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Field-cooled measurements of harmonic generation in magnetization of high-T_c superconductors

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Abstract. We present measurements of harmonic generation in the magnetization of sintered pellets of $YBa_2Cu_3O_7$ and $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$ as a function of DC field. The DC field is applied in the field-cooled mode. Measurements are made at 77 K for various values of the AC field amplitude. A comparison is made with calculations done within the critical state model.

Keywords. Harmonic generation; magnetization; superconductors; critical state model; field-cooled mode.

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1. Introduction

The magnetic response of a sintered pellet of a high-temperature superconductor (HTSC), to a time-varying magnetic field $B(t) = B_{dc} + B_{ac}\cos\omega t$, has been studied in detail in the last few years [1–6]. When B_{dc} is zero, higher odd harmonics $((2l+1)\omega)$ are observed in the magnetisation with their magnitude increasing as B_{ac} is increased. The absence of even harmonics indicates the symmetry of the hysteresis loop. When B_{dc} is non-zero a response is also observed at higher even harmonics. This observation, and the consequent asymmetry of the minor hysteresis loop, was stressed [1] to be a signature of a field-dependent critical current density $J_c(B)$. The dependence of the harmonic magnitudes (M_n) on B_{dc} is again understood [7] as a signature of a field-dependent $J_c(B)$. Calculations of the dependence of various harmonics on B_{dc} and B_{ac}, as predicted by the critical state model with various forms of $J_c(B)$, have been reported in recent literature [1, 4, 8–10].

The transport $J_c(B)$ measured in sintered HTSC samples has been found to depend on the history of application of the magnetic field [11–13]. This, and related history effects in microwave measurements [7], have been understood by realising that a sintered pellet consists of grains and of intergrain regions and the transport $J_c(B)$ is dictated by the response of the intergrain region. The effective magnetic field in the intergrain region (say B_{eff}) has contributions from the externally applied field B_{ext} as well as the field generated by the magnetized grains. The magnetization of the grain depends on the history of application of B_{ext}, and B_{eff} mimics this history effect. The measured $J_c(B)$ is dictated by B_{eff} in the intergrain region, and must depend on the history of application of the external field. The minor hysteresis loop for small B_{ac}

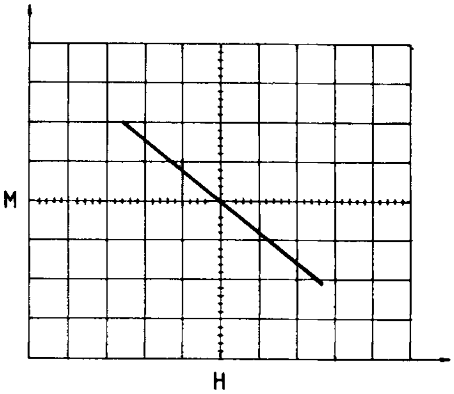

Figure 1a. M vs B loop, as seen on the oscilloscope for a YBaCuO pellet measured at $B_{ac} = 4$ G. Though the vertical scale is arbitrary for magnetization units, the oscilloscope was used in a 5·0 mV/cm configuration for the Y axis.

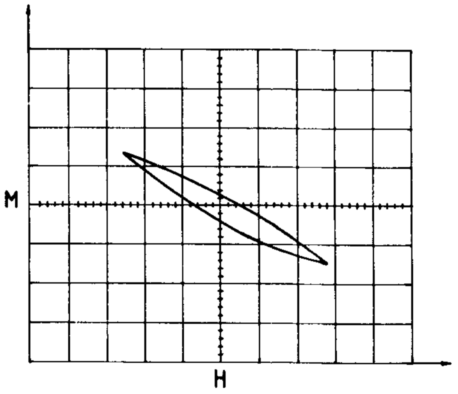

Figure 1b. Same as 1 (a) except that the pellet was momentarily exposed to $B_{dc} = 1$ kG (at 77 K) before measurement.

is attributed mainly to the intergrain region and, being dictated by B_{eff}, its shape should also depend on the history of application of B_{ext}. We have recently confirmed this by careful measurements on various sintered HTSC pellets [14], and figure 1 shows a typical result. Both the hysteresis loops shown are measured, with $B_{dc} = 0$ (earth's field) and $B_{ac} = 4$ G, on a YBaCuO pellet at 77 K. The only difference is that for figure 1 (b) the pellet was momentarily exposed to $B_{dc} = 1$ kG (at 77 K) before measurement in earth's field. This clearly shows that the shape of the minor hysteresis loop depends on the history of application of B_{dc}. The opening up of the hysteresis loop in figure 1 (b) indicates a power loss not seen in figure 1 (a), and provides a caution for accelerator cavity applications since accidental momentary exposure to a DC field can make the cavity show loss.

Since the shape of the minor hysteresis loop is history dependent, this dependence must also be seen in the harmonics. We have systematically demonstrated such history effects [15–17]. We have argued that only those measurements of harmonics in which B_{dc} was varied at $T > T_c$ (referred to as field-cooled data) could be readily compared with theory as only in this case is the DC part of $B_{eff}(t)$ equal to the applied B_{dc}. To our knowledge all reported measurements of M_n vs B_{dc} on HTSC pellets have so far been made with B_{dc} varied isothermally at $T < T_c$. In this paper we report the *first field-cooled data* on M_n vs B_{dc}. The measurements are made at 77 K on pellets of $YBa_2Cu_3O_7$ and $Bi_{1\cdot7}Pb_{0\cdot3}Sr_2Ca_2Cu_3O_{10}$. In the next section we present experimental details and results. We then present a qualitative comparison with theoretical calculations [10].

2. Experimental details and results

Measurements were made on sintered pellets of $YBa_2Cu_3O_7$ and $Bi_{1\cdot7}Pb_{0\cdot3}Sr_2Ca_2 \cdot Cu_3O_{10}$ (Bi-2223). The pellets were prepared by the standard solid state sintering route. For the former, Y_2O_3, $BaCO_3$ and CuO were calcined in air at 900°C (15 h) and 920°C (15 h) interspersed with grinding and mixing. The compacted (10 TS1) pellet was sintered in oxygen at 940°C (15 h) and was held at 400°C (15 h) during cooling. XRD showed no second phase, and oxygen was estimated by iodometry as 6·95. The sample had a resistive T_c of 91 K (confirmed by ZFC magnetization), and a 9 mm dia pellet was thinned to 0·25 mm thickness for the measurements discussed below. (The data shown in figure 1 are with a pellet of thickness 1 mm). For preparation of Bi-2223, $CaCO_3$, $SrCO_3$ and CuO were calcined twice in air at 950°C (15 h each) interspersed with grinding and mixing. $Sr_2Ca_2Cu_3O_7$ was then mixed with Bi_2O_3 and PbO in the right proportion and calcined at 800°C for 2 h followed by grinding and mixing. Pellet compacted at 10 TS1 was flash-heated at 915°C to obtain surface melting. It was crushed, pelletized and sintered at 855°C for 130 h. It was again crushed, pelletized and sintered at 865°C for 140 h. XRD showed no 2212 phase, and the sample has a resistive T_c of 107 K (confirmed by ZFC magnetization). The pellet used had a diameter of 9 mm and thickness of 0·7 mm.

The magnetization of these pellets was measured using a low-field hysteresis loop tracer [18], with a driving frequency of 317 Hz and with B_{ac} variable up to 18 G. The dc bias field was applied by a splitpair Helmholtz coil coaxial with the primary coil of the hysteresis-loop tracer. B_{ac} and B_{dc} were applied perpendicular to the plane of the pellet which was mounted in a glass dewar inside the pick-up coil. The magnetization signal from the output of the hysteresis loop tracer is fed to an HP 3582 A spectrum analyser where the magnitude of various harmonics is recorded. No attempt was made to cancel earth's field. All measurements were made at 77 K with B_{dc} applied with the sample at room temperature, and the sample field-cooled to 77 K. B_{ac} was kept $\leqslant 4$ G ($\leqslant 6$ G) for Bi-2223 (YBaCuO) pellet during field-cooling, and its magnitude was raised to the appropriate values at 77 K. No data were taken by reducing B_{ac} at $T < T_c$, due to associated history effects [14].

We now present the magnitude of various harmonics, as a function of B_{dc}, for various values of B_{ac}. Figures 2 to 5 show the results for Bi-2223, and figures 6 to 9 for YBaCuO. We outline below the salient features seen in these figures.

(i) We note that the second harmonic signal $2f$ (we denote the nth harmonic by nf) has its lowest value at $B_{dc} = 0$. This is also true for the $4f$ signal. This is consistent with the critical state model which requires that all even harmonics vanish for $B_{dc} = 0$. The small signal we observe in figures 2, 4, 6 and 8 can be attributed to the earth's field.
(ii) From figures 3 and 7 we note that $3f$ has a minimum as B_{dc} is raised, and this minimum occurs at higher values of B_{dc} as B_{ac} is raised.
(iii) $2f$ vs B_{dc} for YBaCuO (figure 6) has a small shoulder at 2 G, in addition to the main peak at 10 G, for $B_{ac} = 6$ G. This shoulder gains in intensity as B_{ac} is raised and

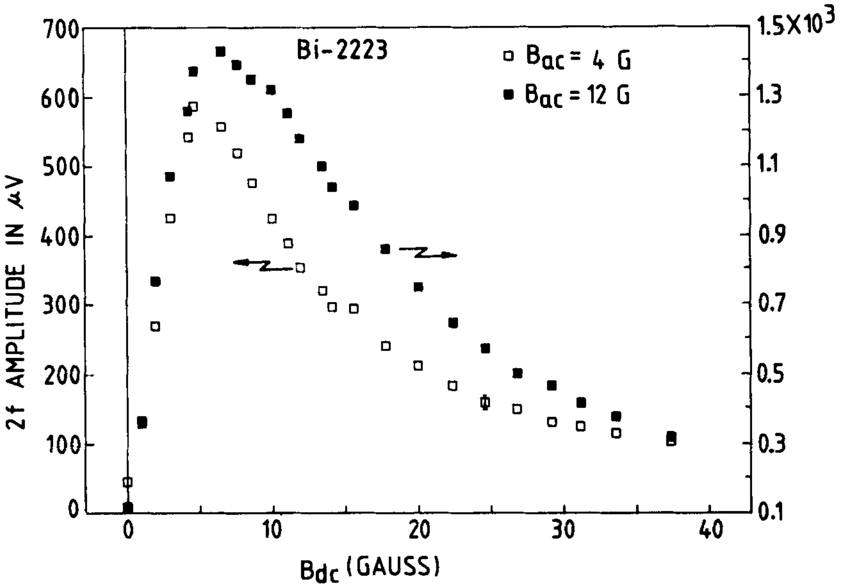

Figure 2. Second harmonic amplitude ($2f$) of Bi-2223 as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 4 and 12 G.

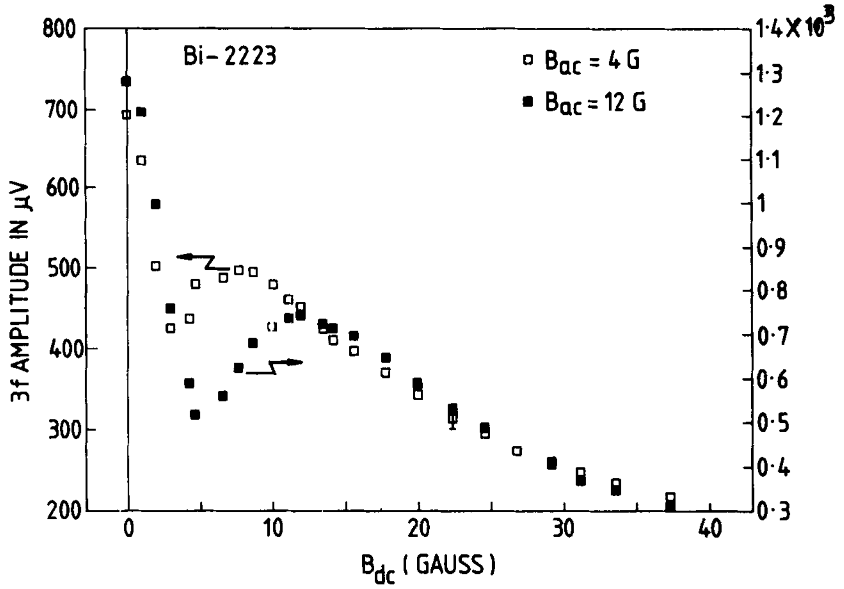

Figure 3. Third harmonic amplitude ($3f$) of Bi-2223 as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 4 and 12 G.

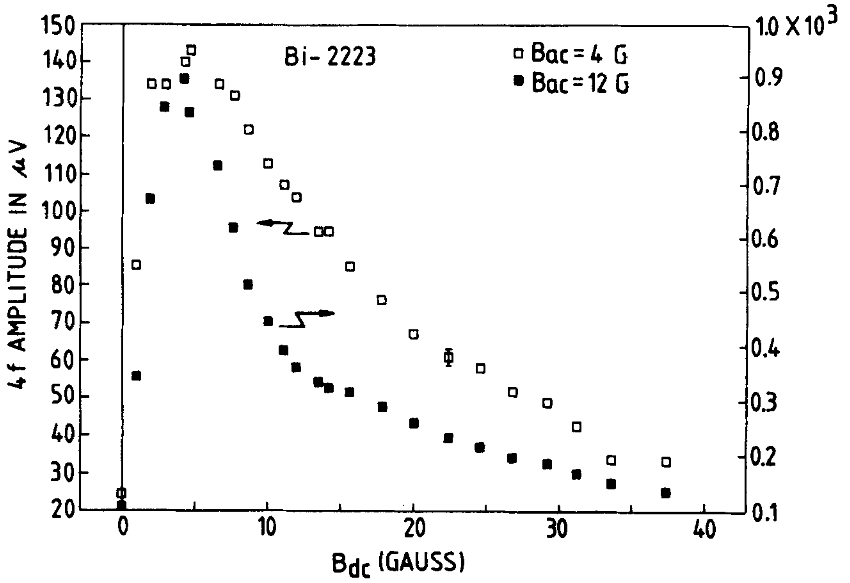

Figure 4. Fourth harmonic amplitude (4*f*) cf Bi-2223 as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 4 and 12 G.

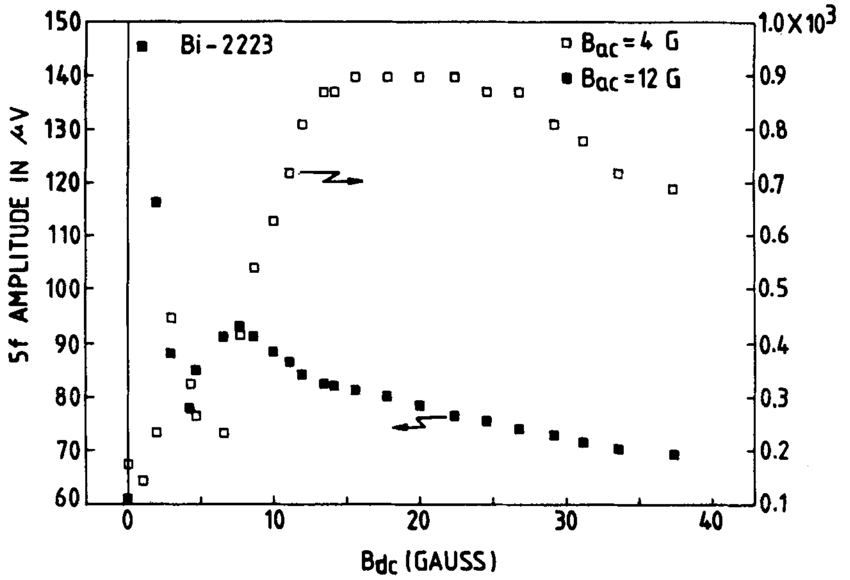

Figure 5. Fifth harmonic amplitude (5*f*) of Bi-2223 as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 4 and 12 G.

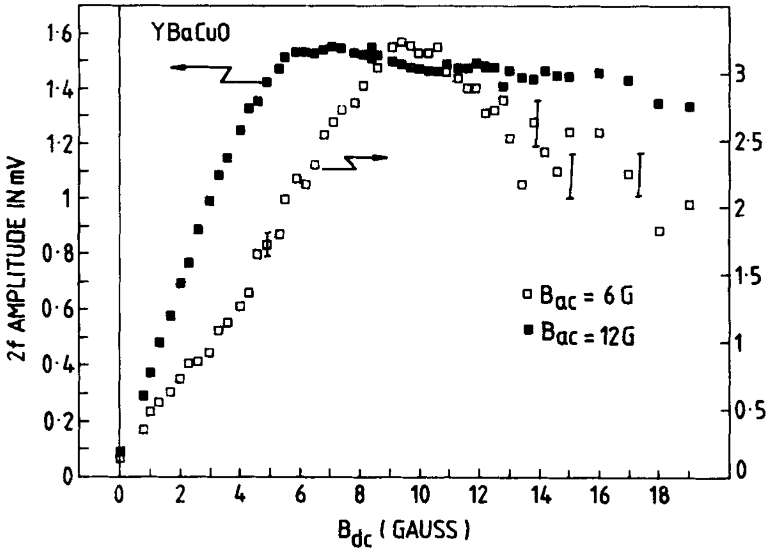

Figure 6. Second harmonic amplitude (2*f*) of YBaCuO as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 6 and 12 G.

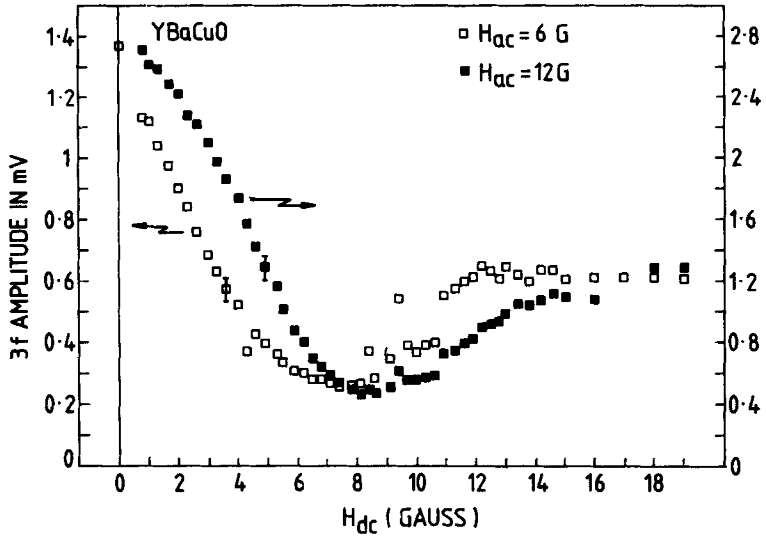

Figure 7. Third harmonic amplitude (3*f*) of YBaCuO as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 6 and 12 G.

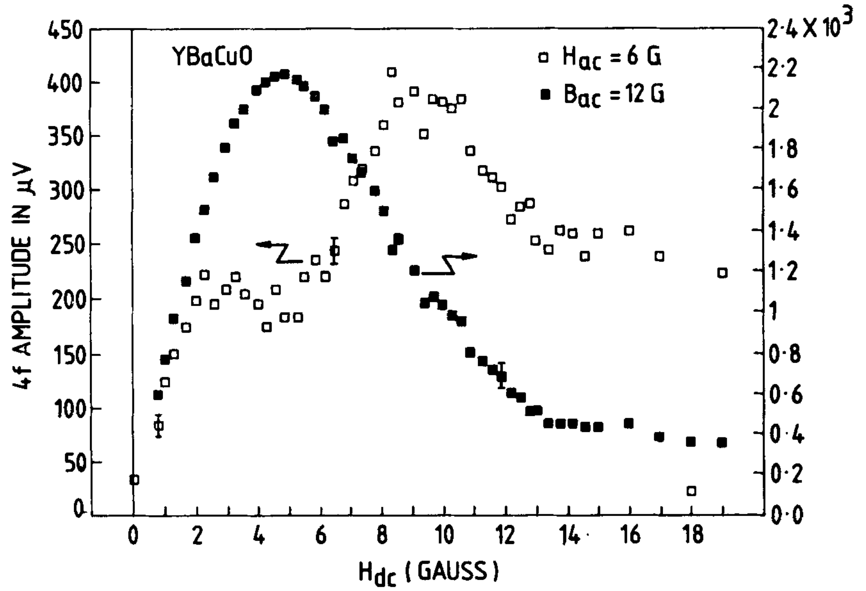

Figure 8. Fourth harmonic amplitude (4f) of YBaCuO as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 6 and 12G.

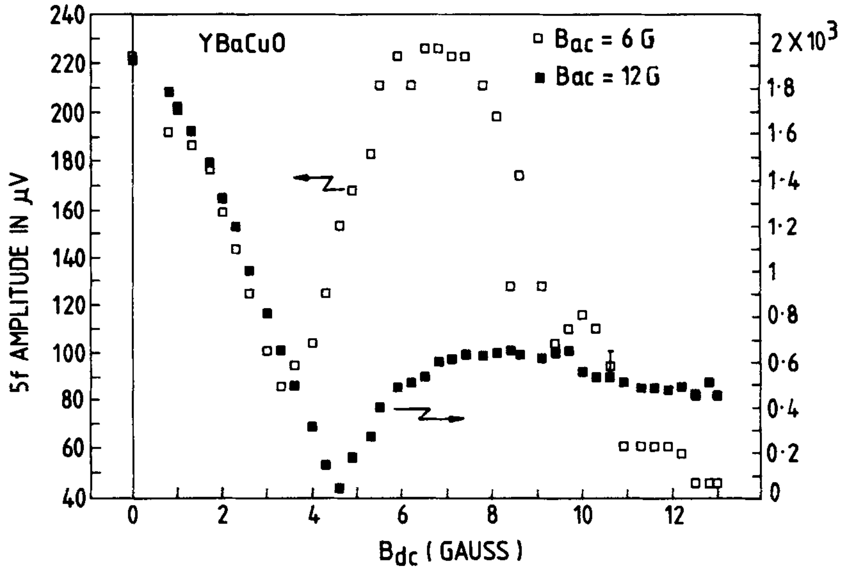

Figure 9. Fifth harmonic amplitude (5f) of YBaCuO as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 6 and 12G.

also moves to larger values of B_{dc} (indicated by arrows). The clear double-peak observed in ZFC data [17] is however, smudged in this FC data.

(iv) The $5f$ data (figure 5 and 9) shows the existence of multiple minima at the lowest value of B_{ac}, but this sharp structure gets smudged for both Bi-2223 and YBaCuO as B_{ac} is raised.

3. Comparison with calculations

As discussed in the introduction, calculations of harmonic magnitude, as a function of B_{dc}, have been performed within the critical state model. One general result [1, 10] is that if the magnitude of nf is plotted as B_{dc} is varied from $-B_{ac}$ to B_{ac}, there should be $(n-1)$ minima. This is consistent with the data in figures 2, 3 and 5 (for

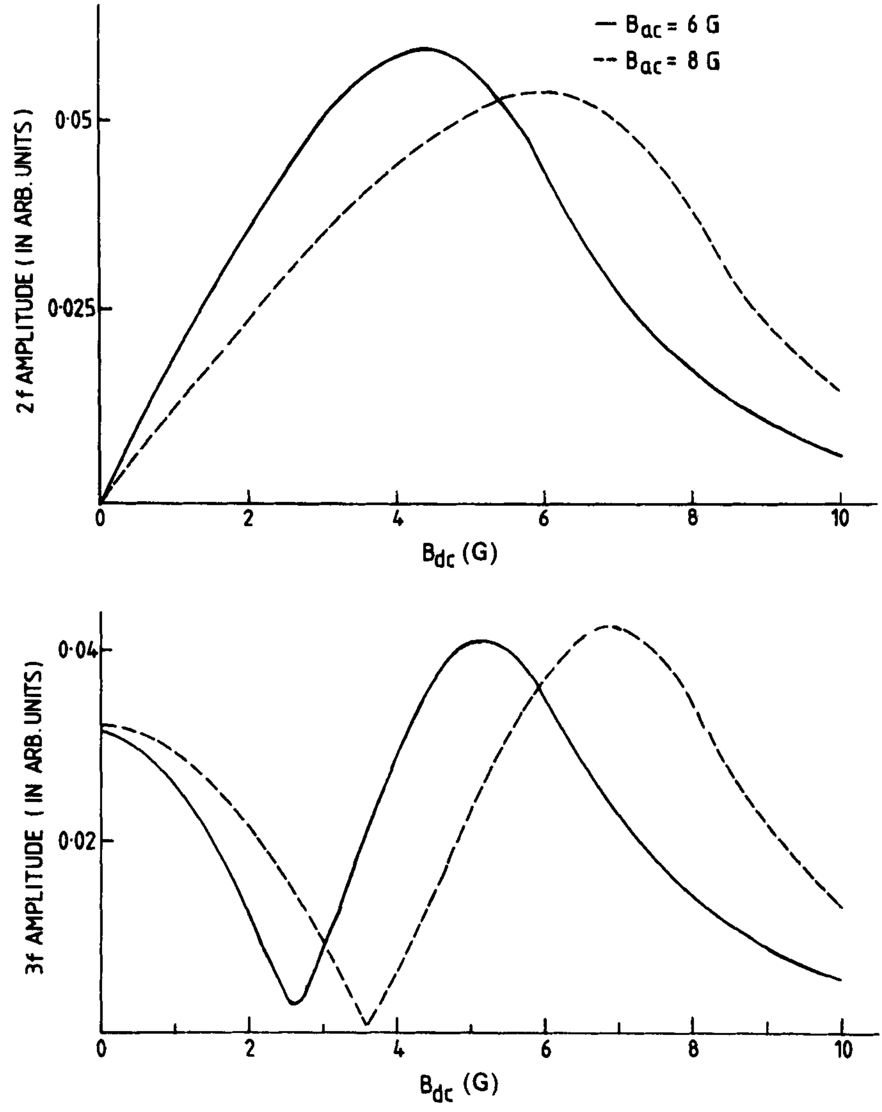

Figure 10. Calculated values of second ($2f$) and third ($3f$) harmonic amplitudes as a function of DC magnetic field (B_{dc}), the ac field (B_{ac}) being fixed at 6 and 8 G.

lowest B_{ac}) for Bi-2223 and with figures 6 and 7 for YBaCuO, but not for the other harmonics. The experimental observation (i) is, nevertheless, explained.

We show in figure 10 the calculated values of $2f$ and $3f$ as B_{ac} is raised. We assume in the calculation that $J_c(B) = J_c(0)\exp[-|B|/2]$. The calculations assume that Bean's parametric field [10] $B^* = 1$, and all fields in the calculation scale linearly with B^*. The peak in $2f$, and the minimum in $3f$, move to larger values of B_{dc} as B_{ac} is raised. This is consistent with the data in figures 2, 3 and 7 but the $2f$ data for YBaCuO in

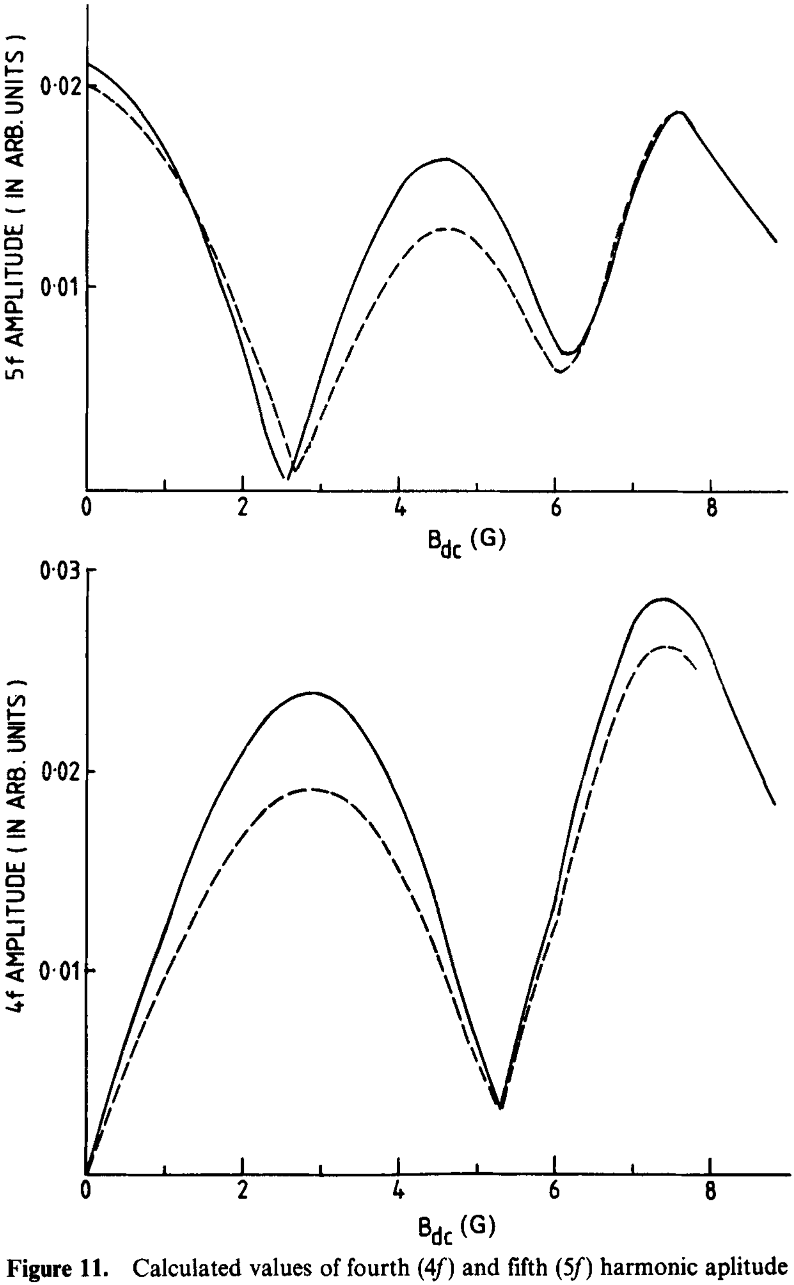

Figure 11. Calculated values of fourth ($4f$) and fifth ($5f$) harmonic aplitude as a function of DC magnetic field (B_{dc}) for two forms of $J_c(B)$ viz. $J_c(0)\exp(-|B|/2)$ (solid line) and $J_c(0)\,[1+|B|/3]^{-2}$ (dotted line).

figure 6 exhibits a shoulder that is not predicted by the calculation. The measured shoulder, as discussed by Roy *et al* [17], may be due to two parallel intergrain components. This is a complexity beyond the scope of existing calculations.

We show in figures 11 calculations of 4*f* and 5*f* for two forms of $J_c(B)$ viz. $J_c(0)\exp(-|B|/2)$ and $J_c(0)[1+|B|/3]^{-2}$. The latter form decays slower with B, and results in a smaller peak-to-valley ratio [10] in *nf* vs B_{dc}. This ratio is however still much larger than the experimental data presented in figures 4, 5, 8 and 9.

4. Conclusions

We have presented measurements of harmonic generation in sintered pellets of Bi-2223 and YBaCuO, with B_{dc} applied in the field-cooled mode. Qualitative features seen in the 2*f* and 3*f* data are consistent with the predictions of the critical state model. The behaviour of the higher harmonics calls for further refinements in the theoretical model.

Acknowledgements

One of the authors (PC) was introduced to the hysteresis loop tracer [18] in Prof. C Radhakrishnamurty's laboratory in 1987. The authors have used this versatile unit as the heart of their magnetization measurements. As Prof. Radhakrishnamurty nears the age of sixty, we wish to gratefully acknowledge the impact of his instrument on our work.

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