Frequency dependence of the intergranular magnetic flux penetration in ceramic $YBa_2Cu_3O_x$ superconductor

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By means of small magnetic field ac susceptibility measurement at 10 kHz we found that the real and imaginary parts of ceramic $YBa_2Cu_3O_x$ susceptibility in presence of the external low frequency field close to 0.1 Hz exhibit frequency dependence. The wide maximum of hysteresis losses and the exponential dependence of the effectiveness of flux penetration with increasing of external field frequency were obtained. We observed a nonlinear dynamic magnetic response in presence dc field and we suggest that this behavior is due to the dynamic and static Josephson vortex-vortex interaction.

1 Introduction

In an early stage of studies of the ac magnetic field response in granular high temperature superconductors (HTSC) many researchers have reported that the temperature $T_{m}'$, indicating the maximum of hysteresis losses of complex magnetic susceptibility $\chi = \chi' - i\chi''$ shifts to the lower values at the applied dc magnetic field on the sample [1-3]. Lobotka and Gomory have reported that the larger dc field values than ac field amplitudes are needed to shift the $\chi''$ peak temperature $T_{m}'$ by the same amount [1]. According to Clem [4] and Gershkenbein, Vinokur, and Fehrenbache [5] the $T_{m}'$ in $\chi''$ corresponds to the temperature at which ac magnetic field with amplitude $h_{ac}$ fully penetrates the sample volume. From this point of view one can expect a frequency dependence of magnetic flux penetration effectiveness in intergranular medium of ceramic superconductor. In [6] some increase of $T_{m}'$ ($\Delta T_{m}' \approx 2$ K) with frequency in the frequencies range of ac magnetic field from 300 Hz to 80 kHz was found and authors explained it by a spin-glass-like behavior of superconducting granular HTCS sample. Other authors [7-10] pointed a weak dependence or full absence [11-13] of frequency dependences of complex magnetic susceptibility in ceramic HTSC materials.

In this paper we present the results of experimental investigations of the frequency dependences of magnetic field penetrations in ceramic $YBa_2Cu_3O_x$ superconductors at non full magnetic flux penetration regime of the sample volume at temperature $T=80$ K for both external sinusoidal magnetic field in frequency range from $f_{ex} = 0.01$ Hz to 90 Hz and amplitude $H_{ac} \leq 40$ Oe and static magnetic field $H_{dc} \leq 40$ Oe. In our experiments we
used the small magnetic field (amplitude 2 mOe) ac susceptibility measurements technique at frequency 10 kHz.

The real part of ac susceptibility has been taken as a deviation $\Delta \chi' = (\chi'_0 - \chi')/\chi'_0$, where $\chi'$ is the current value of the real part and $\chi'_0$ is the value of $\chi'$ at which a full screening state of the sample occurs. The magnitude of $\Delta \chi'$ is proportional to the fraction of the sample volume where the Josephson vortex penetrated. $\Delta \chi' = 0$ corresponds to full screening state of the sample and our measurement $\Delta \chi' = 0.28$ corresponds to the sample state at which Josephson vortexes reaches the center of the sample (see. Fig.1(b) in the text).

### Experimental

Ceramic $YBa_2Cu_3O_x$ samples with cylindrical shape of 3mm in diameter and 8mm long were prepared by standard solid-state reaction method. The real $\chi'$ and imaginary $\chi''$ parts of complex magnetic susceptibility of HTSC samples were measured by small magnetic field ac susceptibility measurements technique at frequency 10 kHz and amplitude 2 mOe in presence both static field or low frequency sinusoidal magnetic field. The ac magnetic field is considered small if its contribution to the measuring values of the susceptibility components $\chi'$ and $\chi''$ are negligible. For each sample and measuring regime exist experimentally determined range of small measuring field amplitude $h_{ac}$ at which both real and imaginary components of ac susceptibility are independent of $h_{ac}$. On the other hand the reduction of the ac field amplitude is limited by the equipment sensitivity. Taking into account these two requirements the optimum ac field amplitude $h_{ac}$ is the high end of the small magnetic field range. For our investigations $h_{ac} = 2$ mOe was obtained (see. Fig.2 in the text).

Complex magnetic susceptibility was measured using a homemade ac inductance bridge working at frequency $f_m = 10$ kHz with amplitude $h_{ac}$ in range 0.5 mOe to 10 Oe. The real $\chi'$ and imaginary $\chi''$ parts of the ac complex susceptibility are determined from the data of measuring coil $L_m$ parameters deviation caused by a superconducting sample which were surrounded by this coil. The $L_m$ is connected in the arm of the ac inductance bridge. Another external larger coil $L_{ex}$ was used for creation of static $H_{dc}$ or sinusoidal $H_{ex}(t)$ magnetic field in frequency rang from 0.01 Hz to 90 Hz and amplitude up to 40 Oe, where t is the time. The axes of both coils were orientated parallel to the cylindrical sample axis. The phase angle adjustment of two Lock-in detectors for both real and imaginary signal components of ac bridge was carried out connecting and disconnecting a calibrating inductance and resistance in series to the measuring coil $L_m$.

In our experiments the influence of the Earth magnetic field on measured values of the real $\chi'$ and imaginary $\chi''$ parts was negligible and all measurements in ambient Earth magnetic field were carried out. Temperature of the sample was monitored with help of a copper wire resistor with relative accuracy of about 0.1 K. The measuring coil $L_m$ was put in the liquid nitrogen and the measurements were carried out in heating regime of the sample at temperature rate about 1 K/min. Our experimental data have appeared reproducible within the sensitivity of our measurements for all investigated samples and their measuring regimes.
3 Experimental results and discussions

In Fig.1 (a) and (b) we show the temperature dependences of the imaginary $\chi''$ and real $\chi'$ components of the ac complex susceptibility $\chi(T) = \chi'(T) - i\chi''(T)$ of the ceramic sample $YBa_2Cu_3O_x$ measured for different ac field amplitudes and superimposed dc field regimes. The curves 1, 2 and 3 in this figure corresponds to the measuring ac magnetic field amplitude $h_{ac} = 2$ mOe, 0.9 Oe and 5.3 Oe respectively, and they have not noticeable features and are similar to the results previously reported by many researchers. However there are many interesting peculiarities at comparison on the curves 2(a), 2(b) and 4(a), 4(b) in a Fig.1, where curve 4(a) and 4(b) are obtained by measuring ac susceptibility at amplitude $h_{ac} = 2$ mOe in the presence of dc field $H_{dc} = 40$ Oe. In the presence of dc field the curves 1(a) and 1(b) in the Fig.1 are transformed and the temperatures $T_{Jm}$ of hysteresis losses peak $\chi''_{\text{max}}$ are shifted to lower temperatures and the slope of a curve $\chi'(T)$ decreases strongly.

In order to compare the ac and static $H_{dc}$ magnetic fields flux penetration effectiveness into the sample volume, from the $\chi'(T)$ and $\chi''(T)$ curve families the curves for which the hysteresis losses peak temperature $T_{Jm}$ have the same value (curves 2(a) and 4(a) in Fig.1) were chosen, i.e. in both cases the magnetic flux front reaches the sample center at temperature $T_{Jm} = 87.2$ K. In case of the presence of a dc field $H_{dc} = 40$ Oe the contribution from a measuring magnetic field $h_m(t)$ with frequency 10 kHz and amplitude 2 mOe to the value of $\chi'(T)$ is negligibly small, because in the amplitude range from 0.5 mOe up to 2 mOe the $\chi'(T)$ at the temperature interval from 80 K to 92K do not vary noticeably. It means that in the latter case the magnetic response is caused by a static magnetic field.

The obtained experimental results show, that the full penetration regime of the magnetic flux in the sample volume at temperature $T=87.2$ K in both cases take place at the ratio of dc field to the effective value of the ac field equal to $\sqrt{2}H_{dc}/h_{ac} \cong 60$. Therefore we can suggest that the effectiveness of the magnetic flux penetration in sample volume for static and ac fields strongly differ. We note that this ratio decreases with increase of $T_{Jm}$ and at $T=88.3$ K is equal 50. The decrease of $T_{Jm}$ in $\chi''$ at a superimposed dc field were reported in [1-3], however the contributions to the measured values of $\chi'$ and $\chi''$ of static field and ac field were not separated and at that case it is very difficult to judge on penetrating ability of dc field and ac field in the sample volume.

Thus, our experimental data show that the Josephson vortexes in $YBa_2Cu_3O_x$ ceramic samples created by static and ac magnetic fields have a different character and we named them static and dynamic Josephson vortex respectively. As can be seen by comparing curve 2(a) and curve 4(a) in Fig.1, the penetration effectiveness of dynamic vortexes in intergranular medium of HTSC ceramic superconducting sample is much more than that for a static vortex and therefore the density of dynamic Josephson vortexes is less than static Josephson vortexes density.

Another feature of the curve 4(a) in comparison with curve 2(a) in Fig.1 is that from temperature $T=83$ K and higher the hysteresis losses $\chi''$ at the presence of static field $H_{dc} = 40$ Oe are noticeably lower than those for $\chi''$ measured by ac field at frequency 10 kHz with amplitude $h_{ac} = 0, 9$ Oe. We suggest that such behavior of the magnetic response is due to vortex - vortex interaction. In other words hysteresis losses at interaction
of dynamic vortexes of the measuring small field with the static vortexes are less than hysteresis losses occurring at interaction of dynamic vortexes with each other at measuring field amplitude $h_{ac} = 0.9$ Oe.

In the Fig. 1(b) the temperature dependences of a real part of complex magnetic susceptibility $\chi'(T)$ measured at different field regimes are shown. As can be seen in Fig. 1 the behaviors of $\chi'(T)$ in curve 2(b) and curve 4(b) in the neighborhood of temperature $T_m'$ differ slightly, whereas at the same temperature range the differences of $\chi''(T)$ are clearly visible. From the Fig. 1 we can see that the hysteresis losses peak $\chi''_{\text{max}}$ for all measuring regimes occurs at value of $\chi' \approx -0.72$ and this corresponds to $\Delta \chi' \approx 0.28$.

The difference in behavior of the magnetic response in HTCS samples in the presence of dc and ac magnetic fields is visible not only at temperature rang of occurrence of hysteresis losses peak, but it is clearly seen in the behavior of $\chi'(f_{ex})$ and $\chi''(f_{ex})$ for low frequency external magnetic field $H_{ex}(t)$. However, before presenting these data, it is necessary to show that the values of $\chi'(f_{ex})$ and $\chi''(f_{ex})$ are caused by action of $H_{ex}(t)$ only, because the ac susceptibility measurements will be carried out at frequency $f_m = 10$ kHz. To do this it is necessary to determine experimentally the range of small measuring magnetic field amplitude and further, to find its optimal magnitude.

With this purpose both the real $\Delta \chi'$ and imaginary $\chi''$ components of complex susceptibility were measured versus ac field amplitude $h_{ac}$ in range from 0.5 mOe up to 10 mOe with frequency $f_m = 10$ kHz for different values of external magnetic $H_{ex}(t)$ field at the temperature $T=80$ K. These data are presented in Fig. 2 (a) and (b), where it is shown that in presence of dc field, starting from the amplitude value $h_{ac} = 3$ mOe and above, there is a noticeable increase of the imaginary $\chi''$ and real $\Delta \chi'$ components (curves 1, 2, 3 in a Fig. 2 (a) and (b)). It is also seen in this figure, that for fixed value of amplitude $h_{ac}$ the increases of the magnitudes of these components depend on static field value and that the higher the magnitude of $H_{dc}$, the more is the magnitude of contribution of the measuring field to these components. In the case $h_{ac}=10$ mOe both components rise almost by factor two whereas at the absence of the dc field the magnitude of imaginary $\chi''$ and real $\Delta \chi'$ parts are equal to zero and remain independent on $h_{ac}$ in the range from 0.5 mOe up to 10 mOe at the temperature of sample $T=80$ K.

This is the most important feature from which we can see that even in the case when the sample state is essentially far from the magnetic flux fully penetration regime ($\Delta \chi' < 0.04$) the magnitude of imaginary $\chi''$ and real $\Delta \chi'$ components of magnetic susceptibility are not a superposition of the contributions of each of these fields, and have more complex character. We suggest that the appearance of such nonlinear magnetic response in this case is caused by the interaction of the dynamic vortexes of ac measuring field with the static vortexes of dc field and this effect becomes noticeable at $h_{ac} \geq 3$ mOe. In other measuring regime, when the external field has the sinusoidal shape, the dependence of $\Delta \chi'$ and $\chi''$ on $h_{ac}$ practically is not noticeable in the range of $h_{ac}$ from 0.5 mOe up to 10 mOe (see curves 4, 5, 6 in a Fig. 2 (a) and (b)). In the last case we suppose that the interaction of two dynamic incoherent Josephson vortexes created by the measuring ac field with frequency $f_m = 10$ kHz and amplitude up to 10 mOe with an external field $H_{ex}(t)$ at frequency $f_{ex} \leq 90$ Hz does not give noticeable contribution to the values $\Delta \chi'$ and $\chi''$.

Thus the experimental data in a Fig. 2 (a) and (b) indicated that: a) the amplitude
$h_{ac} = 2 \text{ mOe}$ of the low magnetic field at frequency 10 kHz is optimal for measuring values of the real $\Delta \chi'$ and imaginary $\chi''$ components and therefore it is possible to state that the magnetic response in ceramic samples in our investigations was caused by external field $H_{ex}(t)$ only; b) the interaction of the dynamic vortexes with the static vortexes, dynamic vortex-vortex and dynamic incoherence vortexes have different behavior. From obtained experimental data we can suggest that the dynamic magnetic response of ceramic $YBa_2Cu_3O_x$ superconductor in none fully magnetic flux penetration regime in intergranular environment is substantially determined by three types of vortex-vortex interactions. Note that it is necessary to take into account as well the superconducting properties of the HTCS sample (pinning centers), because during the penetration of dynamic Josephson vortexes into of the sample bulk a noticeable role can be attributed to the interaction of dynamic vortexes with static vortexes, frizzed in the sample volume at penetration of a sinusoidal magnetic field after the first quarter of the period. More detailed experimental data about vortex-vortex interaction and the penetration dynamics of Josephson vortexes in intergranular environment of HTCS sample will be introduced elsewhere.

Fig. 3 (a) and (b) show a frequency dependence of imaginary $\chi''$ and real $\Delta \chi'$ components of complex susceptibility for some values of external magnetic field $H_{ex}(t)$ at the temperature $T=80 \text{ K}$, measured by ac field with the amplitude $2 \text{ mOe}$ and frequency 10 kHz.

As can be seen the magnitude of hysteresis losses $\chi''(f_{ex})$ at the presence of a static field and a sinusoidal field up to frequency about $f_{ex} = 0.1 \text{ Hz}$ does not exhibit a frequency dependence (Fig. 3 (a)). Further $\chi''$ starts to rise and exhibits a wide maximum versus external magnetic field $H_{ex}(t)$ frequency and the higher amplitude of a magnetic field, the lower the frequency of occurrence of the $\chi''$ peak. For example, the value of $H_{ac} = 40 \text{ Oe}$ corresponds to the $\chi''$ peak frequency $f_{ex}^{\text{max}} \approx 0.5 \text{ Hz}$, and the value $H_{ac} = 10 \text{ Oe}$ corresponds to $f_{ex}^{\text{max}} \approx 5 \text{ Hz}$.

Fig. 3(b) shows that the magnitude of $\Delta \chi'$ up to frequency about $f_{ex} = 0.1 \text{ Hz}$ does not depend on the frequency of $H_{ex}(t)$, but with the increase of $f_{ex}$, for each amplitude value of magnetic field $H_{ac}$, there is a threshold frequency, starting from which the $\Delta \chi'$ exhibits a frequency dependence. The increase of $\Delta \chi'$ in this frequency range is described by exponential law $\Delta \chi' \propto f_{ex}^{0.62}$. Thus the increase of the frequency of the external magnetic field from $f_{ex} \approx 0.1 \text{ Hz}$ for fixed amplitude $H_{ac}$ leads to the increase of the effectiveness of the magnetic flux penetration in the sample volume.

Finally we note that qualitatively similar experimental results were obtained for the bismuth based ceramic HTCS sample ($Bi_{1.7}Pb_{0.2}Sr_{0.1}Ca_2Cu_3O_x$; $T_c = 108 \text{ K}$) at the temperature $T=80 \text{ K}$.

4 Conclusions

We have shown that the real and imaginary parts of the small magnetic field ac susceptibility at frequency 10 kHz and amplitude 2 mOe in presence of low frequency external magnetic field at non fully penetration regime in the ceramic $YBa_2Cu_3O_x$ sample at $T=80 \text{ K}$ close to $f_{ex} = 0.1 \text{ Hz}$ begin to exhibit frequency dependence. With increasing of external magnetic field frequency the wide maximum of hysteresis losses and the ex-
ponential dependence ($\Delta \chi' \propto f_{ex}^{0.62}$) of the effectiveness of flux penetration in the sample volume were obtained.

It is established that the penetration effectiveness of ac field at frequency 10 kHz is much more than that for dc field and the magnetic flux in both cases reaches the cylindrical sample center at temperature $T=87.2$ K at the ratio of the magnetic fields $\sqrt{2}H_{dc}/H_{ac} \approx 60$.

It is found that the dynamic magnetic response in ceramic $YBa_2Cu_3O_x$ samples in presence of dc magnetic field is not a superposition of the contributions of measuring ac field and dc field. We suggest that in this case the nonlinear dynamic magnetic response was caused by dynamic and static Josephson vortex-vortex interaction.

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Figure 1: Temperature dependences of the imaginary $\chi''$ (a) and real $\chi'$ (b) parts of ac susceptibility of the ceramic sample $YBa_2Cu_3O_x$ at frequency 10 kHz for different measuring ac field amplitude $h_{ac}$: 1 - 2 mOe, 2 - 0.9 Oe, 3 - 5.3 Oe, 4 - 2 mOe + $H_{dc}$; $H_{dc}$=40 Oe.
Figure 2: Dependence of imaginary $\chi''$ (a) and real $\Delta \chi' = (\chi'_a - \chi'_o)/\chi'_0$ (b) parts of $YBa_2Cu_3O_x$ sample complex susceptibility versus measuring ac field amplitude $h_{ac}$ at frequency $f_m = 10$kHz at $T = 80$ K in the presence of external static field $H_{dc}$: 1 - 7.1 Oe, 2 - 14.2 Oe, 3 - 28.3 Oe; and external sinusoidal field with amplitude $H_{ac}$ and frequency $f_{ex}$ respectively: 4 - 10 Oe, 5 Hz; 5 - 20 Oe, 2 Hz; 6 - 40 Oe, 1 Hz.
Figure 3: Dependence of the imaginary $\chi''(a)$ and real $\Delta\chi' = (\chi'_0 - \chi')/\chi'_0$ (b) parts of ceramic sample $YBa_2Cu_3O_x$ ac susceptibility versus external field frequency, measured at frequency 10 kHz and amplitude 2 mOe at $T=80$ K for different external field amplitudes $H_{ac}$: 1-10 Oe, 2 - 20 Oe, 3 - 40 Oe. The values of $\chi''$ and $\Delta\chi'$ at $f_{ex} = 0$ correspond to the dc field $H_{dc} = H_{ac}/\sqrt{2}$. 