Non-Universal Temperature Dependencies of the Low Frequency ac Magnetic Susceptibility in High $T_c$ Superconductors

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Abstract

The complex ac magnetic susceptibilities ($\chi_n = \chi'_n + i\chi''_n$) of high $T_c$ superconductors in absence of dc fields have been studied by numerically solving the non-linear diffusion equation for the magnetic flux, where the diffusivity is determined by the resistivity. In our approach the parallel resistor model between the creep and flux flow resistivities is used, so that the crossover between different flux dynamic processes (thermally activated flux flow, flux creep, flux flow) can naturally arise. For this reason we remark that, as the frequency increases, the presence of a different non linearity in different regions of the I-V characteristic determines non-universal temperature dependencies of the $\chi_n$, i.e. the $\chi_n$ are found to be not universal functions of a frequency and temperature dependent single parameter. Moreover, the actual frequency dependent behavior is also shown to be strictly related to the particular pinning model chosen for the simulations. Indeed, for large values of the reduced pinning potential ($U/KT \geq 220$) and for increasing frequency, a transition has been observed between dynamic regimes dominated by creep and flux flow processes. On the other hand, for smaller reduced pinning potentials, a transition from the thermally activated flux flow (Taff) to the flow regime occurs. In qualitative agreement with available experimental data but in contrast with previously used simpler models, the amplitude of the peak of the imaginary part of the first harmonic is shown to be frequency dependent. Moreover the frequency dependence of its peak temperature shows large discrepancies with approximated analytical predictions. Finally, the shape of the temperature dependencies of the higher harmonics are found to be strongly affected by the frequency.

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1 – INTRODUCTION

Dissipative effects in the mixed state of high T_c superconductors have been investigated by different dynamic techniques, such as "ac magnetic" susceptibility\(^1\), "ac transport" measurements, mechanical oscillator\(^2,3\). Furthermore, the low frequency complex susceptibility (\(\chi_n = \chi'_n + i\chi''_n\)) is also used to determine the critical current density \(^4\). It is known that \(\chi_n\) can be regarded as the Fourier coefficients of the steady magnetization cycles in the presence of an external oscillating magnetic field, being such coefficients determined by the magnetic flux entering and leaving the sample. Therefore, it is necessary to study the non-linear diffusion-like equation\(^5\) which governs the spatial-temporal evolution of the local magnetic field \(B\), where the role of the flux diffusivity is played by the resistivity \(\rho\), which is a function of temperature \((T)\), local field \((B)\) and local current density \((J)\). In such description the various regimes of flux dynamics are introduced through the I-V characteristic, which in turn depends on the different pinning mechanisms operating in the material. As a matter of fact, considerable efforts have been devoted to the development of theoretical models for the description of flux pinning and dynamics in high T_c materials: from the Kim - Anderson\(^6\) to the novel vortex-glass\(^7\)/collective pinning models\(^8,9\). The common feature of such models is the highly non-linear I-V characteristics.

The general non-linear diffusion problem cannot be solved analytically. In longitudinal geometry (long slab or cylinders) analytical solutions are available for:

a) the linear limit, corresponding to the regimes of thermally activated flux flow\(^10\) and flux flow when the magnetic field dependence is neglected;

b) highly non-linear (stepwise) I-V characteristics as in the Bean model\(^11\), where the solution is described by the critical state which should be recovered in the zero frequency limit.

In transverse geometry (thin platelets or films in a perpendicular magnetic field) recent analytical results have extended the Bean model to thin strips\(^12\), disks and squares\(^13-15\), accounting also for the finite thickness of specimen\(^16\).

In the presence of ac fields, a discussion of the frequency and amplitude dependence of the threshold between a linear and non-linear response has been reported for the vortex liquid and vortex glass states\(^17,18\).

Some of the experimental features of the temperature dependence of \(\chi'_n\) and \(\chi''_n\) have been successfully explained by the critical state model and its generalizations\(^19-21\). However, the observed frequency dependence of the fundamental\(^22-25\) and higher harmonics\(^26-29\) cannot be described within the framework of the critical state. As a consequence, the simultaneous presence of hysteretic and dynamic losses has to be included in the model description. Numerical methods have therefore to be applied for solving the non-linear magnetic diffusion problem. Within such approach, the time evolution of flux profiles and magnetization curves have been calculated by many authors\(^30,31\). Moreover, results have been also reported in literature for the complex rf magnetic permeability in a parallel static magnetic field\(^32\) and for the ac susceptibility in presence of a dc bias magnetic field, calculated in the flux creep regime\(^33\). Recently, the ac response of thin superconductors has been studied in the flux creep regime by numerically solving the integral equation which describe the flux diffusion in the transverse geometry\(^34\). Some authors suggested\(^21,35\) the possibility of an universal behavior described by the single scaling parameter \(\delta(\omega,T)\), i.e. the effective penetration length, which is
related to a frequency dependent critical current. In this approach the susceptibilities can be written as: \( \chi_n = f_n(\delta(\omega, T)) \). Such behavior has been also considered\(^ {26} \) on the basis of a comparison between the experimental temperature dependence of the third harmonic susceptibility and a semi-analytical approach.

To our knowledge, while the frequency dependence of the peak temperature has been extensively studied\(^ {22,36-38} \), an issue that has not received a careful inspection is the frequency dependence of the peak amplitude \( \chi_1'' \), which has been reported in a few experimental works\(^ {39-44} \). Furthermore, also the theoretical description of the frequency dependence of the higher harmonics\(^ {26,27} \) has not been fully developed.

In this paper we shall focus on the temperature and frequency dependence of the ac susceptibility \( \chi_n(T) \) (fundamental and its harmonics) without dc bias magnetic fields, starting from the numerical solution of the non-linear magnetic diffusion equation. As a novelty with respect to previous literature works, the diffusivity has been described in terms of a "parallel resistor model"\(^ {45-48} \), incorporating both flux creep and flux flow resistivities. In such way different non-linear behaviors naturally arise in different regions of the I-V characteristic: at very low current values the "Taff" regime corresponds to a linear behavior, while for currents close to the critical one an exponential increase of the voltage appears, leading again to a linear behavior for \( J > J_c \) in the flux flow regime. In general, the approach usually reported in literature\(^ {49,50} \) is to consider the same non-linear behavior for the overall I-V characteristic (for instance a power law, \( V \propto I^n \), with \( n > 1 \)). On the contrary, our approach accounts for changes of the non-linear behavior produced by variations of the currents induced by the ac magnetic driving field. As a consequence a non-universal behavior appears, especially in the general shape of the temperature dependence of higher harmonics.

The paper is organized as follows. In Sec. II the non-linear diffusion problem is formulated in terms of a partial differential equation, together with the parallel resistor model for the I-V characteristics. To study in some detail the effects of thermally activated processes in different cases, we have chosen different temperature functional dependencies for the pinning potential, \( U_p(T) \), and the critical current density, \( J_c(T) \), related to particular pinning models. Local magnetic field profiles, magnetization cycles and \( \chi_n(T) \) are discussed in Sec.III. Moreover, a comparison of numerical results with available experimental data and analytical approximated predictions is also presented. Finally, Sec.IV is devoted to summary and conclusions.

2 – THE NON-LINEAR DIFFUSION EQUATION

We consider a hard superconductor in the geometry of an homogeneous-infinite slab with thickness \( 2d \), in presence of an ac external magnetic field, \( B_{\text{ext}}(t) = B_0 \sin(2\pi vt) \), applied parallel to the sample surface. In such a one-dimensional case the non-linear diffusion equation for the local magnetic field \( B \) inside the sample is reduced to:

\[
\frac{\partial B}{\partial t} = \frac{\partial}{\partial x} \left[ \rho(B, J) \frac{\partial B}{\partial x} \frac{\mu_0}{\partial x} \right]
\]
where $\rho(B,J)$ is the resistivity which, in absence of a dc magnetic field, strongly depends on the local field $B$\textsuperscript{[17]}). Such resistivity is taken as the parallel ($\rho_{\text{par}}$), between the "flux creep" ($\rho_{\text{cr}}$\textsuperscript{[51,52]} and the "flux flow" ($\rho_{\text{ff}}$\textsuperscript{[53]} resistivities:

$$\frac{1}{\rho(B,J)} = \frac{1}{\rho_{\text{par}}} = \frac{1}{\rho_{\text{cr}}} + \frac{1}{\rho_{\text{ff}}}$$

$$\rho_{\text{cr}}(I) = 2\rho_c\left(\frac{J_c(t)}{J}\right)e^{-\frac{U_p(t)}{kT}} \sinh\left(\frac{JU_p(t)}{J_c(t)kT}\right)$$

$$\rho_{\text{ff}} = \rho_n(T) \frac{B}{B_{c2}(t)}$$

where $t = T/T_c$ is the reduced temperature, $U_p(t)$ is the pinning potential, $J_c(t)$ the critical current density, $J$ the current density deduced by the local magnetic profile and $B_{c2}(t)$ is the upper critical field written as\textsuperscript{[32]}:

$$B_{c2}(t) = B_{c2}(0) \left(1 - t^2\right) \left(1 + t^2\right)$$

The prefactor\textsuperscript{[54]}, $\rho_c$, in eq.3 is determined by the condition $\rho_{\text{cr}}(J_c) = \rho_{\text{ff}}$, so that $\rho_c = \rho_{\text{ff}}$.

For $JU_p/J_cK_B T << 1$, the "Taff" resistivity limit ($\rho_{\text{Tf}}$) of $\rho_{\text{cr}}(I)$ is recovered:

$$\rho_{\text{Tf}} = 2\rho_n\left(\frac{B}{B_{c2}(t)}\right) \left(\frac{U_p(t)}{KT}\right) e^{-\frac{U_p(t)}{kT}}$$

Within this approach the fluctuations effects in the resistivity are neglected; indeed, since $U_p(t)$ and $J_c(t)$ vanish as $T \to T_c$, the normal state value of the resistivity is recovered in such a limit. The resistivity decrease due to fluctuations should lead only to a smearing of the temperature dependencies near $T_c$.

Moreover, to identify the dominant dissipative process in different temperature and/or frequency regions, the diffusion equation has been also studied using some limits of eq.2 i.e. $\rho_{\text{Tf}}, \rho_{\text{cr}}$ or $\rho_{\text{ff}}$.

The description of the total resistivity by means of a parallel resistor model is based on the assumption that dissipation processes are a sequence of independent flux creep and flux flow events\textsuperscript{[46]}. Within such approach the different regimes of flux motion ("Taff", "Creep", "Flow") are smoothly connected in the E-J characteristic. As the increase of the frequency of the magnetization cycle corresponds to an increase of the electric field, the crossover from a weak ("Creep") to a strong ("Flow") frequency dependence, is recovered in a natural way.

Equation (1) is numerically solved by means of the FORTRAN NAG\textsuperscript{[55]} routines, where adimensional variables have been introduced: $\tilde{x} = \frac{x}{d}$; $\tilde{t} = \frac{t}{t_0}$; $t_0 = \frac{\mu_0 d^2}{\rho}$; $\rho' = 1\mu\Omega m$.

Moreover, the normalized frequency $v^*$ is related to the frequency $v$ of the applied magnetic
field by the following: \( \nu^* = \frac{\mu_0 d^2 \nu}{\rho^*} \) where \( \nu \) is the frequency in Hertz and \( \mu_0 = 4\pi \times 10^{-7} \text{ N/} \text{A}^2 \).

The boundary and initial conditions are respectively:

\[
B(1, t) = B(-1, t) = B_0 \sin(2\pi \nu^* t)
\]

\[
B(\bar{x}, 0) = 0
\]  

(7)

where \( B_0 \) and \( \nu^* \) are the amplitude and frequency of the magnetic field respectively.

The algorithm computes the time evolution of the local field profile by integrating the discrete version of eq.(1) using the method of lines and Gear's method for a fixed number of spatial meshes (typically 100). The periodic steady magnetization loops \( M(B) \) are calculated from the difference between \( \langle B(t) \rangle \), that is the volume average of the profile \( B(\bar{x}, \bar{t}) \), and the instantaneous value of the applied magnetic field \( B_{ext}(\bar{t}) \). In particular, the calculated magnetization loop is considered as a steady state when the difference \( \delta B \equiv \langle B(\bar{t}_{n+1}) \rangle - \langle B(\bar{t}_n) \rangle \)

is lower than \( 10^{-3} B_o \), being \( \bar{t}_n = \left( \frac{1}{\nu^*} \right) (n + \frac{1}{4}) \) and \( n \) an integer. The complex susceptibilities \( \chi'_n \) and \( \chi''_n \) are then calculated as:

\[
\chi'_n = \frac{1}{\pi B_0} \int_0^{2\pi} M(\omega^* \bar{t}) \sin(\omega^* \bar{t}) d(\omega^* \bar{t}) \quad (8a)
\]

\[
\chi''_n = \frac{1}{\pi B_0} \int_0^{2\pi} M(\omega^* \bar{t}) \cos(\omega^* \bar{t}) d(\omega^* \bar{t}) \quad (8b)
\]

where \( \omega^* = 2\pi \nu^* \).

In order to account for the temperature dependence of the susceptibility, the temperature dependencies of \( U_p(t) \) and \( J_c(t) \) have to be specified. A natural choice is to rely on pinning models invoked in the literature for explaining experimental data on irreversible magnetic properties. In particular, three functional temperature dependencies have been considered, corresponding to different \( \frac{dU_p(T)}{dT} \) and \( \frac{dJ_c(T)}{dT} \) in the limit \( T \to T_c \).

In the first one \( P^{9,56} \), we have:

\[
U_p(B,t) = U_0 \left( 1 - t^4 \right) \quad (9a)
\]

\[
J_c(t) = J_0 \left( \frac{1-t^2}{1+t^2} \right)^{3/2} \quad (9b)
\]
where $U_0 \equiv U_p(T=0)$. Such temperature dependencies of $U_p$ and $J_c$ arise within the collective pinning model, where vortices are supposed to be pinned by randomly distributed weak pinning centers, possibly related to local variations of the electronic mean free path. Such a model has been used to describe the behavior of stoichiometric yttrium-based thin films.

In the second form (II), assuming the Ginzburg-Landau temperature dependencies for the thermodynamical critical magnetic field $B_c(t)$ (eq.5) and the coherence length

$$\xi(t) = \frac{\xi_0 (1 + t^2)^{1/2}}{(1 - t^2)^{1/2}},$$

we have the following:

$$U_p(B,t) = U_0 \frac{(1 - t^2)^{1/2}}{(1 + t^2)^{1/2}} \quad (10a)$$

$$J_c(t) = J_0 \frac{1 - t^2}{1 + t^2} \quad (10b)$$

Indeed in such model, the pinning potential $U_p$ is estimated\(^{57,58}\) as the condensation energy density $H_{c2}(t)$ times a volume $\xi^3$. In this case it has been assumed that a fluxoid intersects a small pinning site of volume $\xi^3$ (core interaction). The elementary pinning force is given by $f_p = U_p / \xi$ and the macroscopic force $F_p$ results from a procedure of direct summation of elementary forces $f_p^{51,59}$.

In the last pinning model (III), a fast decrease of $U_p(t)$ with the temperature is assumed, whereas a decrease of $J_c(t)$ intermediate between the first two cases has been chosen:

$$U_p(B,t) = U_0 \frac{(1 - t^2)^{3/2}}{(1 + t^2)^{1/2}} \quad (11a)$$

$$J_c(t) = J_0 \left(1 - t^2\right)^2 \quad (11b)$$

Similar dependencies have been introduced to account for the existence of the "giant flux creep"\(^{60-61}\), taking the pinning potential as $U_p = H_{c2}^2(t) * (a_0^2 \xi)$, where $a_0^2 = \phi_0 / B$ is the area of a unit cell of the flux lines lattice\(^{59}\). The macroscopic force $F_p$ results also in this case from a direct summation procedure of elementary pinning forces, $f_p = U_p / \lambda$, where $\lambda$ is the London penetration depth. The temperature dependence of $U_p$ and $J_c$ in Eq. 10a,b stems from the temperature dependence of $H_{c2}(t)$, $\xi(t)$ and $\lambda(t)$. In a first approximation, a weak temperature dependence of $\kappa$, $\kappa = \kappa_0 (1 + t^2)^{-\nu}$ and the Gorter-Casimir temperature dependence of $\lambda$ have been assumed. It should be noted however, that for high $T_c$ superconductors experimental evidence exists\(^{62}\) in favour of temperature dependencies other than the "two-fluid model". Nevertheless, experimental data reported by different groups are not entirely consistent with a unique temperature dependence of $\lambda$; therefore the "two-fluid model" has to be regarded as a useful starting point. The comparison of the different temperature dependencies of the pinning potentials (normalized to the zero temperature value), reveals that:
1) \( U_p(I) \gg U_p(III) \) for all the reduced temperatures;
2) \( U_p(I) > U_p(II) \) up to \( t=0.9 \), while near \( T_c \) the opposite behavior occurs;
3) \( U_p(I) \) is quite constant up to \( t=0.6 \); for \( t > 0.6 \) it decreases very quickly if compared to the other cases;
4) \( U_p(III) \) describes a fast decrease of the pinning potential with temperature.

For \( J_c \) we have: \( J_c(I) < J_c(III) < J_c(II) \) at any temperature.

3 – RESULTS AND DISCUSSION
3.1 – Magnetic field profiles and stationary magnetization cycles

The material parameters used for the simulations pertain to an YBCO slab of thickness \( 2d = 2\text{cm} \), \( T_c = 92.3\text{K} \), \( B_c(0) = 112\text{T} \), \( U_0(0)/K = 2*10^4\text{K} \), \( J_c(0) = 10^{10}\text{A/m}^2 \). Neglecting fluctuations around \( T_c \), the normal state resistivity is:

\[ \rho_n(T) = \rho_0\left(1 + \alpha\frac{T}{T_0}\right), \]

where \( \alpha = 5.5*10^{-3}\text{K}^{-1} \), \( \rho_0(T_0) = 2 * 10^{-6}\Omega\text{m} \) and \( \hat{T} = T - T_0 \).

Our analysis has been restricted to the case a small applied ac magnetic field in absence of a superimposed large dc component; therefore the magnetic field dependencies of \( J_c \) and \( U_p \) are very weak and they can be neglected. Moreover, any explicit spatial dependencies of the pinning parameters have not been considered. In any case, being thermally activated processes negligible at low temperature, we have firstly verified that at low temperature and for low frequencies, the solutions of the diffusion equation reproduce the critical state picture corresponding to an effective critical current density \( J_c^* \) close but lower than \( J_c \).

As suggested by several authors\(^{35} \), the increase of the frequency results in the increase of the electrical field, corresponding to higher values of \( J_c^* \). Nevertheless, at higher frequencies, significant deviations from the critical state description are present due to the flux flow component of the parallel model, in such way that, at \( \nu^* = 1 \), the parallel result is practically equal to the flux flow one.

The field profiles for \( \rho_{par} \) at different frequencies are reported in Fig.1, where the external field is equal to \( B_0 \) and \( U_p/KT=260\text{K} \) and \( J_c = 2.4\text{KAm}^{-2} \) corresponding to the pinning model I at \( T = 91.5\text{K} \). The profiles corresponding to the critical state, to \( \rho_{cr} \) at \( \nu^* = 10^{-4} \) and to \( \rho_{ff} \) at \( \nu^* = 10^{-1} \) are also plotted for comparison.

At the lowest frequency (\( \nu^* = 10^{-4} \)) the creep profile is practically identical to the parallel one. For frequencies larger than \( \nu^* = 10^{-1} \) the field profiles determined by the "parallel resistivity" are practically identical to the flux flow ones. It should be noted that the "parallel resistivity" gives at \( \nu^* = 10^{-4} \) a constant field gradient which apparently suggests a behavior similar to the critical state with a value of \( J_c^* < J_c \). However, the inspection of profiles at different times of the cycle shows, also in such a case, large differences from the critical state determined by \( J_c^* \).
FIG.1 - Magnetic field profiles at T=91.5K for $B_0=2\text{mT}$ and $\omega t=\pi/2$, evaluated for different normalized frequencies, $\nu^*=\mu_0d^2\nu/\rho^*$, in the case of the parallel resistivity, $\rho_{\text{par}}$, and collective pinning model I. The parallel is calculated between creep and flow resistivity, i.e. $\rho_{\text{par}}=\rho_{\text{c}}/\rho_{\text{ff}}$. The value of $\rho^*$ is $1\mu\Omega\text{m}$. Symbols refer to: (\textcolor{red}{$\Delta$})$\nu^*=10^{-4}$; (\textcolor{blue}{$\square$})$\nu^*=10^{-3}$; (\textcolor{green}{$\triangle$})$\nu^*=10^{-2}$; (\textcolor{black}{$\times$})$\nu^*=10^{-1}$, (\textcolor{purple}{$\circ$})$\nu^*=1$.

As the transition temperature ($T_c=92.3\text{K}$) is approached, the contribution of the flux flow component has a larger weight even at lower frequencies. In Fig.2a we report the frequency dependence of the magnetization cycles at 92K, whereas Fig.2b shows the magnetization cycles, computed at $\nu^*=10^{-4}$ for different resistivities with the pinning model I. The critical state result is plotted too, but just for reference purpose. As a result, the field diffusion at low frequency is determined, close to $T_c$, by the flux flow resistivity.

We wish to remark the unexpected and somewhat surprising shape of the cycles, which seems to be the result of a critical state with a magnetic field dependence of critical current density. Such result confirms what previously reported by other authors 31. We ascribe such an anomalous behavior mainly to the field dependence of the flux flow resistivity, i.e. $\rho_{\text{ff}}=\rho_{\text{n}}B/B_{c2}(t)$: physically, the collapse of the cycle for increasing absolute values of the applied field can be explained by the increasing of the resistivity with the field. By the way, the field dependence is present also in the creep resistivity through the coefficient $\rho_{\text{c}}$. From the analysis of the field profiles it comes out that the origin of the bump corresponding to $B_{\text{ext}}=0.26\text{mT}$ in Fig.2b, is related to the presence of regions of the sample in which the field and the corresponding resistivity are close to zero, so that a vanishing diffusion coefficient determines larger difference between the internal and the external field. However, the presence of the anomalous bump in the magnetization cycle appears only at low frequency and close to $T_c$. 
FIG. 2 - Magnetization cycles calculated at $T=92K$ and $B_0=2mT$ in the pinning model I case: (a) with $\rho_{\text{par}}$ for different normalized frequencies $\nu^*$: ($\cdots\Delta\cdots$) $10^{-4}$; ($\cdots\square\cdots$) $10^{-3}$; ($\cdots\bullet\cdots$) $10^{-2}$; ($\cdots\times\cdots$) $10^{-1}$; ($\bullet\cdots$) $1$; (b) for $\nu^*=10^{-4}$ with different resistivities $\rho$: ($\cdots\Delta\cdots$)$\rho_{\text{par}}$; ($\cdots\square\cdots$)$\rho_{\text{cr}}$; ($\bullet\cdots$)$\rho_{\text{ff}}$. The Bean prediction is also shown for comparison (solid line).

3.2 – Susceptibilities
3.2.1 – Fundamental harmonic

In this section we analyze the temperature dependence of the first harmonic ac susceptibility ($\chi'_1, \chi''_1$) at different frequencies by comparing the behaviors predicted by the three pinning models. The "parallel" resistivity, $\rho_{\text{par}}$, will be used in the numerical analysis as well as the resistivities describing single loss regimes ("Taff" resistivity, $\rho_{\text{Tf}}$; "creep" resistivity, $\rho_{\text{cr}}$; "flux flow" resistivity, $\rho_{\text{ff}}$). To clarify the notation, we remark that the symbols $\chi'_n (\alpha;\beta)$ and $\chi''_n (\alpha;\beta)$ denote the susceptibilities, with $\alpha = \text{I, II, III}$, (pertaining to the pinning models) and $\beta = \text{par, Tf, cr, ff}$ (pertaining to the resistivity loss mechanism).

In general, from the analysis of magnetic field profiles and magnetization cycles it is expected that at low frequency and low temperature, the behavior of susceptibilities should be critical state-like. On the contrary, as the frequency or/and the temperature increases, the
diffusion of the magnetic field due to $\rho_{TF}$ or $\rho_{cr}$ and $\rho_{ff}$ becomes more and more relevant. Moreover, near $T_c$ or at higher frequencies the $\rho_{ff}$ dominates the diffusion process. Since the relevance of thermally activated processes is strictly related to the value of the ratio $U/KT$, we expect that in the model III at any temperature, the influence on the susceptibilities of diffusive phenomena should be larger than for the other two models. The model II should exhibit a larger influence of activation processes up to 85K if compared to the model I. However, an opposite behavior is expected between 85K and the transition temperature.

Our simulations have been performed for two values of the ac magnetic field amplitude $B_0=2\text{mT}, 20\text{mT}$. The 2mT results are closer to accessible experimental data, whereas, due to the well known amplitude dependence of the susceptibilities, the 20mT simulations are intended to give a better understanding of the behavior at lower temperatures. The temperature dependencies of the ac susceptibilities in the model I, $\chi'_1(I;\text{par})$ and $\chi''_1(I;\text{par})$ are shown in Fig.3a,b at different frequencies for $B_0=20\text{mT}$. The critical state predictions are also reported in the same plots in order to stress the relevance of thermally activated processes.

The main features can be summarized in the following:

- at low frequency the calculated temperature behavior is critical-state like; however the zero frequency limit of the ac susceptibility is different from the critical state prediction and, in particular, the peak temperatures $T_p$ is lower than the Bean predicted value;
- the maxima of $\chi''_1(I;\text{par})$ are always higher than the critical state value ($\chi''_1(\text{Bean})_{\text{max}}\approx 0.24$);
- both the peak temperatures and amplitudes increase with frequency.

At the highest frequency considered, the peak amplitude is $\chi''_1(I;\text{par})_{\text{max}}\approx 0.5$ whereas for a constant resistance the value is $\chi''_1(\rho)_{\text{max}}\approx 0.42$. As previously shown in Sec.3.1, the magnetic flux diffusion at high frequency is essentially dominated by the flux flow; therefore the explanation of the large value of the peak amplitude should be found in the dependence of $\rho_{ff}$ on the local magnetic field.

All the features found for 20mT appear for 2mT at higher temperatures, where the thermally activated processes become more relevant and enhance the differences respect to the critical state behavior.

As far as the temperature dependence of $\chi'_1(I;\text{par})$ is concerned, the transition is sharper at lower amplitudes $B_0$ and higher frequencies. The analysis of $\chi'_1(I;\text{par})$ as a function of frequency at fixed temperature reveals that the relative variation with frequency (i.e. $\frac{d\chi'_1(I;\text{par})}{d\nu}$) is much higher near $T_c$. Indeed, near $T_c$, the shielding effects are only due to the frequency induced dissipative currents, which vanish in dc conditions; however, a significant shielding exists at low temperature also in the zero frequency limit, due to the critical current. Moreover, a distinctive feature with $\frac{d\chi'_1(I;\text{par})}{dt^2}<0$ appears at $T=90.6\text{K}$ and $\nu^*=10^{-2}$ (Fig.3a): such behavior is related to the crossover between regimes dominated by "creep" and "flux flow" phenomena.
FIG. 3 - First harmonic susceptibilities vs temperature for $B_0=20\text{mT}$, calculated for different normalized frequencies $v^*$ in the case of $\rho_{\text{par}}$ and pinning model I: (a) real parts, ($\chi'_{1}$); (b) imaginary parts, ($\chi''_{1}$). Symbols refer to: ($\bullet\square\bullet\}$ $v^*=10^{-4}$; ($\bullet\blacktriangle\bullet\}$ $v^*=10^{-3}$; ($\blacktriangle\blacktriangle\blacktriangle\}$ $v^*=10^{-2}$; ($\blacktriangle\textbullet\blacktriangle\}$ $v^*=10^{-1}$; ($\bullet\bullet\bullet\}$ $v^*=1$. The Bean prediction is also shown (solid line).

Indeed, at $v^*=10^{-2}$ and for $T<90\text{K}$, the $\chi''_{1}(I;\text{par})$ curve merges with the pure $\rho_{\text{cr}}$ result, which appears closer to the critical state prediction in the whole temperature range. On the contrary for $T>90.7\text{K}$, the $\rho_{\text{ff}}$ contribution becomes more and more relevant.

At frequencies lower than $v^*=10^{-3}$ the behavior of $\chi''_{1}(I;\text{par})$ is described by $\rho_{\text{cr}}$, whereas for frequencies above $v^*=10^{-1}$ $\chi''_{1}(I;\text{par})$ coincides with $\chi''_{1}(\text{ff})$, in the whole temperature range investigated. By the way, in the model I, the $\rho_{\text{ff}}$ resistivity is ruled out in describing the temperature dependence of $\chi''_{1}$.

A similar analysis has been performed for the models II and III (not shown for sake of brevity). In both cases the susceptibilities in different resistivity regimes have been compared to understand the dominant resistivity regime.

In the model II, for $t>0.3$, $J_c$ is higher compared to the model I, particularly near $T_c$. For this reason at each frequency the peak of $\chi''_{1}(I;\text{par})$ is sharper in the model II and the transition
width of $\chi'_1(\Pi;\text{par})$ is narrower, all such features developing in a very narrow temperature range around $T_c$.

By the way the main features obtained are:

a) at low frequencies, i.e. $\nu^* = 10^{-3} - 10^{-4}$, the behavior of $\chi''_1(\Pi;\text{par})$ above $T_p$ is essentially determined by the "$\rho_{Tf}$". To strengthen such a statement, the "Taff condition", $J/J_c(t) \times U/(K T) << 1$, has been also checked. For $T < T_p$, the behavior of $\chi''_1(\Pi;\text{par})$ is essentially the same of $\chi''_1(\Pi;\text{cr})$;

b) at intermediate frequency $\nu^* = 10^{-2} - 10^{-1}$ the $\chi''_1(\Pi;\text{par})$ merges in $\chi''_1(\Pi;\text{cr})$ for all temperatures;

c) for $\nu^* = 1$ near $T_c (\pm 0.1 \text{K})$ the "flux flow resistivity " gives a small contribution to $\chi''_1(\Pi;\text{par})$.

In the pinning model $\Pi$, the temperature dependence of $J_c$ is intermediate between models $I$ and $\Pi$; on the contrary, the pinning potential vanishes in a quicker fashion, so that the giant creep phenomenon is expected to be more relevant also at lower temperatures ($T \gtrsim 80 \text{K}$), inducing large differences between the critical state and the actual diffusion of the magnetic field. The main features of the $\chi''_1(\Pi)$ temperature dependencies are:

a) for $\nu^* = 10^{-4}$, the temperature dependencies of $\chi''_1(\Pi;\text{par})$, $\chi''_1(\Pi;\text{cr})$ and $\chi''_1(\Pi;\text{Tf})$ are very similar;

b) differences between $\chi''_1(\Pi;\text{par})$, and $\chi''_1(\Pi;\text{Tf})$ (which is close to $\chi''_1(\Pi;\text{cr})$) appear for $10^{-3} \leq \nu^* \leq 10^{-2}$: such differences are amplified just above the peak temperatures and the behavior could be ascribed to a significant $\rho_{Tf}$ contribution;

c) the differences between $\chi''_1(\Pi;\text{par})$ and $\chi''_1(\Pi;\text{cr})$ seems to be negligible for $10^{-1} \geq \nu^* \geq 1$.

It should be noted that an anomalous behaviors occurs at low frequencies in a few tenth below $T_c$: as the temperature increases, a decreasing of $\chi'_1(\Pi;\text{par})$ as well as an increase of $\chi''_1(\Pi;\text{par})$ occur. Such results are due to the unphysical non-monotonic increase with temperature of the $\rho_{Tf}$ resistivity in the model $\Pi$, showing a maximum, $\rho_{Tf}^\text{Max} (91.5 \text{K}) \sim 3 \times 10^{-2} \Omega \text{m}$. For such reason the results obtained at $T > 91.5 \text{K}$, where the "Taff" is relevant, have to be rejected.

The frequency dependence of the $\chi''_1$ peak amplitude and peak temperature ($T_p$) for the models $I, II, \Pi$ are summarized in Figs. 4 and 5.
FIG. 4 - Calculated peak amplitude $\chi''(T_p)$, vs frequency for different pinning models in the $\rho_{\text{par}}$ case. The symbols are ( ----- ) I for collective, ( - - - - - - ) II for core interactions, ( - - - - ) III for giant flux creep. The parameter values are $U_0(0)/K = 2\times10^4\, \text{K}$, $J_c(0) = 10^{10}\, \text{A/m}^2$, $B_{c2}(0) = 112\, \text{T}$, $B_0 = 20\, \text{mT}$ is the amplitude of the ac applied magnetic field $B_{ac}$.

As far as the peak amplitude is concerned, its frequency variation is the first clear evidence of the absence of a universal behavior, which should otherwise lead to a constant peak value.

In the pinning model I, the low frequency behavior is close to the critical state with $\chi''(T_p) \approx 0.3$. On the contrary, as the frequency increases, the increasing contribution of the flux flow resistance leads to a smooth growth of $\chi''(T_p)$ up to values larger than 0.5. As previously stated, such large value is determined by the implicit non-linearity present in the magnetic field dependence of the flux flow resistivity.

By the way, in pinning model II such a high value of the peak of $\chi''(\text{II};\text{par})$ appears for each frequency; thus such feature is again related to a "magnetic field dependent" linear or a "weakly moderate non-linear" I-V characteristic, as in the case of the $\rho_{Tf}$ dominant resistivity. In such case however, as the frequency increases, the exit from the Taff condition towards a stronger non-linear behavior causes a reduction of the peak.

Finally, in the model III the overlap of the Taff and the flux flow resistivities induces an almost constant peak value $\approx 0.5$.

For these reasons, in the analysis of experimental data, the different frequency dependencies of the peak amplitude for the three models can give first indication of the actual dominant dynamic process.

Usually ac susceptibility data are often reported in arbitrary unit, making a quantitative comparison between experimental and numerical results very difficult to perform. Therefore, the analysis has to be necessarily restricted to a qualitative discussion of the trend of data with frequency and/or temperature. Furthermore, a quantitative comparison with published experimental data in absence of an high dc magnetic field is not achievable. Nevertheless the increase of the peak amplitude of $\chi''(T_p)$ with the frequency is evident in some papers $^{39-41}$.

As far as the peak temperature is concerned, a general result common to the critical state
descriptions, is its relation to the value of the critical current density $J_c$: the peak temperature is closer and closer to $T_c$ as $J_c$ increases.

In the analysis of the frequency dependence of peak temperature, using an approach similar to that of the present paper, many authors suggested an Arrhenius-like relationship between the frequency and the inverse of $T_p$:

$$\frac{1}{T_p} = C - \frac{K}{U_p(T_p)} \ln(f)$$  \hspace{1cm} (12)

where $C$ is a constant. In a very small frequency range, corresponding to very small $T_p$ variations, $U_p$ can be considered constant, so that the linear dependence of $1/T_p$ on $\ln(f)$ has been used to derive the value of the pinning potential. For larger frequency ranges, as $T_p$ increases with the frequency, $U_p$ decreases, so that eq.12 predicts a slope increasing with the frequency. On the contrary, the results of the numerical solutions of the diffusion equation, reported in Figs.5a,b for the three pinning models, show a monotonic increasing slope only for the model III.

As expected from eq.12, the comparison among the numerical frequency $T_p$ dependencies shows that larger slopes are present in pinning models with lower pinning potentials, so that the slope analysis can give a qualitative indication of the actual pinning model. Conversely, a quantitative proportionality of the slope with $1/U_p$, predicted by eq.12, is not verified. In fact in figs.5a,b for the three models the linear dependencies predicted by the eq.12 (determined by the value of $U_p$ corresponding to $T_p$ (0.8Hz) ) are depicted by full lines intersecting the corresponding numerical results. Large slope differences with the numerical solutions clearly appear. Finally in fig.5b the numerical results are also compared with the analytical frequency dependence of $T_p$ derived from the condition yielding the $\chi''$ peak for a pure resistance $\rho$:

$$d \left( \frac{\omega \mu_0}{\rho} \right)^{1/2} = 1.2 \sqrt{2}.$$  

In the case of the Taff resistivity, the peak frequency $f_T$ is:

$$f_T = f(T_p) \cdot e^{\frac{U_p(T_p)}{KT_p}}$$  \hspace{1cm} (13)

where $f(T_p) = 2.88 \frac{\rho_o}{\pi d^2 u_o} B_{c2}(T_p) \frac{U_p(T_p)}{KT_p}$

For magnetic field $B$ values comparable to the amplitude of the ac field, the agreement between the numerical solutions and eq.13 is reasonable. Considering an effective value of $B$ equal to $1/2$ of the maximum amplitude $B_0$, the agreement is very good.

For such reason the large difference between eq.12 and eq.13, shown in fig. 5, implies that the factor $f(T_p)$ cannot be neglected, since it strongly changes the slope of the $1/T_p$ curve. Following the above discussion, a qualitative comparison with experimental data can be performed: fig.5a shows data referring to $Y_1Ba_2Cu_3O_7$ grown by melt-powder-melt-growth method and $YBaCuO$ single crystal. As a matter of fact, only few published measurements...
have been taken in absence of large dc magnetic fields. In spite of the somewhat arbitrary choice of the parameters \( U_0(0)/K = 2 \times 10^4 \text{K}, \quad J_c(0) = 10^{10} \text{A/m}^2, \quad d=1 \text{cm}, \quad \rho_c=\rho_{\text{ff}} \) used in our numerical analysis, the behavior of the melt-grown sample appears to be similar to the II model dependence. Due to the error bars appearing in published data, a best fit procedure is meaningless, however in order to fit the melt-grown sample data with the other two models, too large values of \( J_c(0) \) and \( U_0(0) \) should be supposed.

For the single crystal the increase of the slope with the frequency appears to be close to the collective pinning prediction (model I). However, also in this case, the uncertainty of the actual sample size and the presence of non-negligible geometrical effects gives only a qualitative meaning to the fit procedure. By assuming the size of the single crystal equal to \( d \approx 1 \text{mm} \) (1/10 of the sample thickness used in our numerical simulations), a two order of magnitude frequency shift should be expected.

FIG. 5 - a) Calculated \( T_c/T_p(\chi''_1) \), vs frequency for different pinning models, the symbols are: \((-\cdots-)\ \text{I}, \ (-\cdots-\cdots-)\ \text{II}, \ (-\cdots-)\ \text{III}.\) The three short full lines represent the Arrhenius-like dependencies (eq.12), obtained by the \( U_p(T_p) \) values corresponding to intersecting points with the numerical results in the respective pinning model. The literature data for YBCO single crystal, \((\square)\) and YBCO semi-fused data \((\blacktriangle)\) are also shown. b) a detail of the \( T_c/T_p(\chi''_1) \) vs \( f(\text{Hz}) \) for model II \((-\cdots-)\), where \((\times)\) are \( T_p(\chi''_1) \) values calculated with "Taff resistivity", equation (6). The solid line present the Arrhenius-like relationship.
As reported in fig.5a a qualitative agreement appears. Once again, in order to fit the single crystal data with the model III, too large values of $J_c(0)$ and $U_0(0)$ should be supposed. Moreover the single crystal data cannot be fitted with the model II since, as shown in fig.5b, this model predicts a decrease of the slope with the frequency increase in opposition to the experimental results and pinning model I. In any case, our key result is the large difference between numerical solutions and the slope of the $1/T_p$ curve (eq.12); therefore, the derivation of the pinning potential from the frequency dependence of the first harmonic susceptibility should be exploited very carefully.

### 3.2.2 – Third harmonic

As expected, in absence of a dc magnetic field, even harmonics are found to be equal to zero. The analysis of profiles, cycles and first harmonic has shown that for the pinning models considered, the magnetic flux diffusion is in many cases dominated by the $\rho_{ff}$ or $\rho_{Tf}$ resistivity: in such linear cases, (i.e. a linear I-V characteristic) higher harmonics\(^1\) should be absent. However, the magnetic field dependence of the two resistivities, $\rho_{cr}$ and $\rho_{ff}$ can induce, even in presence of an ac field only, a non-linear behavior in the diffusion equation. Therefore, in this case too, higher harmonic components are present in the susceptibility. As an example, $\chi'_3(ff)$ and $\chi''_3(ff)$ calculated for $\rho_{ff}$ are reported in Figs. 6a,b, whereas $\chi'_3(I;Tf)$ and $\chi''_3(I;Tf)$ calculated for $\rho_{Tf}$ are shown in Figs. 7a,b for the pinning model I. The real parts display negative values and bell-like shapes, with peak temperatures increasing as the frequency increases; at the highest frequency, $n^*=1$, $\chi'_3(I;Tf)$ and $\chi'_3(ff)$ are negligible except that in a few tenths of a Kelvin around $T_c$. On the other hand, the imaginary parts show an oscillatory behavior: it displays positive values on approaching $T_c$ and negative values at lower temperatures. As the frequency increases, such behavior of $\chi''_3(I;Tf)$ and $\chi''_3(ff)$ occurs closer and closer to $T_c$.

Following the scheme of the previous section, we start the study of odd harmonics in the pinning model I using the parallel resistivity.
FIG. 6 - Third harmonic susceptibilities vs temperature for $B_0 = 20$ mT, evaluated with the flux flow resistivity, $\rho_{ff}$, for different normalized frequencies $v^*$: (a) real part, $(\chi_3')$, (b) imaginary part, $(\chi_3'')$. Symbols refer to: (♦•••) $v^*=10^{-4}$; (♦●●) $v^*=10^{-3}$; (♦▷▷) $v^*=10^{-2}$; (♦★★) $v^*=10^{-1}$; (♦←←) $v^*=1$. 

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>$\chi_3'$</th>
<th>$\chi_3''$</th>
</tr>
</thead>
<tbody>
<tr>
<td>88.0</td>
<td>-0.12</td>
<td>0.06</td>
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<td>90.5</td>
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<td>91.5</td>
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</tr>
<tr>
<td>92.0</td>
<td>0.04</td>
<td>0.06</td>
</tr>
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</table>
FIG. 7 - Third harmonic susceptibilities vs temperature evaluated for $B_0=20\,\text{mT}$ and different normalized frequencies $\nu^*$, with the "Taf" resistivity, in the pinning model I: a) real parts, $(\chi'_3)$; b) imaginary parts, $(\chi''_3)$. Symbols refer to (•••) $\nu^*=10^{-4}$; (––) $\nu^*=10^{-3}$; (––) $\nu^*=10^{-2}$; (– – – – –) $\nu^*=10^{-1}$; (– – – – –) $\nu^*=1$.

Even in such a case, the single components of the third harmonic $(\chi'_3, \chi''_3)$ are analyzed separately, because their features cannot be unfolded from the analysis of the modulus.

The temperature dependencies of the real and imaginary part of the third harmonic $\chi'_3(\text{I;par})$ and $\chi''_3(\text{I;par})$ for the parallel resistivity, are reported in Figs. 8a and 8b for different frequencies and $B_0=20\,\text{mT}$; the Bean critical state result is also plotted for comparison. In such cases, the increase of the frequency gives rise to meaningful qualitative differences in the temperature dependencies and, as a consequence, it is not possible to describe the susceptibility components in terms of a single scaling parameter.\(^{21}\)
FIG. 8 - Third harmonic susceptibilities vs temperature evaluated for $B_0=20\text{mT}$ and different normalized frequencies $\nu^*$, with $\rho_{\text{par}}$ in the pinning model $I$: (a) real parts, $(\chi_3')$; (b) imaginary parts, $(\chi_3'')$. Symbols refer to: $(-\triangle--) \nu^*=10^{-4}$; $(-\square--) \nu^*=10^{-3}$; $(-\blacktriangle--\blacktriangle-\blacktriangle-) \nu^*=10^{-2}$; $(-\blacktriangle--\blacktriangle-\blacktriangle-\blacktriangle-\blacktriangle-) \nu^*=10^{-1}$.

In particular, for $\nu^*=10^{-4}$, the overall shape of the curve resembles the critical state behavior but with some differences. In the critical state, the real part is always positive whereas the diffusion calculation yields negative values for $T<90.7\text{K}$ (Fig.8a). Moreover, for temperatures around $T_c$, the critical state $\chi''_3(I)$ has a negative value whereas the diffusive process yields small positive values.
FIG. 9. Third harmonic susceptibilities vs temperature evaluated for $B_0=20 \text{mT}$ and different normalized frequencies $v^*$, with the "creep" resistivity in the pinning model $I$: 
a) real parts, ($\chi_3'$); b) imaginary parts, ($\chi_3''$). Symbols refer to: (\textbullet\textbullet\textbullet\textbullet\textbullet) $v^*=10^{-4}$;  
(\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet) $v^*=10^{-3}$; (\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet) $v^*=10^{-2}$; (\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet\textbullet) $v^*=10^{-1}$.

The temperature dependence of the third harmonics, calculated with $\rho_{\text{par}}$ for $v^*=10^{-2}$ and $v^*=10^{-1}$ (Fig.8 a,b), show large differences from the critical state prediction. Indeed, $\chi_3'(I)$ is always positive for the critical state, while for $v^*=10^{-2}$, $\chi_3'(I;\text{par})$ has large negative values with the presence of two minima around $T=90.5\text{K}$ and $T=92\text{K}$ . For $v^*=10^{-1}$ only a negative peak appears just below $T_c$.

As far as $\chi_3''(I;\text{par})$ at $v^*=10^{-2}$ is concerned, qualitative deviations from the critical state behavior appear above $T=90.2\text{K}$, with the presence of a positive maximum around $T=92.2\text{K}$ whose amplitude increases with the frequency.

As previously stated for the first harmonics, such behaviors are essentially determined by the flux flow resistivity; in fact for $v^*>10^{-2}$ and $T>91.5\text{K}$ the results of the parallel model (Figs. 8a,b) coincide with the flux flow one (Figs.6a,b).

It can be shown (Fig.9a,b) that results obtained at different frequencies using the creep
resistivity only yield a mere shift ($\Delta T=0.5K$) of the curve toward $T_c$. Furthermore, if larger ac fields are applied or a lower critical current density is assumed, all the structures of the curves shift to lower temperature. For values such that the $\chi_3'(I;cr)$ peak is placed below 85 K, the diffusion result is practically identical to the critical state one.

![Diagram](image)

**FIG. 10** - Third harmonic susceptibilities vs temperature evaluated for $B_0=20mT$ and different normalized frequencies $\nu^*$, with $\rho_{par}$ in the pinning model II:

- (a) real parts, ($\chi_3'$) 
- (b) imaginary parts, ($\chi_3''$). Symbols refer to: ($\cdot\square\cdot$) $\nu^*=10^{-4}$; ($\cdot\blacksquare\cdot$) $\nu^*=10^{-3}$; ($\cdot\blacksquare\cdot$) $\nu^*=10^{-2}$; ($\blacktriangledown$) $\nu^*=10^{-1}$.

Within the framework of the pinning model II, the temperature dependencies of the third harmonics, are shown in Figs.10a,b for different frequencies at $B_0=20mT$. The Bean critical state prediction is also plotted for comparison. The analysis of $\chi_3'(II;par)$ and $\chi_3''(II;par)$ confirms the results obtained for the first harmonic, but with some differences: indeed, the shift of $\chi_3'(II;par)$ and $\chi_3''(II;par)$ with frequency develops in a narrower temperature range ($\Delta T=0.2K$).

An immediate feeling of the dominant contribution to the diffusion process comes from the $\chi_3'(II;par)$ plots. Indeed $\chi_3'(II;par)$ always takes negative values with a large negative peak
similar to "Taff" and "flux flow" behaviors, opposite to the Bean model result. In particular, the following statements can be made on $\chi_3^{\text{(II)}}$:

a) at the lowest frequencies, the parallel result is close to the "Taff" one
b) as the frequency increase, the parallel result is similar to the flux creep behavior

Therefore for the pinning model II, as the frequency increases, the "parallel resistivity" show a transition from "Taff" to "creep" regime, with a significant contribution of the flux flow at the highest frequency investigated.

Even for the pinning model III, the analysis of higher harmonic is confirmed to be a more stringent test with respect to the fundamental harmonic. In such case, due to the lower values of both critical current and pinning energy, the main features of the third harmonic appear in a temperature range wider than for the other pinning models.

Results for $\chi'_3^{\text{(III;par)}}$ and $\chi''_3^{\text{(III;par)}}$, are presented in Figs.11a,b. The $\chi'_3^{\text{(III;par)}}$ plot displays negative values in a wide temperature range, clearly differing from the positive critical state prediction. For frequencies above $\nu^* \geq 10^{-3}$, the negative peak of $\chi'_3^{\text{(III;par)}}$ and the oscillations of $\chi''_3^{\text{(III;par)}}$ disappear. By analyzing the behaviors of $\chi''_3^{\text{(III;par)}}$ obtained with different resistivities forms it can be argued that:

a) for $10^{-4} \leq \nu^* \leq 10^{-2}$ the behavior of $\chi''_3^{\text{(III;par)}}$ is similar to that of $\chi''_3^{\text{(III;Tf)}}$ up to 88.8K, differing for higher temperatures; for the same frequencies "creep" and "Taff" yield very similar results;

b) for $10^{-1} \leq \nu^* \leq 1$ the $\chi''_3^{\text{(III;par)}}$ and $\chi''_3^{\text{(III;cr)}}$ curves merge; only very near $T_c$ the "flux flow" give some contribution.

A general fact common to the different pinning models, is that the increase of the frequency determines a change in the non linearity of the dominant resistivity and of the related magnetic flux diffusion coefficient. Such variation produce significant qualitative changes of the shape of the temperature dependencies which cannot be described by a single scaling parameter, $\delta(\omega,T)$. In other terms, the susceptibilities cannot be written as, $\chi_n = f_n(\delta(\omega,T))$, since any frequency change determines variations of the functions $f_n(\delta(\omega,T))$.

As far as a comparison with experimental data is concerned, to the best of our knowledge susceptibility data collected in absence of dc fields are not available at the present time.
FIG.11 - Third harmonic susceptibilities vs temperature evaluated for $B_0=20\text{mT}$ and different normalized frequencies $\nu^*$, with $\rho_{\text{par}}$ in the pinning model III: a) real parts, ($\chi'_3$); b) imaginary parts,($\chi''_3$). Symbols refer to: 
(- - - - ) $\nu^*=10^{-4}$; ( - - - ) $\nu^*=10^{-3}$; ( - - - - ) $\nu^*=10^{-2}$; ( - x - ) $\nu^*=10^{-1}$.

4 – SUMMARY AND CONCLUSIONS

The temperature dependencies of the first and higher harmonics of the complex magnetic susceptibility have been analyzed by numerically solving the diffusion equation for the local magnetic field, using a parallel resistivity model which takes into account both creep and flux flow processes. Within such approach different non-linear behaviors are present in different regions of the I-V characteristic. The simulations refer to an YBCO slab in presence of longitudinal ac magnetic fields of 2 and 20 mT, in the frequency range $0.8 \text{ - } 8*10^3 \text{ Hz}$. The absence of a dc field restricts our analysis only in a few K below $T_c$. The material parameters relevant for the calculation are: the critical current density at $T=0\text{K}$ ($J_c(0)=10^{10} \text{ A/m}^2$), the upper critical field ($H_{c2}(0)=112\text{T}$), and the pinning potential at $T=0\text{ K}$ ($U_p(0)/K_B=2 \times 10^4 \text{ K}$). In any case, the main results are not affected by the exact parameters values chosen for the calculations.
Three distinct pinning models have been considered, which determine different temperature dependencies of the pinning potential and the critical current density. The first (I) is the collective pinning model; the third (III) resembles the "giant flux creep" model, while the second (II) shows larger pinning potentials and very larger critical current densities in a temperature range of 10K below \( T_c \).

In general, the performed analysis points out that the current densities generated by the ac fields determine different regimes of flux motion ("Taff", "creep", "flow") in temperature and frequency ranges, which depend on the pinning models. The transition between different regimes induces a non universal behavior in the temperature dependence of the susceptibilities. This crossover cannot be analyzed in simpler models previously used in the literature.

In particular, for the collective pinning model I and for the lowest investigated frequencies, a behavior similar to the critical state is found in some regions of the magnetization cycle, with an effective current density \( J_{c^*}(T,v) < J_c(T) \). Such differences are more significant close to \( T_c \). In any case, the value of \( J_{c^*}(T,v) \) increases with the frequency. In such cases the diffusion process is determined by the creep phenomenon with the presence of currents high enough to drive the system far from the "Taff" limit. In such conditions the overall behavior can be described, in a first approximation, by a single scaling parameter. However, as the frequency increases, the induced currents increase in such way that the flux flow contribution cannot be neglected, so that at the highest frequencies and close to \( T_c \), the magnetization cycles are completely determined by the flux flow process. In these conditions an overall behavior far from the critical state is found so that the scaling procedure is not allowed.

For the pinning model II, the higher values of the critical currents are such that the main features of the susceptibilities develop within 1K below \( T_c \), so that thermally activated processes become relevant. In this case at low frequencies the parallel resistivity is in the "Taff" limit, which in turn goes to the "creep" regime for temperatures above 92.1K. Only at the highest frequencies investigated and within a few tenth of K near \( T_c \), the "flow" component gives some contribution.

In the model III the lower values of the pinning potential determine a larger relevance of the thermally activated processes also at lower temperatures, so that the main features of the low field susceptibilities appear in a range of \( \approx 10K \) below \( T_c \). Even in such a case, at the lowest frequency and below 87.5K, the susceptibilities are determined by the "Taff" resistivity whereas, at higher temperatures and higher frequencies, the diffusion process is no more in the Taff limit and a flux flow contribution appears.

A key new result of this work is the frequency dependence of the amplitude of the peak of \( \chi^\prime_1 \), which is related to different non-linear behaviors of the different ranges of the I-V characteristic, determined by the current values induced by the magnetic field variations. Indeed, the peak amplitude generated by a field dependent linear resistivity (flux flow or Taff) is of about 0.5, whereas the highly non-linear behavior of the "creep" resistivity gives rise to a peak value closer and closer to the critical state prediction (0.24) as the non-linearity increases. For such reason in our conditions for the model I, as the frequency increases, the transition from creep resistivity to a flux flow one generates an increase of the peak value. On the contrary, in the II model, the transition from "Taff" to "creep" regime, generated by the increase of the frequency, yields a reduction of the peak. Finally, a constant peak value \( \approx 0.5 \) is
found in the model III for the "Taff" to "flux flow" transition.

These three different behaviors give a first experimental criterion for the identification of the actual pinning model; however, for this purpose, the absolute calibration of the susceptibility apparatus is necessary. Anyhow, a more pressing experimental criteria for the choice among different pinning models can be given by the inspection of the frequency dependence of both peak amplitude and temperature.

Indeed, as far as the frequency dependence of the peak temperature is concerned, $T_p$ clearly increases within any model since it is related to the increase of the induced currents driven by the field frequency. However such dependencies appear to be qualitatively different in the different pinning models which show a similar frequency dependence of the amplitude. For instance, the amplitude analysis can give similar results ($\chi''_1(\text{peak})=0.5$) even for very different dynamic regimes (Taff, flux flow) determined by different pinning models; however these different regimes can be discriminated by observing the frequency dependence of the peak temperature.

As a matter of fact the numerical behavior found for the different model is in a qualitative agreement with experimental data found in literature for different materials.

A second key result is the large difference observed between the numerical slope of $1/T_p$ vs $\ln \omega$ and the value $-K/U_p$ predicted by the analytical approximated approach \(^2\). Thus, a lot of care should be taken in the determination of $U_p$ from the frequency dependence of the peak temperature, experimentally observed.

As far as higher harmonic components are concerned, the crossover between different dissipative regimes, induced by the frequency increase, strongly affects the shape of their temperature dependence.

A third novel key result is the relevant presence of high harmonics even for diffusion processes dominated by $\rho_{ff}$ and $\rho_{Tf}$ resistivities, as long as the dependence on the local field is introduced. These resistivities are responsible for large qualitative differences in the temperature dependencies of the third harmonics. In this way, the analysis of real and imaginary components of higher harmonics is confirmed to be a more stringent test for the choice of the pinning model with respect to the fundamental harmonic.

In conclusion, the presented analysis describes temperature and frequency dependencies of the susceptibilities in contrast with simpler models which leads to functional dependencies of the susceptibilities $\chi_n=f_n(\delta(\omega,T))$ depending only on a single scaling parameter $\delta(\omega,T)$. Therefore we believe that no critical state description, even refined for the frequency dependence, can describe the physical phenomena underlying low frequency - low fields complex ac susceptibility measurements. Indeed, as the frequency increases, our numerical analysis shows significant variations of the functions $f_n(T)$. Such variations are due to the transitions between different dissipative processes (taff, creep, flow).

For these reasons, the experimental criteria to discriminate among different pinning models can be given by the frequency dependencies of the susceptibilities and in particular of the absolute amplitude of the peak, and of the peak temperature for the imaginary part of the first harmonic. However for a deeper analysis a full best fit procedure will be necessary, using numerical simulations taking into account the different losses mechanisms in the different pinning models.
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