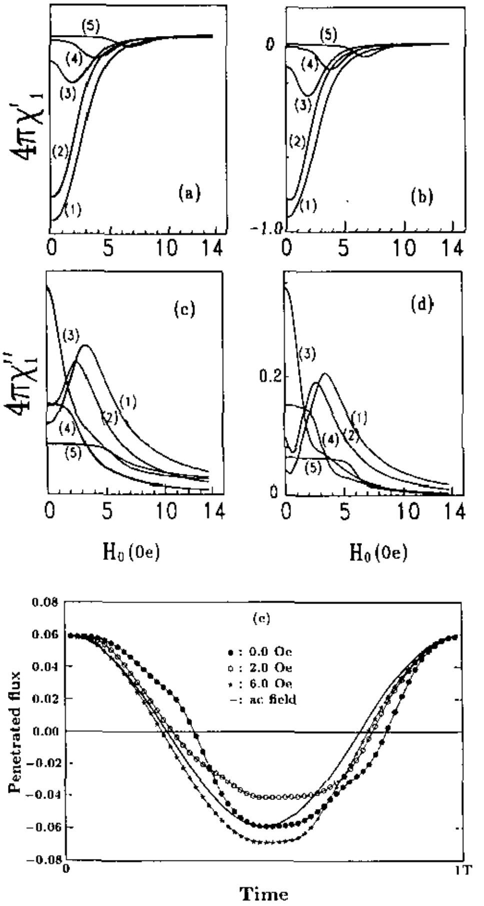

Figure 1. (*a*) and (*c*) Measured real and imaginary component χ' and χ'' respectively of the complex AC magnetic susceptibility χ_1 at fundamental frequency $f = 394$ Hz for a $NdBa_2Cu_3O_7$ sample at $T = 77$ K for various values of AC magnetic field H_1; (*b*) and (*d*) Theoretical curves calculated by the modified critical-state model; (*e*) Penetrated flux versus time during one cycle period of $H_a = H_0 + H_1 \cos(2\pi f t)$ as calculated from the modified critical state model for $H_1 = 2.9$ Oe and $H_0 = 0.0$ (•), 2.0 (o), 6.0 Oe (⋆). The parameters used for the calculations are $\beta = 2.5$, $H_s = 3.0$ Oe, and α' of equation (4). The numbers on the curves represent the different values of H_1: (1) 0.6 Oe, (2) 1.0 Oe, (3) 2.9 Oe, (4) 4.9 Oe, (5) 7.8 Oe.

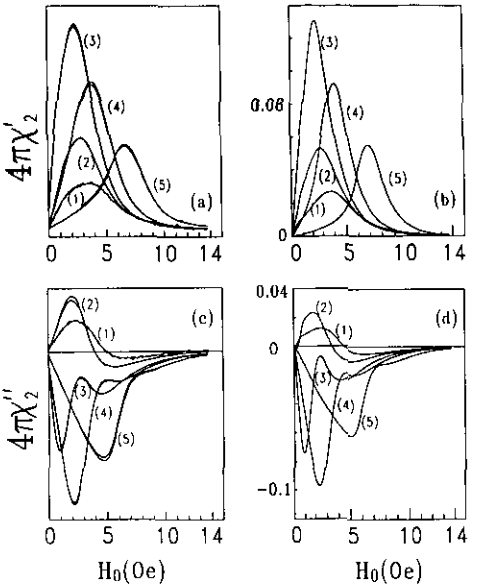

Figure 2. (*a*) and (*c*) Measured complex AC susceptibility at the second-harmonic frequency χ_2; (*b*) and (*d*) Calculations using the modified critical-state model. Conditions for the measurement and parameters for the calculation are the same as in figure 1.

The modified critical-state model applied to the superconductor $YBa_2Cu_3O_7$ has employed the field dependence of $J_c(H)$ for the intergrain as (Lam *et al* 1990, Kim *et al* 1991)

$$J_c = c\alpha'/(|H| + H_s)^{\beta} \tag{4}$$

with $\beta \simeq 2$, which means steeper field dependence than $\beta = 0$ (Bean 1964) or $\beta = 1$ (Anderson and Kim 1964) as used for conventional type II superconductors.

The parameter α' is related to the vortex pinning force density $\alpha(H)$ of the form $\alpha'/(|H| + H_s)^{\beta-1}$ where H_s is introduced to avoid the singularity at $H = 0$.

We apply the modified critical-state model in the form of equation (4) to fit the experimental data of our present work as shown in figures 1–4.

For the $NdBa_2Cu_3O_7$ sample the best fitting parameters are obtained as $\beta = 2.5$, $R = 0.8$ mm and $H_s = 3.0$ Oe. We can obtain from the same model a relationship

$$\alpha' = [(H_s + H^*)^{\beta+1} - H_s^{\beta+1}]/[4\pi(\beta + 1)R] \tag{5}$$

where H^* is the value of the applied field at which we have a maximum of $\chi_1''(H_1)$ at $H_1 = H^*$. This value of $H_1 = H^*$ can be measured experimentally and corresponds to that for which the flux front reaches the centre $r = 0$. We find $H^* = 2.4$ Oe for the $NdBa_2Cu_3O_7$ sample. Compared with $H^* = 8$–10 Oe for $YBa_2Cu_3O_7$ a much smaller value of H^* is obtained for $NdBa_2Cu_3O_7$. This parameter gives $\alpha' \simeq 90$ and thus $\alpha(H = 0) \simeq 17\,\mathrm{Oe}^2\,\mathrm{cm}^{-1}$ which is also much smaller than $\alpha(H = 0) \simeq 174\,\mathrm{Oe}^2\,\mathrm{cm}^{-1}$ for $YBa_2Cu_3O_7$ (Kim *et al* 1991). We find that the parameter β controls the overall shape of the susceptibility curve whereas the parameter H_s affects mostly the region near $H_0 = 0$. This is as expected because H_s is introduced to keep $J_c(H)$ finite at $H = 0$.

For a magnetic field $H_a = H_0 + H_1\cos(2\pi f t)$ applied to the superconducting sample, the AC flux penetration during one cycle period was calculated from the modified critical-state model as shown in figure 1(*e*), where the parameters are chosen as H_1=2.9 Oe, H^*=2.4 Oe, H_s=3.0 Oe, β=2.5 in conformity with $NdBa_2Cu_3O_7$ so that it may be directly compared to the curves of figure 1(*b*) and (*d*). The minimum of χ_1' at H_0=2.0 Oe in figure 1(*b*) (curve (3)) corresponds to the minimum penetrated flux per cycle period, that is, the minimum area span of the curve in figure 1(*e*). For $H_0 > 5.0$ Oe the point where the flux tends toward maximum saturation corresponds to the maximum χ_1' of the susceptibility curves. The maximum of χ_1'' at H_0=0.0 Oe corresponds to the maximum phase difference between the flux change and the AC driving field. We can also see that the increasing DC field results in the flux change being in phase with the AC driving field so that χ_1'' tends to decrease. For AC fields greater than the penetration field H^* the flux penetration is completed up to the centre of the sample without a DC field as can be seen in curve (3) of figure 1. For AC fields smaller than H^* (see curve (1) of figure 1) the flux front reaches the centre of the sample only when the DC field is increased to H_0=3.8 Oe, and χ_1'' obtains its maximum value. With further flux penetration χ_1' will also increase. However, as the DC field is further increased the superconducting diamagnetism deteriorates and the flux change in time becomes in phase with the AC driving field resulting in a decrease of χ_1''. Even without the DC field χ_1'' is observed to increase with increasing AC amplitude H_1 until H_1 reaches H^* but to decrease as H_1 is increased to above H^*. In order to compare the measured data with the model calculations we used an arbitrary unit in which the range of $4\pi\chi_1'$ was set from -1 (fully diamagnetic) to zero (fully penetrated).

The calculations of figure 1(*b*) and 1(*d*) show remarkably good agreement with the measured data for a wide range of H_1 and H_0. Figure 2 shows the data and the calculations of the second-harmonic susceptibility χ_2. Good agreement between the data and the calculations, using the same values of fitting parameters as in figure 1, is obtained. This indicates that the field dependence of the form $J_c(H) \sim H^{-n}$ (n = 1.8–2.5) for the intergrains is valid for most of the granular high-T_c superconductors and is compatible with the field dependence for a random array of Josephson junctions (Peterson and Ekin 1989).

Close examination of the theoretical curves (1) and (2) in figure 1(*c*) and figure 1(*d*) for $H_0 < 3$ Oe shows some discrepancy between the data and the calculations. Similar discrepancy is noticed in figure 3 for the third-order susceptibility χ_3 and also in figure 4 for the fourth-order susceptibility χ_4. Although part of the discrepancy may arise from the experimental difficulty in adjusting the correct phase of the lock-in amplifier we speculate that it may be due to inappropriateness of the intergrain field dependence $J_c(H)$ in equation (4) at very small field. Equation (4) gives larger values of J_c at small field than observed.

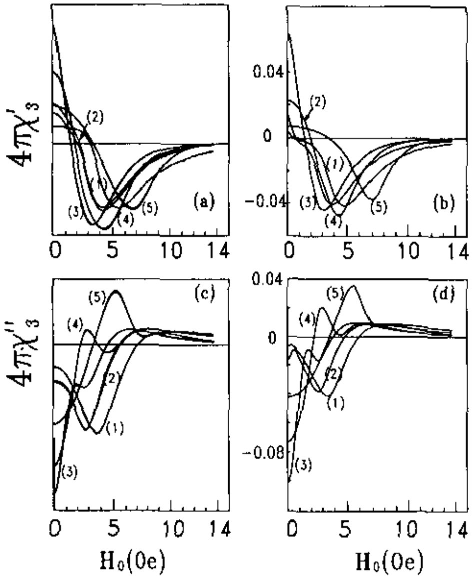

Figure 3. (*a*) and (*c*) Measured complex AC susceptibility at the third harmonic χ_3; (*b*) and (*d*) – Calculations using the modified critical-state model. Conditions for the measurement and parameters for the calculation are the same as in figure 1.

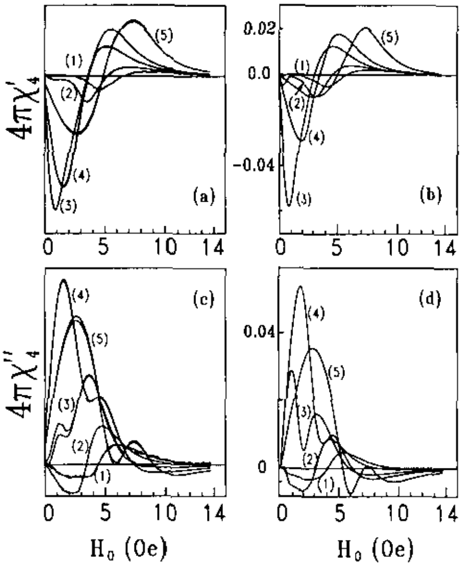

Figure 4. (*a*) and (*c*) Measured complex AC susceptibility at the fourth harmonic χ_4; (*b*) and (*d*) Calculations using the modified critical-state model. Conditions for the measurement and parameters for the calculation are the same as in figure 1.

Since we do not apply the transport flow of current but only the shielding current in the sample we do not have the problem of the self-field effects. The current-induced self-field effects as encountered in the transport measurement of the critical current density give rise to a decrease (or increase) of J_c with increasing (or decreasing) thickness of the sample, and the local value of J_c is masked by self-field effects (Campbell and Blunt 1990).

Reduction of $J_c(H)$ in the sample at small field is expected to be due to defects or impurities in the intergrains so that $J_c(H)$ is different from that of ideal Josephson junctions. Study of further details of the susceptibility at small field values will be necessary to understand their effects on the intergrain J_c. We also observed the hysteresis in the AC susceptibility for the $NdBa_2Cu_3O_7$ sample. The hysteresis is found to be more evident in the higher-order susceptibility as can be seen in figure 4. The origin of the hysteresis may be ascribed to the irreversibility of the critical current associated with the trapped magnetic vortex and microstructures in high-T_c superconductors (Evetts and Glowacki 1988, Jeffries *et al* 1989, Kim *et al* 1991). More experimental work should be done for a quantitative understanding of the observed hysteresis.

The critical current behaviour of polycrystalline high-temperature superconductors varies with the grain size (Sennoussi *et al* 1991). In general the high-T_c bulk superconductors have a broad grain-size distribution dependent on the sample preparation process. However, experimental results for various $YBa_2Cu_3O_7$ samples of different origin report almost the same value of $\beta \simeq 2$. We have also obtained $\beta \simeq 2.2$ for $YBa_2Cu_3O_7$ prepared by the same process applied to the $NdBa_2Cu_3O_7$ and $\beta \simeq 1.0$ for BiSrCaCuO in fair agreement with $\beta \simeq 1.3$ of Müller *et al* (1992). These experimental observations seem to indicate the important contributions of the different intergrain characteristics intrinsic to each of the different high-T_c superconductors.

4. Summary and conclusion

Higher-order AC magnetic susceptibility of the polycrystalline $NdBa_2Cu_3O_7$ sample was investigated as a function of the applied magnetic field to study the non-linear magnetic properties of the intergrains. We used a two-coil technique for measurements of the higher-order AC susceptibility with respect to frequency from fundamental f to the fourth harmonic $4f$. Although the experimental data showed a very complicated field dependence, the theoretical calculations on the basis of a modified critical-state model, with an assumption of field dependence of the intergrain current density J_c as $J_c(H) \sim H^{-2.5}$, could reproduce the experimental data. Although quantitative details differ between $NdBa_2Cu_3O_7$ and $YBa_2Cu_3O_7$ in both the critical current density $J_c(H)$ and the penetration field strength H^* the same form of the modified critical-state equation applies equally well for both of them. It is speculated that this may be common to high-T_c superconductors where the intergrains are basically of Josephson-junction type but with random orientations and varying strengths of the junctions.

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