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DETERMINATION OF FIELD AND TEMPERATURE DEPENDENCES OF THE INTERGRAIN CRITICAL CURRENT DENSITY $J_c^{\omega}(H,T)$ IN HIGH-TEMPERATURE SUPERCONDUCTORS

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I. Introduction

High-temperature superconductors (HTS) having a short coherence length, should have a large current carrying capacity without any losses, i.e., a large value for the critical current density J_c , if the main mechanism limiting J_c is depairing Cooper pairs (e.g., the theoretically evaluated value of the depairing J_c for $YBa_2Cu_3O_{7-\delta}$ (Y123) at 4.2K is about $10^9A/cm^2$ [1]).

Contrary to the low- T_c superconductor, whether a single chemical element or a solid solution of some elements, the HTS is like a "mixture" of two systems: a system of isolated grains and a built-in system of intergranular weak links.

While the phase structure is determined by the grain system, the superconducting properties are determined by the whole "mixture". Especially at low value ranges of the three main parameters: temperature, magnetic field and current, the superconducting properties are commonly determined not by the grain system but the system of intergranular weak links.

The above means, that in HTS the depairing mechanism is not the case and the critical current density J_c might therefore be controlled by the depinning or decoupling mechanism. The critical current J_c then becomes not an intrinsic property of the material but depends on the way it was prepared. Thus it becomes very topical to elaborate a method to determine J_c and its field and temperature dependences. Classifying J_{c} -determination methods used in practice can be made by noting the kind of current passing through the samples:

1. Transport current method

In this method, the current (direct or pulse) passing through a sample comes from external current sources. The current is determined as critical when a voltage appears between the two potential probes (usually with the criterium V=0.1 $\mu V/mm$). Because of influences of the self-magnetic field related to this current, the critical current value is lowered and is dependent on sample size. Thus, in this case it is difficult to determine the value of J_c and its field and temperature dependences exactly. 2. Magnetization current method

The supercurrent circulating in a sample is created by an external excitation magnetic field (dc or ac). In a dc-magnetic field H_{dc} generating a sample magnetization M, J_c is proportional to the gap between the major hysteresis curve $M^+(H_{dc}^{\dagger}) - M^-(H_{dc}^{\dagger})$ (M^+, H_{dc}^{\dagger} and M^-, H_{dc}^{\dagger} correspond to the ascending and descending branches of the magnetization curve, respectively) in a large field range, where M^+ and M^- are weakly dependent on H_{dc} [2]. This J_c -determination can ignore any influences of reversible magnetization M_{eq} but yields only the value of J_c for the grain system standing fully in a critical state and ignores any information concerning the system of intergranular weak links. Theoretically, J_c could be evaluated by using the minor loops limited between the major branches of the magnetizations curves [3], but this is not easy to perform experimentally.

In the case when the exciting field is an ac-field $H_{ac} = H_o \sin\omega t$, J_c can be estimated by using the peaked H_{ac}^p , which is the full penetration of the ac-field into a sample and appears in the imaginary part of the ac-susceptibility at a temperature T_p and dc-field H_{dc}^p [4]. This kind of J_c -evaluation is usually complicated whenever the sample is of not the right geometry or is not homogeneous.



It is supposed that J_c could also be determined by measuring the screen effect of an external ac-field [5] provided that a suitably thin sample could be made so that the penetration field profile is linear. A practice widely in use is the method of fitting the theoretical curves of the field and temperature dependent ac-susceptibility $\chi', \chi''(H,T)$ with the proposed $J_c(H,T)$ over $\chi', \chi''(H,T)$ -experimental data [6]. Although the latter method seems to be more hopeful, it strongly depends on the choice of functions $J_c(H,T)$. These $J_c(H,T)$ functions should not be primary in the integral evaluating the dependences $\chi', \chi''(H,T)$ but in the effects on χ', χ'' through the magnetization averaged over the ac-field period and the sample volume that lowers precisely this kind of J_c -determination.

In this work we present our own version of determining the intergrain critical current density $J_c^w(H,T)$ using the field and temperature dependences of the acsusceptibility, $\chi', \chi''(H_{dc},T)$, measured in a low ac-field $H_{ac}(H_q << H_{ac}^p)$. Here and further the upper index w is omitted. The method is presented in detail in the second section. The use of this method to determine $J_c(H,T)$ and discussions of $YBa_2Cu_3O_{7-\delta}$ (Y123) and $Bi_{1.65}Pb_{0.35}Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223) ceramics as well as a Y123:8% Ag thick film are given in the third section.

II. Working model

Assuming that the samples have the right form (cylinder of radius R or parallelepiped with a square cross section $4R^2$), with one axis long enough to neglect the demagnetization effect placed in the external axial H_{ac} and H_{dc} fields, our assumptions are as follows (see Fig.1):

1. The ac-field penetration length $(R - r_p)$ is smaller than the parameter R.

2. The investigated samples are homogeneous. Therefore $J_c(T, H_{dc})$ determined by $\frac{dH_i}{dr}|_{r=R-\lambda_L} (\lambda_L \text{ is the London length}, H_i \text{ is the penetrated magnetic field dependent} on T and <math>H_{dc}$) presents the $J_c(T, H)$ of samples. This determination together with the first assumption allows the linear internal H_{ac} profile to be assumed.

3. The surface effects are negligible.

4. The measuring ac-field is smaller than H_{dc} .

Provided that the H_{ac} field is larger than the first critical field of the intergranular weak link system H_{cl}^{w} , the ac-field penetrates the sample radially. For the given sample geometry (see Fig.1) we have the critical state equation:

$$\frac{dH_i}{dr}\Big|_{r=R_o=R-\lambda_L} = J_{co}(T, H_{dc}) = tg\alpha, \tag{1}$$

where J_{co} is the critical current density dependent on T and H_{dc} . This formula takes into account the reversible penetration of external fields (dc- or ac-field of low frequency $\omega < 1MHz$ [7]) along the length of λ_L . In the case of $\lambda_L << R$ it may be considered that $R_o = R$.

The real and imaginary parts of the fundamental ac-susceptibility, measured at a temperature T and dc-field H_{dc} with the $H_{ac} = H_o sin\omega t$ for the sample volume unit,

can be expressed mathematically as :

$$\chi' = \frac{1}{\pi H_o} \int_o^{2\pi} M(t) \sin\omega t d\omega t$$
 (2)

$$\chi'' = \frac{-1}{\pi H_o} \int_o^{2\pi} M(t) \cos\omega t d\omega t \tag{3}$$

Here M(t) is the time-dependent magnetic moment averaged over a sample volume. Between the M(t) and the bulk local supercurrent $J_c(r, t)$ exists the relation:

$$M(t) = \frac{-1}{R^2} \int_0^R J(r,t) r^2 dr,$$
 (4)

which can be rewritten as:

$$M(t) = \frac{-1}{R^2} J_{co}(T, H_{dc}) \int_o^R r^2 dr,$$
 (5)

when the four assumptions mentioned above are valid. Thus, it is obvious, that one can estimate the dependences of J_c on T and on H_{dc} by means of measuring the field and temperature dependences, $\chi', \chi''(T, H_{dc})$.

Evaluating the averaged moment M(t) after equation (4) is given in the Appendix. Substituting expressions A4, A5 of M(t) into equations (2, 3) followed by intergration gives us the expression for $\chi', \chi''(T, H_{dc})$ as follows:

$$\chi' = -1 + \frac{H_o}{RJ_{co}(T, H_{dc})} - \frac{5}{16} \left(\frac{H_o}{RJ_{co}(T, H_{dc})}\right)^2 \tag{6}$$

$$\chi'' = \frac{4}{3\pi} \frac{H_o}{RJ_{co}(T, H_{dc})} - \frac{2}{\pi} (\frac{H_o}{RJ_{co}(T, H_{dc})})^2$$
(7)

Expressions analogous to equations (6, 7) with some modifications were also obtained in [8]. In accordance with the first assumption we can write

$$\chi' + 1) = \frac{H_o}{R} \frac{1}{J_{co}(T, H_{dc})}$$
(8)

$$\chi'' = \frac{4}{3\pi} \frac{n_o}{R} \frac{1}{J_{co}(T, H_{dc})}$$
(9)

 $\chi'' = \frac{4}{3\pi}(\chi' + 1) \tag{10}$

Equations (8-10) form the basis for evaluating the field and temperature dependences of J_c . The experimental procedure consists of the following operations:

1. Measure $\chi', \chi''(T, H_{dc})$ dependences of investigated samples of the right shape. In our study $H_o = 0.2 - 2.5Oe$, $\omega = 68Hz$, T = 77 - 120K, $H_{dc} = 0 - \pm 320Oe$. The samples geometries are listed in Table 1.

2. Plot the graphs of $\chi''(H_{dc})$ as a function of $\chi'(H_{dc}) + 1$ at a given temperature, or of $\chi''(T)$ as a function of $\chi'(T) + 1$ in a given field H_{dc} . The validity of the above four assumptions must be approved by the presense of the initial sections on these plots where χ''_1 is linearly dependent on $(\chi'_1 + 1)$ with the slope $4/3\pi \approx 0.425$. Moreover,

these linear sections estimate the ranges of the field $(H_{dc} = H_{dc}^{min} - H_{dc}^{max})$ as well as the temperature $(T = T_{min} - T^{max})$, at which the evaluation of the $J_c(T, H_{dc})$ after (8) or (9) could be performed.

3. Determine $J_c(H_{dc})$ at given T with equation (9) and $J_c(T)$ at a given H_{dc} with equation (8). The absolute value of J_c has to be determined by calibrating the obtained $J_c(T, H_{dc})$ function to values of $J_c(T = 77K, H_{dc}^p)$ estimated by means of the peaked dc-fields H_{dc}^p observed on the curves of $\chi''(T = 77K, H_{dc})$ measured with various ac-fields H_{ac} .

III. Experimental and Discussion

III.1. Sample preparation

Sample S1

Sample S1 is a slab $0.9 \times 0.205 \times 0.205 cm$ in size with a density $d = 6.2 \text{ g/cm}^3$ cut from the $YBa_2Cu_3O_{7-\delta}$ ceramic prepared by the standard ceramic technology from $BaCO_3$, CuO, Y_2O_3 of the 99.99 % purity. Since the main mechanism forming the 123-phase is diffusion, many-fold grindings and pressings must be applied. The oxygen content in the unit cell was taken up to the equilibrium value of ~ 7 by annealing the samples for a long time at 350°C in a 1 atm oxygen atmosphere. X-ray diffraction confirmed a single 1-2-3 phase in the sample and the value $\delta \approx 0$ (after the unit cell parameter c).

Sample S2

This sample is also of single phase Y123, but is prepared from the precursors Y_2BaCuO_5 , $BaCuO_2$ and CuO. Unlike the standard ceramic technology, a mixture of these precursors weighted in the right ratio was quickly heated to 985° C, held at this temperature 3for some minutes to melt the mixture of $BaCuO_2 + CuO$, then slowly cooled from 985° C to 920° C at a rate about 1 deg/min to form the Y123 phase. Because of local melting, sample S2 had a lower density, about ~ 5 g/cm³. To measure ac-susceptibilities one slab, $0.92 \times 0.22 \times 0.22cm$ in size, was cut from the pellet.

Sample S3

Sample S3 is a Y123:8% Ag thick film 300 μm thick prepared by painting a $(3BaCuO_2 + 2CuO)$ mixture on the cylindrical Y_2BaCuO_5 -substrate of radius $R \simeq 0.11cm$ and length 0.87cm, followed by synthesizing described in detail in [9]. The 8% Ag-doping of this sample was made by adding Ag to the $(3BaCuO_2 + 2CuO)$ mixture in a mass ratio of 8%. The film has a very high density (about that of X-ray density) confirmed by studying the sample morphology on the JSM-840 electron microscope.

Sample S4

The last sample is a $Bi_{1.65}Pb_{0.352}Sr_2Cu_3O_{10+\delta}$ ceramic slab $0.72 \times 0.2 \times 0.2cm$ in size with a density of about $5.5g/cm^3$. To get the single 2223 phase, a precusor mixture of $Ca_2Sr_2Cu_3O_8$ and $(Bi_2O_3 + PbO)$ was synthesized by a three-fold thermal shock at 930°C followed by a long annealing time in air at 845°C with intermediate grindings. That sample is single 2223 phase was verified by X-ray diffraction and by the fact that the resistance versus temperature curve showed only one sharp drop at $T_c \sim 110K, \Delta T = 5K.$

III.2. J_c -evaluation

The $\chi', \chi''(T, H_{dc})$ measurements of the investigated samples were carried out on a home-made ac-susceptometer [10]. Every measurement started by heating the sample to a temperature larger than T_c , ZF-cooling it to 77K, then either heating it again by a rate of about 1 deg/min to measure the temperature dependences or exciting it by increasing the external H_{dc} from zero to 320 Oe to measure the field dependences.

Fig. 2 shows the mutual relations between χ'' and $(\chi'+1)$ measured with various ac-field amplitudes $H_o = 0.25 - 2.5Oe$ at T = 77K, in $H_{dc} = 0 - 320Oe$ (Fig.2a, 2b, 2c) and in $H_{dc} = 0$ at T = 77 - 92K (Fig. 2d, 2c, 2f). It is noted that while the criterium $\chi_1'' = 0.425(\chi_1'+1)$ is observed for samples S1 and S2, there are deviations for S3 and S4 (the slopes are 0.7 and 0.5 instead of 0.425, respectively). The deviation for S3 seems to be reasonable, taking into account that S3 is not a solid but rather a hollow cylinder of thickness $d \approx 300 \mu m$. In the latter case, if $d \ll R$ equation (10) must be modified as $\chi'' = 0.425(R/2d)(\chi_1'+1)$. Therefore, the slope becomes $\sim 0.425(0.11/2 \times 0.03) \approx 0.7$. The slight deviation for S4 is thought to be due to the more complicated structure of intergranular weak links of the Bi-system (see below).

Following the above explanation one uses equation (9) to determine the field dependences of J_c for the investigated samples. A typical plot of χ''_1 as a function of H_{dc} at T = 77K is presented in Fig.3. The solid lines are power law fitting curves of the experimental data. It is worth noting that for sample S1, for various ac-fields, the function $J_c(H)$ can be expressed as $H^{-\beta}$ with $\beta = 1.43 \pm 0.07$ at T = 77K

This Kim-like functional, $H^{-\beta}$, also appears to be valid for samples S2-4. The values of the factor β , estimated by fitting in this way, are equal 1.43, 1.46, 1.62 and 1.43 for S1-4, respectively.

Estimating the temperature dependence $J_c(T)$ in a zero H_{dc} field was performed in a similar way but by using equation (8) rather than equation (9). The fitting treatment has shown that the $(\chi''(T)+1)$ (or $J_c(T)$) of samples S1-4 measured with a low ac-field $(H_o \sim 0.25Oe)$ can be best fitted by a function of the kind $(1-T/T_c^w)^n$ (see Fig.4). The values of the parameter n extrapolated to zero ac-field 2.18, 1.79, 2.62, 3.13 for S1-4, respectively. Here the temperature T_c^w is the onset critical temperature for the system of intergranular weak links and is determined as the temperature at which the low temperature peak of $\chi''(T)$ vanishes.

The explicit $J_c(T, H)$ function of the investigated samples which has the form:

$$J_c(T,H) = (1 - T/T_c^w)^n / (H_1 + |H_i|)^{\beta},$$
(11)

was obtained by calibrating the fitting results using the peaked dc-fields H_{dc}^p as mentioned in Section II to estimate the parameters α and H_1 , which remain free after the fitting procedures. The relation between $J_c(77K, H_{dc}^p)$, the full penetration ac-field H_{ac}^p and the penetration length (i.e. the sample parameter R) agrees:

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$$J_c(77K, H_{dc}^p) = (H_{ac}^p)^{\beta+1} / ((\beta+1)R).$$
(12)

Using equations (11) and (12) the explicit functions $J_c(T, H)$ of the samples S1-4 are estimated to be:

For S1:

$$J_{c}(\frac{A}{cm^{2}}) = 4.93 \cdot 10^{7} (1 - T(K)/89.2)^{2.18} / |H(Oe)|^{1.43}, \quad (13)$$
For S2:

$$J_c(\frac{14}{cm^2}) = 9.34 \cdot 10^{\circ}(1 - T(K)/90.3)^{110}/|H(Oe)|^{110},$$
 (14)
For S3:

$$J_{e}(\frac{A}{cm^{2}}) = 1.16 \cdot 10^{9} (1 - T(K)/90.1)^{2.62} / |H(Oe)|^{1.62},$$
(15)

$$J_c(\frac{A}{cm^2}) = 1.45 \cdot 10^7 (1 - T(K)/99)^{3.13} / |H(Oe)|^{1.43}.$$
 (16)

In the calibrating process the parameter H_1 seemed to have a large sparseness (from 1 to 10 Oe), and for definition it was taken equal zero.

III.3.Discussion

To discuss the $J_c(T, H)$ of the investigated samples the main results are listed in Table 1.

An ac-magnetic field with a flux density H_o above the lower critical field H_{c1}^{w} enters the system of intergranular weak links, the permeability of which, is affected by the grain system. The profile of the ac penetrated field results from the balance of the Lorentz force and the pinning force disturbed by thermal activation processes such as flux creep or flux flow. Generally, this profile is described by the diffusion-like equation (e.g. for the cylinder geometry)[11]:

$$\frac{\partial H_i}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} (rD \frac{\partial H_i}{\partial r}) \tag{17}$$

where the diffusion coefficient expresses the above mentioned balance.

In the stationary case (our case, in which there are low dc- and ac-fields, as well as low ac-field frequency), $\frac{\partial H}{\partial t} = 0$, Thus, one has the critical state equation:

$$\frac{dH_i}{dr} = sgn(H_i)J_c(T, |H_i|) = sgn(H_i)\frac{\alpha_p}{|H_i|}$$
(18)

and the critical current density $J_c(T, |H_i|)$ depends on the temperature and field dependent pinning ability of the sample pinning centers. As the pinning force α_p is proportional to H_i , J_c is a constant in the critical state (Bean model). Any deviation of this linearity results in a field dependence of J_c . The Kim model, $J_c \sim 1/H_i$, is the case when α_p is independent of H. Thus, if the depinning mechanism is dominated, the parameter β migh be varied in the interval 0-1.

As Clem et al. [12] appreciated, in granular superconductors, the order parameter is depressed by the supercurrent flowing through the sample. This depression changes the balance, which creates an inner field profile, but at any point of the sample the Ampere law holds. Thus the J_c determination process presented in Section II remains true also for this case, where J_c can be drastically reduced by a thin layer (insulating or normal), through which the supercurrent has to tunnel (decoupled current). The critical current density controlled by the Josephson effect is highly sensitive to magnetic fields, in contrast to that limited by depinning or by depairing.

Judging from the experimental data of the critical current field dependences given in Section II (see the values of the parameter β in Table 1) we can conclude that in the investigated samples, S1-4, the critical current density J_c is limited by the decoupling mechanism.

According to [13], if the barriers between the superconducting grains are of an insulating nature (S-I-S) the decoupling current temperature dependence, near T_c , is $J_c(T) \sim (1 - T/T_c^w)$. In the case of (S-N-S) barriers (N is normal metal), close to T_c one finds $J_c(T) \sim (1 - T/T_c^w)^2$.

As estimated in Section II, the values of the parameter n are equal 2.18 and 1.79 for samples S1-2, respectively, that means the critical current density of these samples could be described in terms of arrays of S-N-S type of weak links.

The granular (S-N-S) model remains valid for the Ag-doped YBCO-thick film (sample S3), but Ag-doping, in which Ag atoms are arranged within the grain boundaries, gains the temperature dependence of the critical current density. By doping the 8% Ag the n value of sample S3 is increased to 2.62. For sample S4 the n value is larger and equals 3.13. This situation might be more understandable if one takes into consideration the fact that because the Bi2223 phase is created from the Bi2212 phase, the system of intergranular weak links in sample S4, therefore, is more complicated than that in samples S1-3.

IV. Conclusion

We have shown how to determine $J_c(T, H)$ using experimental data of the real and imaginary parts of the fundamental ac-susceptibility. Our working model is valid for the assumptions given in Section II. This validity must first be checked by equation (10). Then temperature and field dependences can be estimated by equations (8) and (9), respectively. The absolute value of $J_c(T, H)$ is calibrated using the peaked appearance at H_{dc}^p on the curve $\chi_1''(T = 77K, H_{dc})$, taking into account that the relation between H_{dc}^p , J_c and the penetration length must be considered in the context of the real field dependence of J_c .

The $J_c(T, H)$ functions for the Y123-bulk ceramic sample (S1), the Y123-bulk sample prepared from precursors (S2), the painted-on thick film Y123 - 8% Ag (S3) and the Bi2223-bulk ceramic sample (S4) have been determined by the above method and have the explicit form as expressed by equations (11-14).

These $J_c(T, H)$ functions show that, in these samples, the main mechanism limiting J_c^w is decoupling, and the layers, by which the superconducting grains are coupled, are of a normal metal nature (S-N-S type weak links). It is observed the role of the Ag atoms doped in the YBCO-system in gaining the temperature dependence of J_c . In the Bi2223-system the functional $J_c(T)$ is strongly deviated from the standard form $J_c(T)$ of the granular (S-N-S) model, that it thought to connect with a more complicated structure of intergranular weak links in this system in comparison with one of YBCO-systems.

Table 1. Summary of the sample characters, the fitting parameters n, β and α of the explicit forms of $J_c(T, H) = \alpha (1 - T/T_c^w)^n / |H|^\beta$. Here J_c in A/cm^2 , T in K, H in Oe.

N°	Sample	Form	density	dimensions	T_c^w	n	β	α
			(g/cm^3)	(cm)		1997		:
S1	Y123-bulk ceramic	Slab	6.2	$0.9 \times 0.205 \\ \times 0.205$	89.2	2.18	1.43	4.93×10^{4}
S2	Y123-bulk	Slab	5	$0.92 \times 0.22 \times$	90.3	1.79	1.46	9.34
	prepared from precursors		e a contra	0.22		ti e lag	ta e g	×10*
S3	Y123:8%Ag -	hollow	X - ray	0.87 imes 0.11 imes	90.1	2.62	1.62	1.16
	thick film	cylinder		0.03	· ·	en for in de Robert de		$\times 10^{6}$
S4	Bi2223-	Slab	5.5	0.72 imes 0.2 imes	99	3.13	1.43	3.17
	bulk ceramic			0.2	** -			$\times 10^{5}$

n dalah di berekara di kelik di berepan ngana ngang pertangkaran ang pangkaran dan di berekara (sa pang) panga Bana karabaran kelik di seria di baga na pertangkaran kelik di pangkaran kelik sa pertangkaran di baharan di ba Bana pertangkaran kelik di pertangkaran kelik sa pertangkaran kelik sa pertangkaran kelik sa pertangkaran sa pe



Fig.1 The penetration picture of an ac-magnetic field at various dc-fields H_{dc} (curve 1 and 3) and temperatures (curve1 and 2). The slope $tg\alpha$ is the J_{co} and depends on T and H_{dc} .

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Fig.3 The plot of χ''_1 as a function of H_{dc} for sample S1 at T = 77K. The solid lines are power law $(\chi'' \sim H_{dc})$ fitting curves of the experimental data. The β -values are indicated for each of curves.



Fig.4 The plot of $(\chi'_1 + 1)$ as a function of $(1 - T/T_c^{\omega})^n$ for sample S1 at $H_{dc} = 0$. The solid lines are power law $(\chi'_1 + 1) \sim (1 - T/T_c^{\omega})^{-n}$ fitting curves of the experimental data. The values of the factor n are also indicated.

Appendix

Generally the magnetization M(t) created by the external ac-magnetic field $H_{ac} = H_{osinwt}$ and averaged over the volume of a sample, which is in the critical state, is related to the distribution of the critical current density. For a cylindrical sample with radius R (or a slab with the square cross section $4R^2$) M versus $J_c(r)$ function takes the following simple form (in the SI unit system):

$$M = -\frac{1}{R^2} \int_o^R J_c(r) r^2 dr$$

Provided that the four assumptions mentioned in Section II are valid, $J_c(r)$ is a constant, which equals $\frac{dH}{dr}|_{r=R}$, and depends on temperature T and the external H_{dc} field, equation A.1 is rewritten as:

(A1) (A1)

$$M = -\frac{J_{co}(T,H_{dc})}{R^2} \int_o^R r^2 dr. \quad (A2)$$

The zero-field cooled sample being subjected to the ac-field H_{ac} has a virgin magnetization $M_{vir}(t)$ in the interval $0 \le wt \le \pi/2$ as follows:

$$M_{vir}(t) = \frac{-J_{co}R}{3} (1 - [1 - (1 - r_p/R)sin\omega t]^3)$$
 (A3)

where $r_p = H_o/J_{co}$. In the interval $\frac{\pi}{2} < wt \leq 3/2\pi$ the ac-field descends from $+H_o$ to $-H_o$, accordingly:

$$M_{cyc}^{-} = -\frac{J_{co}R}{3} (2[1 - 1/2(1 - r_p/R)(1 - \sin\omega t]^3 - (1 + r_p^3/R^3))). \quad (A4)$$

In the interval $\frac{3\pi}{2} < wt < 5/2\pi$ one has

$$M_{cre}^{+} = +\frac{J_{co}R}{2}(2[1-1/2(1-r_{p}/R)(1+sin\omega t)]^{3} - (1+r_{ac}^{3}/R^{3})).$$
(A5)

The real χ' and imaginary χ'' parts of the fundamental ac-susceptibility are determined as:

$$\chi' = \frac{2}{\pi H_o} \int_{\pi/2}^{3\pi/2} M_{cyc}^- \sin\omega t d\omega t \qquad (A6)$$

$$\chi'' = -\frac{2}{\pi H_o} \int_{\pi/2}^{3\pi/2} M_{cyc}^{-} \cos\omega t d\omega t \qquad (A7)$$

and finally one has:

$$\chi_{1}' = -1 + \frac{H_{o}}{J_{co}R} - \frac{5}{16} (\frac{H_{o}}{J_{co}R})^{2}$$
(A8)
$$\chi_{1}'' = \frac{4}{3\pi} \frac{H_{o}}{J_{co}R} - \frac{2}{\pi} (\frac{H_{o}}{J_{co}R})^{2}.$$
(A9)

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