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Even harmonics of AC susceptibility in some sintered pellets of high temperature superconductors

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Abstract. The real (χ_n') and imaginary (χ_n'') parts of even harmonic susceptibility ($n \leqslant 6$) are measured as a function of external DC field (H_{dc}) in the field increasing ($H^{\uparrow}$) and decreasing ($H_{\downarrow}$) cycle. Hysteresis is observed between $H^{\uparrow}$ and $H_{\downarrow}$ cycles. In the $H_{\downarrow}$ cycle, at a field, $H_{\text{comp}} = 4{\cdot}2\,\text{G}$, both χ' and χ'' of all the even harmonics vanish indicating a true cancellation of fields in the intergrain region ($H_{\text{eff}} \approx 0$) caused by the balance between H_{dc} and the remanent magnetization of the grains. The position of the extrema and the zero of the various harmonics undergo a shift proportional to the remanent magnetization of the grains at that particular field.

Keywords. AC Magnetic susceptibility; even harmonics; high T_c superconductors; thermomagnetic hysteresis; remanent magnetization.

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1. Introduction

In sintered pellets of high temperature superconductors, the grains are interconnected through grain boundaries or weak links. AC susceptibility studies reveal interesting features linked with inter-grain and intra-grain flux penetration [1]. The observation of thermomagnetic hysteresis effects in $J_c(H)$ [2], harmonic voltages [3, 4] and minor loop magnetic hysteresis [5] in such materials have attracted attention.

One can understand the thermo-magnetic hysteresis in the following way. When a low value of external magnetic field, H_a, is applied, this field penetrates the weak links but is excluded from the grains, if it is below H_{c1} of the grains. The exclusion of the flux from penetrating the grains causes a concentration of flux in the intergrain region. This phenomenon termed flux compression causes the effective magnetic field in the intergrain region, H_{eff}, to be more than the applied field. When the applied field is reduced from a value large enough to ensure flux penetration within the grains, the remanent magnetization in the grains causes a field opposite in direction to the applied field in the weak link region. Thus the effective magnetic field is now lower than the applied field. Since J_c is a function of H_{eff} one sees hysteresis when J_c is plotted as a function of the applied field.

The occurrence of even harmonics in low amplitude AC susceptibility measurements requires (a) J_c to be a function of the effective magnetic field and (b) the presence of a non-zero DC magnetic field superposed on the AC field. In the decreasing cycle of the magnetic field one would reach a stage when H_{eff} will become zero. This will be signalled by the vanishing of all even harmonics at a non-zero value of the DC magnetic field. Reitz *et al* [6] have used the amplitude of the second harmonic signal to detect when an exact compensation was achieved in the following two situations: (a) The sample is field-cooled (FC) in a large value of H_{dc}. After cooling the field is switched off. Then a small DC magnetic field is applied and its value is adjusted to get a minimum in the second harmonic signal; (b) The sample is zero-field cooled (ZFC); then a large DC field is applied for 100 s and switched off. After a delay of 100 s the second harmonic is monitored as a function of applied DC field till its amplitude vanishes. The two values of the field at which the second harmonic vanishes are different, the value in case (a) being larger than in the case (b). The value of the field at which the second harmonic vanishes is a measure of the remanent magnetization and this was verified by a direct determination of the remanent magnetization in FC and ZFC cases.

In the present study we have verified that the real and imaginary parts of all the even harmonics up to the sixth vanish at nearly the same value of H_{dc} indicating that the H_{eff} in the inter-granular region is really zero.

2. Experimental details

The sample studied is $Y_1Ba_2Cu_{2\cdot8}Ag_{0\cdot2}O_7$ prepared by the conventional ceramic route; and is in the form of a cylinder of about 2 mm diameter and 5 mm length. The T_c (onset) and $T_c(0)$ determined from χ_{ac} and resistivity measurements are 88 K and 87 K, respectively. The harmonic voltages are measured as a function of DC field (H_{dc}) at an excitation field of $H_{ac} = 1\cdot9$ G, using a lock-in amplifier (SRS 850), which is able to lock into the various harmonics of the fundamental frequency, without the requirement of any additional external circuitry. The fundamental frequency of measurement is kept at 66·666 Hz and the measurements are done at 77 K, having the sample, primary, balanced secondary and the coaxial DC coil assembly immersed in liquid nitrogen bath. The primary excitation field is a pure sine wave and no measurable harmonic voltages were found in the absence of sample and at the highest excitation fields. The phase adjustment is done by making $\chi_1'' = 0$ in 0·015 G AC field. All the measurements are done in the presence of the earth's magnetic field. When we are mentioning about ZFC measurements it is actually FC to 77 K in earth's magnetic field. The absolute magnitude of the even harmonics is very small but is much above the noise and experimental resolution. Since the measured voltages are not calibrated with respect to a standard sample the units are maintained as microvolt (μV).

3. Results and discussion

Figure 1 shows the variation of χ_2' and χ_2'' as a function of the applied DC field. Figure 1(a) shows the variation with increasing DC field ($H^{\uparrow}$) while figure 1(b) shows the results in the decreasing cycle ($H_{\downarrow}$). The maximum applied DC field is 53 G. In the $H^{\uparrow}$ cycle both χ_2' and χ_2'' exhibit a maximum around $H_{dc} \approx 5$ G. The maximum in χ_2' is broader than in the χ_2'' and appears to show some structure. The hysteresis in the values of χ_2' and χ_2'' with $H^{\uparrow}$ and $H_{\downarrow}$ is apparent from the figure. For the same value of applied field, χ_2' and χ_2'' are more in the decreasing cycle than in the increasing cycle and are also shifted to higher values of the applied field. As the field is reduced below the value at which the peaks are observed, both χ_2' and χ_2'' appear to go to zero (within the experimental uncertainty) at $H_{dc} \approx 4\cdot2$ G. The vanishing of the real

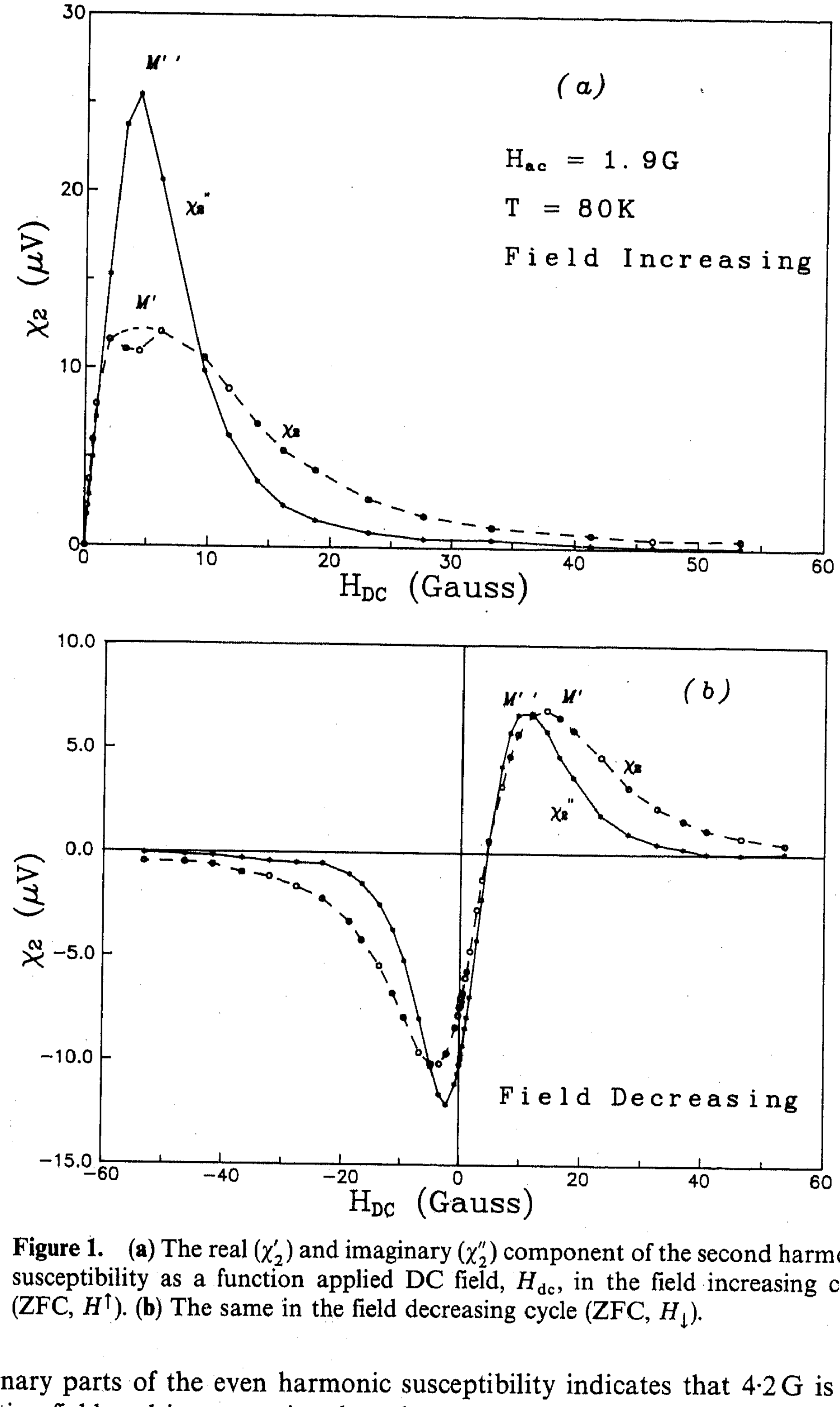

Figure 1. (**a**) The real (χ_2') and imaginary (χ_2'') component of the second harmonic susceptibility as a function applied DC field, H_{dc}, in the field increasing cycle (ZFC, $H^{\uparrow}$). (**b**) The same in the field decreasing cycle (ZFC, $H_{\downarrow}$).

and imaginary parts of the even harmonic susceptibility indicates that 4·2 G is the compensating field and is proportional to the remanent magnetization in the ZFC sample when it is exposed to a maximum field of 53 G. We find that the sign of both χ_2' and χ_2'' changes when H_{dc} is reduced below 4·2 G. With a spectrum analyzer, which will give only the amplitude, one could see a rise in amplitude below a minimum in the second harmonic signal [3]. However, the signal is not symmetric about the compensation field. The non-symmetric character can possibly be consequence of flux compression once the field in the intergrain region is reduced to zero and reversed.

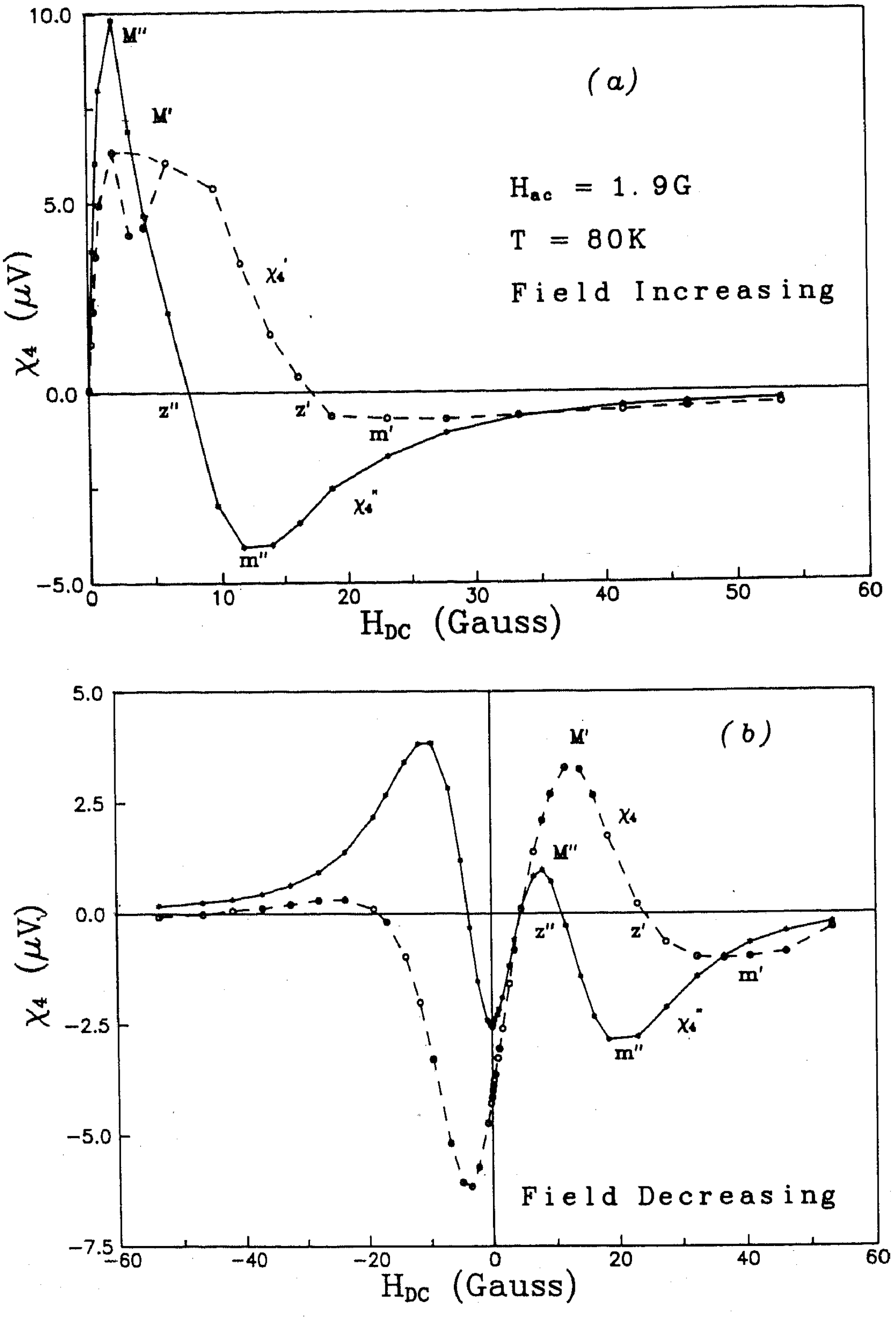

Figure 2. (a) The real (χ_4') and imaginary (χ_4'') component of the fourth harmonic susceptibility as a function applied DC field, H_{dc}, in the field increasing cycle (ZFC, $H^{\uparrow}$). (b) The same in the field decreasing cycle (ZFC, $H_{\downarrow}$).

Below H_{comp}, the effective field will be negative and as long as its magnitude is less than H_{c1} of the grain, there will be flux compression. So if we shift the origin of the magnetic field to the compensation point, the curve to the left will be compressed towards the y-axis resulting in an asymmetry. This is what is observed in the figure 1(b).

Figures 2(a) and 2(b) depict the variation of χ_4' and χ_4'' as a function of H_{dc} in the increasing and decreasing magnetic cycles respectively. Figures 3(a) and 3(b) show the variation of χ_6' and χ_6'' with increasing and decreasing fields, respectively. Both

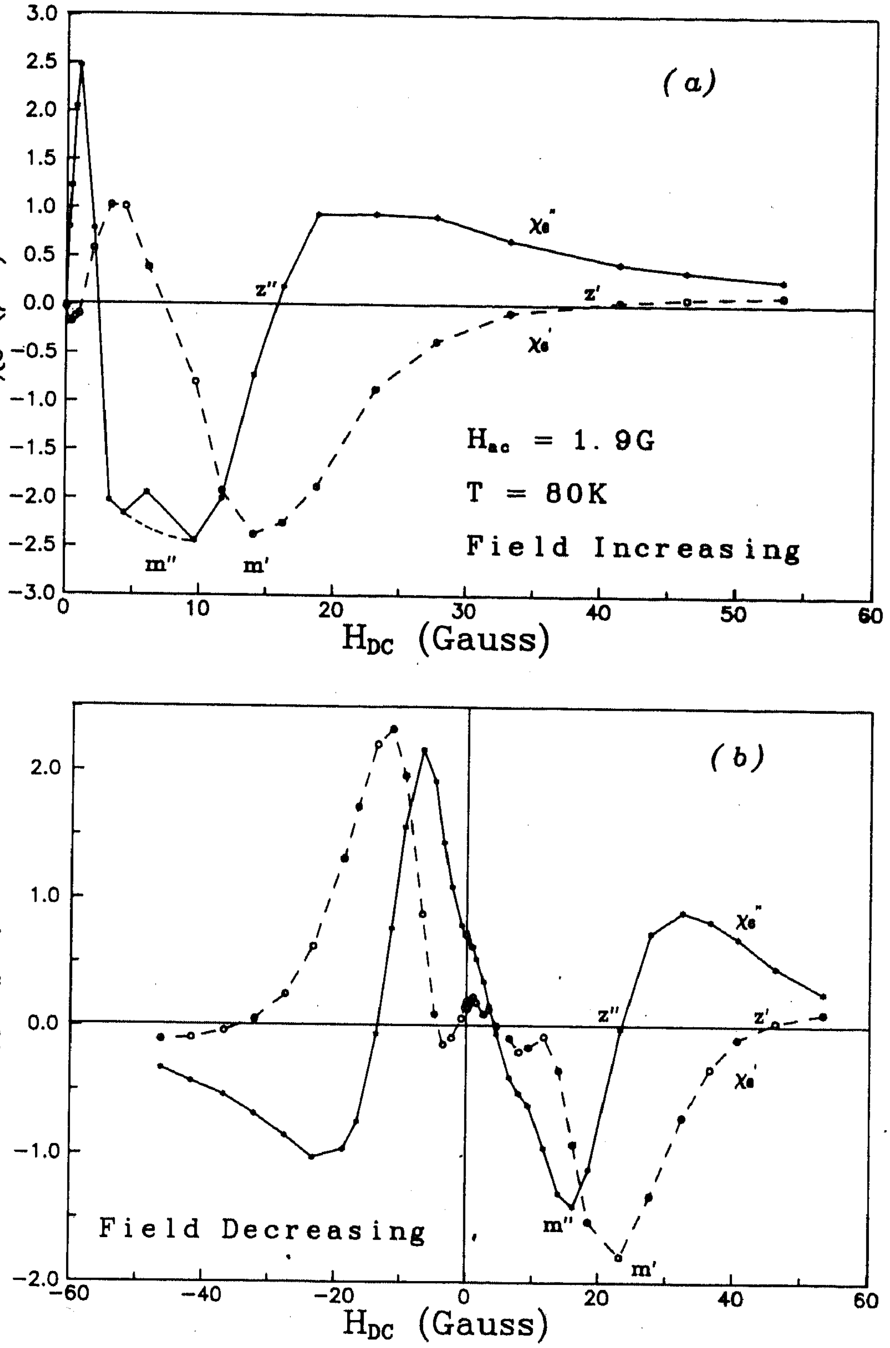

Figure 3. (**a**) The real (χ_6') and imaginary (χ_6'') component of the sixth harmonic susceptibility as a function applied DC field, H_{dc}, in the field increasing cycle (ZFC, $H^{\uparrow}$). (**b**) The same in the field decreasing cycle (ZFC, $H_{\downarrow}$).

χ_4' and χ_4'' change sign once in the increasing cycle. χ_6' and χ_6'' change signs twice with increasing field. However, the field at which the real part crosses the axis is different from that of the imaginary part both for χ_4 and χ_6. Whereas, on decreasing the field there is one value at which both real and imaginary parts vanish simultaneously. This occurs for χ_4 and χ_6 at 4·2 G which is the field at which the real and imaginary parts of χ_2 also vanish. This provides a clear indication that compensation of the intergrain field occurs when the magnetic field is at 4·2 G.

Table 1 gives the value of $H_{\rm dc}$ at which maxima, minima and zero of the real and imaginary parts of χ_2, χ_4 and χ_6 occur with increasing and decreasing fields. The difference in the position of either the extrema or zero, between increasing and decreasing field, at a particular field, is a measure of the remanent magnetization of the grains at that particular field. In figure 4 we plot these differences as a function

Table 1. DC fields at which maxima (M), minima (m) and zero values (z) occur in the real and imaginary parts of the even harmonics. The zero at the compensation field is omitted.
(Those fields which are larger than the compensation field and which can be reasonably well fixed are collected in the table).

Component	Figure	Field increasing	Figure	Field decreasing
2′	1a(M')	4·4	1b(M')	13·8
2″	1a(M'')	3·5	1b(M'')	10·0
4′	2a(M')	4·2	2b(M')	12·7
4′	2a(z')	17·6	2b(z')	24·2
4′	2a(m')	Broad	2b(m')	Broad
4″	2a(M'')	1·8	2b(M'')	7·9
4″	2a(z'')	7·8	2b(z'')	10·8
4″	2a(m'')	12·4	2b(m'')	20·0
6′	3a(m')	13·7	3b(m')	23·0
6′	3a(z')	38·1	3b(z')	44·0
6″	3a(m'')	7·0	3b(m'')	15·8
6″	3a(z'')	15·7	3b(z'')	23·0

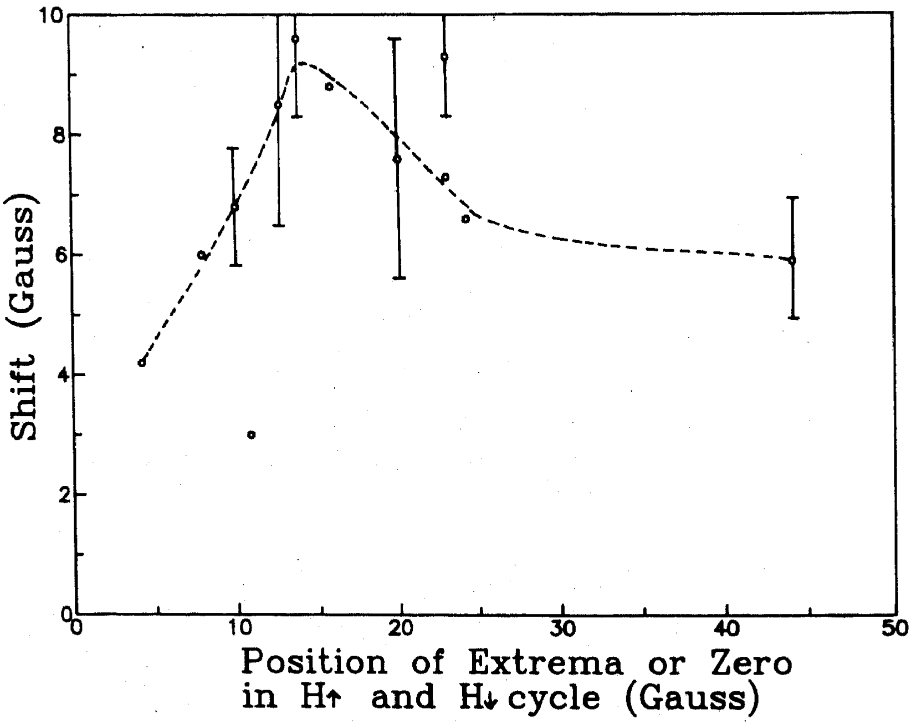

Figure 4. Plot of the shift in the position of the extrema and zero between $H^{\uparrow}$ and $H_{\downarrow}$ cycles as a function of the position of the extrema and zero in $H^{\uparrow}$ and $H_{\downarrow}$ cycles.

of the position of the extremum and zero. We find that the remanent magnetization shows a maximum at $H_{dc} \approx 15\,G$. A similar behavior would be obtained if a plot of the difference in the magnetization $M(\mathrm{ZFC}, H^{\uparrow})$ and $M(\mathrm{ZFC}, H_{\downarrow})$ is made.

4. Conclusions

The even harmonics exhibit hysteresis between the field increasing and decreasing cycle. The hysteresis is due to $H_{\mathrm{eff}}(\mathrm{ZFC}, H^{\uparrow}) > H_{\mathrm{eff}}(\mathrm{ZFC}, H_{\downarrow})$. The vanishing of the real and imaginary parts of all the even harmonic susceptibilities at H_{comp} gives an unambiguous indication that H_{eff} in the intergrain region is truly zero at this field. The maximum cancellation of fields in the intergrain region occurs when H_{dc}, equals in magnitude, with the field caused by the trapped magnetization of the grains. The position of the extremum in the real and imaginary parts of the various harmonic susceptibility undergoes a shift which is proportional to the remanent magnetization of the grains at that particular field.

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