Geometry effect on the nonlinear magnetic response of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

PARSHANT BATHAM$^1$, S B ROY$^2$, SHAILENDRA KUMAR$^2$, A K PRADHAN$^2$, A K GROVER$^1$, P CHADDAH$^2$, RAM PRASAD$^3$ and N C SONI$^3$

$^1$Centre of Advanced Study, Department of Physics, Panjab University, Chandigarh 160014, India
$^2$Low Temperature Physics Group, Centre of Advanced Technology, Indore 452013, India
$^3$Metallurgy Division, Bhabha Atomic Research Centre, Bombay 400085, India

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Abstract. Results are presented for study of nonlinear magnetization of a sintered $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ pellet of different thickness of a sample subjected to very low magnetic field. On cooling the sample below $T_c$ in zero field a change, in the oscillatory structure of harmonics in increasing dc field is observed in very low ac magnetic field. The effect of finiteness of the sample on the oscillatory structure and on the hysteresis of harmonics is also studied. The results are explained qualitatively.

Keywords. Nonlinear magnetization; critical current density; demagnetization factor.

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1. Introduction

Nonlinear magnetization response of hard type II superconductors (including the high temperature superconductors) is a well studied phenomenon [1–7]. According to Bean's Critical State Model [1] the origin of the harmonics is hysteresis in magnetization and the nonlinear relation between magnetization and applied field due to pinning of the flux lines in hard type II superconductors. Bean's model assumes that a hard type II superconductors can carry a limiting macroscopic superconducting current density $J = J_c$ and further, that any electromotive force, however small, will induce the macroscopic superconducting current of density $J_c$ to flow locally. Given the distribution of current that are set up and the applied field, one can calculate the flux density $B(r)$ at various point in the sample. The average magnetization induced by an applied field can be calculated by averaging the local flux density. If we apply an ac magnetic field [$H(t) = H_{ac}\cos\omega t$], we have $M(\omega t) = -M(\omega t + \pi)$ i.e. magnetization which has time reversal symmetry. This is shown schematically in figure 1a and one observes only the odd harmonics in accordance with Bean's critical state model [1]. In the limit that critical current density is independent of applied field, this time reversal symmetry is maintained even when a static magnetic field is superimposed as depicted in figure 1b. The field dependence of critical current density leads to generation of even harmonics, when we superimpose the static magnetic field i.e. when $H(t) = H_{ac} + H_{dc}\cos\omega t$. It also breaks the time reversal symmetry of the magnetization loop as indicated in figures 1c–1e [8]. Ji et al [2] and Navarro et al [3] reported observation of the even harmonics when the dc field was superimposed on time varying magnetic field in high temperature superconductors. The amplitude of harmonics was calculated as a function of external
dc field and it was found that [4] magnitude of nth harmonics should show oscillatory structure with \((n - 1)\) minima when the superimposed \(H_{\text{app}}\) field varies between \(+ H_{\text{ac}}\) and \(- H_{\text{ac}}\). The history effect of the static magnetic field [6, 7] on the magnitude of harmonics in the case of ceramic samples of high temperature superconductors is a much more complicated phenomenon. The history effect is explained using the two component behaviour of ceramics ascribed to grains and intergrain boundaries. Further, the observation of a sharp structure of second harmonic in remnant state [9] can be explained in terms of flux trapping in grain and using this memory effect of magnetization in second harmonics, measurements can be made to estimate the field for first flux penetration into grains, viz. \(H_{\text{cl}}\).

In the present investigation of harmonics generation, we have focused attention onto an aspect no specially studied in any detail so far, namely the effect of the sample shape (i.e. demagnetization factor) on the generation of harmonics in a given ceramic preparation of a compound. Magnetization studies in ceramic sample are routinely used to elicit informations of field dependence of intergrain and intragrain critical current density. Our present studies demonstrate that for the quantitative information on the current densities, the effect of demagnetization factor is necessary. We report the 2nd and 3rd harmonic as a function of static magnetic field in zero field cooled state of \(\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}\) sintered pellet sample with different demagnetization factors with very low ac driving field at a temperature of 77 K. These samples with different demagnetization factors were obtained by progressive thinning of the same pellet.

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2. Experimental

The results reported here are on a sintered pellet of YBa$_2$Cu$_3$O$_{7-\gamma}$ which were prepared by standard solid state sintering process. Y$_2$O$_3$, BaCO$_3$ and CuO in appropriate proportion were calcinated in air at 900 °C for 15 hr, interspersed with grinding and mixing. The powder was compacted (at 10 tons/sq in) and sintered in oxygen at 940 °C for 15 h and was held at 400 °C during cooling. No second phase was detected in X-ray diffraction and oxygen was estimated by iodometry as 6.95. The sample has a resistive $T_c$ of 90 K and this was confirmed by magnetization data as well. For measuring the ac magnetization a low field hysteresis loop tracer [10] with driving frequency of 317 Hz and driving field $H_{ac}$ 1 to 2 Gauss was used. The field is applied perpendicular to the plane of the pellet to study the effect of the demagnetization factor of the pellet. A dc biasing field upto 45 G can be provided by a split pair of Helmholtz coils, coaxial to the coil of hysteresis loop tracer.

An AD3522 FET analyzer was used to measure the amplitude of harmonics in the AC magnetization. The possibility of spurious contribution at secondary coil (in absence of sample) from the driving frequency was checked. No contribution from higher harmonics signal was observed in the absence of the sample, or with the sample at $T > T_c$. The noise level was about 15 mV. For calculating the demagnetization factor we measure the ac magnetization at a low ac field value (1 G) such that no hysteresis was observable. With two different orientation that is field parallel to plane of pellet (of volume V) and field perpendicular to the plane of the pellet, we obtain values $M_1$ and $M_\perp$ respectively [11].

$$\frac{M_1}{V} = -\frac{H}{(1 - N_\parallel)}; \quad \frac{M_\perp}{V} = -\frac{H}{(1 - N_\perp)} \quad (1)$$

We also note that the trace of demagnetization factor is one i.e.

$$N_\parallel + 2N_\perp = 1 \quad (2)$$

where $N_\parallel$ and $N_\perp$ are the demagnetization factors when applied field is perpendicular and parallel to the plane of the pellet. Using (1) and (2) and the observed $M_1$ and $M_\perp$, we find that demagnetization factors are $N_1 = 0.745$, $N_2 = 0.792$ and $N_3 = 0.842$ for three different pellets of thickness are $t_1 = 1.61$ mm, $t_2 = 1.41$ mm, and $t_3 = 0.92$ mm, respectively, where the magnetic field is perpendicular to the plane of sample. Each of these sample had a diameter of 8.08 mm. We are interested in generation of harmonics in zero field cooled (ZFC) state. The sample is cooled to 77 K in absence of any dc bias ($H_{dc}$) and then the $H_{dc}$ is switched on. Measurement are made for various $H_{dc}$ field in small value of constant $H_{ac}$ field.

3. Results and discussion

3.1. Structural change in oscillatory behaviour of harmonics

In figure 2, we present the magnitude of third harmonic signal (3f) as a function of $H_{dc}$ with $H_{ac}$ fixed at 1, 1.5, 2 and 3 Gauss for sintered pellet YBa$_2$Cu$_3$O$_{7-\gamma}$ with demagnetization factors $N_1 = 0.74$. Figures 3 and 4 show the same data for sample with demagnetization factors $N_2 = 0.792$ and $N_3 = 0.842$ respectively. In figure 2, the magnitude of third harmonic shows a minimum as $H_{dc}$ varies from zero to maximum
value (5 G) at driving field $H_{ac} = 3 \text{ G}$. But, at driving fields $H_{ac} = 1.5 \text{ G}$ and 1 G, the $3f$ signal increases monotonically on varying the dc bias from 0 to maximum value 5 G which is quite unusual. The observation of one minimum in $3f$ signal for $H = 3 \text{ G}$ is consistent with theory given by Ji et al [2] in which they have calculated that the magnitude of $n$th harmonic should show oscillatory behaviour with $(n - 1)$ minima on varying the $H_{dc}$ from $-H_{ac}$ to $H_{ac}$. This discussion is, however, valid only for $H_{ac} > H^*$, where $H^*$ is the field for full penetration of the sample. In our observation of the M–H loops on this sample, the field for full penetration is about 2.0 G. The change in the oscillatory behaviour for demagnetization factor $N_1 = 0.74$ shown in figure 2 appears to be at driving field at $H_{ac} = 2 \text{ G} = H^*$. For driving field $H_{ac} < H^*$, we observe a behaviour different from that predicted in ref. [2] in that $3f$ signal has only one minimum and that is at $H_{dc} = 0$. This is also seen from the calculation of ref. [4] when $H_{ac} < H^*$. The change in the oscillatory structure in $3f$ signal for demagnetization factors $N_2 = 0.79$ and $N_3 = 0.84$ is measured to be at field values $H = 1.5 \text{ G}$ and 1 G, as shown in figures 3 and 4 respectively. These results are new and call for further refinement in the existing theoretical models.

3.2 Effect of geometry on the harmonics

From figures 2 to 4, the change in oscillatory behaviour of $3f$ vs $H_{dc}$ shifts to lower field values on increasing the demagnetization factor. This confirms the fact that on
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![Graph showing the relationship between f_3 (microvolt) and H_{dc} (G)]

**Figure 3.** Third harmonic amplitude (3f) as a function of dc magnetic field, the ac field being fixed at 1, 1.5, 2 and 3 G for demagnetization factor $N_2 = 0.792$.

![Graph showing the relationship between f_3 (microvolt) and H_{dc} (G)]

**Figure 4.** Third harmonic amplitude (3f) as a function of dc magnetic field, the ac field being fixed at 1, 1.5, and 2 G for demagnetization factor $N_3 = 0.89$.
Figure 5. The hysteresis in the amplitude of second harmonic (2f) of a ZFC pellet as function of $H_{dc}$ raised to 39 G and lowered to zero value in $H_{ac} = 4 G$ for demagnetization factor $N_1 = 0.745$.

Figure 6. The hysteresis in the amplitude of second harmonic (2f) of a ZFC pellet as function of $H_{dc}$ raised to 39 G and lowered to zero value in $H_{ac} = 4 G$ for demagnetization factor $N_2 = 0.792$. 

Figure 7. Unique functional dependence of amplitude of second harmonic (2f) as a function of scaled field ($H_{\text{ac}}$). (■) curve denote scaling of applied field for demagnetization factor $N_1 = 0.745$ and (●) curve signifies the scaling of field for demagnetization factor $N_2 = 0.792$ for the unidirectional increase of the applied field.

Increasing the demagnetization factor, the effective field experienced by the sample increases. The generation of even harmonics is the consequences of the field dependence of the critical current density [2–6]. The irreversible behaviour of the nonlinear response of the YBa$_2$Cu$_3$O$_{7-y}$ is attributed to the two component network of sample i.e. integrain and intragrain region. In figures 5 and 6, we observe that 2f signal shows a history effect with the applied magnetic field due to its ceramic nature [5, 6] for demagnetization factors $N_2 = 0.74$ and $N_3 = 0.79$. We have used a large ac field $H_{\text{ac}} = 4$ G and we observe a significant difference in the amplitude and in the field dependence of 2f for different demagnetization factors.

However, scaling the applied field with the demagnetization factor $H_{\text{scal}} = H/\{1 - N(1 - V_f)\}$ [12], where $N$ is demagnetization factor and $V_f$ is effective volume of sample into which flux is able to penetrate. This scaling becomes useful once the flux starts penetrating the intergrain region and our eq. (1) no longer holds. We observe in figure 7 2f signal show a unique functional dependence on scaled field for unidirectional increase in applied field and in figure 8 for unidirectional decrease in applied field, where following ref. [12], $V_f$ is taken to be 0.4. Therefore, for quantitative measurement of critical current density from magnetization studies, demagnetization correction is very important. Our results show that when this is taken into account, a unique dependence on applied field is obtained.
4. Conclusion

In conclusion, we observed that the $3f$ shows consistent behaviour as theories predict at ac field $H_{ac} > H^*$. However at field smaller than $H^*$, $3f$ increases monotonically. The ac field at which the shift in structure observed also decreases on thinning the sample progressively i.e. on increasing the demagnetization factor. We measured the significant difference in the second harmonics at different demagnetization factor of the sample. But, scaling the applied field with demagnetization factor and the total volume experiencing the field, the $2f$ shows a unique field dependence.

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References

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