Manifestation of oxygen ordering in the magnetic relaxation of YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals

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Abstract.

The temperature dependence of the relaxation rate $S = -d \ln M/d \ln t$ for YBa$_2$Cu$_3$O$_{7-\delta}$ samples with different oxygen content was measured. The most important result consists in the qualitative change of the topology of the curve $S(T)$ when the oxygen deficiency $\delta$ passes through some threshold value $\delta_{th} \simeq 0.37$. For the oxygen contents corresponding to $\delta < 0.37$ ($T_c > 60$ K), the function $S(T)$ has a usually observed maximum at $T/T_c \sim 0.4$. For a sample with $T_c = 51$ K, this maximum transforms to a plateau and a new sharp maximum at $T/T_c \sim 0.1$ arises. It is known that the transition from disordered to ordered oxygen state occurs in YBa$_2$Cu$_3$O$_{7-\delta}$ samples with $\delta \simeq \delta_{th}$ ($T_c \simeq 60$ K). We consider that the change in the form of our experimental curve $S(T)$ is a macroscopic manifestation of this transition.

 Shortly after the discovery of high-temperature superconductivity, an intensive study of the dynamics of the vortex system in high-$T_c$ superconductors (HTS) started. A lot of experimental and theoretical works was devoted to the problem of the relaxation of the HTS magnetization in the non-equilibrium state. The measured logarithmic relaxation rate of the magnetization, $S = -d \ln M/d \ln t$, provides important information about the effective barrier height $U$ in the interaction between the magnetic-flux vortices and inhomogeneities (the pinning centers), $S = -d \ln M/d \ln t \sim kT/U(T)$.

A pronounced maximum in the temperature dependence $S(T)$ was observed in numerous experiments. One of the possible approaches to explain this maximum invokes the distributions of various kinds of the energy barriers [1]. According to Refs. [1], the maximum of $S(T)$ is caused by a coexistence of strong and weak pinning centers. The strong centers play the main role in the magnetic relaxation at high temperatures and provide a relatively small relaxation rate. If a sample is magnetized at lower temperatures, the weak centers also take part in pinning that results in a relatively high relaxation rate, though the term $kT \ll U$. Thus, the relaxation rate $S(T)$ should be sensitive to the internal structure of a sample and to a possible phase separation in a superconductor.

A great deal of attention of many research groups is currently being focused on the problems of the phase separation and the origin of different superstructures in the strongly correlated systems, in particular, in oxide superconductors. The phase separation plays a key role in the phenomenon of the colossal magnetoresistance in new oxide magnetic materials (manganites, cobaltites, etc.) [5]. The phase coexistence and the oxygen ordering are also
very important for many physical properties of HTS. Even the nature of the high-temperature superconducting state itself is connected \[3, 4\] to the existence of the oxygen superstructures. The phase separation in the compounds YBa$_2$Cu$_3$O$_{7-\delta}$ can be controlled by the oxygen content. According to Ref. \[6, 7, 8, 9, 10\], the transition of the oxygen vacancies in the yttrium systems to the ordering state occurs at $\delta \geq \delta_{th} \approx 0.37$.

The oxygen content is well known to define the concentration of the free carriers and, therefore, affects main superconducting properties of the oxide superconductors. In particular, the oxygen deficiency leads to the decrease of the critical temperature $T_c$. The detailed study of the dependence of $T_c$ on $\delta$ was made in Refs. \[11, 12\]. According to the obtained results, the curve $T_c(\delta)$ contains a plateau at $T_c \approx 60$ K for $\delta = 0.3 - 0.4$, see Fig. 1 taken from Ref. \[12\]. This plateau corresponds to the region of transition from the disordered oxygen state to an ordered one, possibly in the form of charge and/or spin stripes or other superstructures \[13\].

The intrinsic superstructures in high-$T_c$ superconductors with the oxygen deficiency attract attention of many research groups. To search and study the superstructures, a number of modern methods including low-angle neutron scattering, tunnel and atom-force microscopy, precise X-ray methods have been used. It seems to look into a possible influence of the formed superstructures upon the macroscopic properties of the superconducting oxides. In this paper, we call our attention to the fact that the study of the relaxation rate $S(T)$ can be an efficient macroscopic method for detecting the transition of the oxygen vacancies in HTS to the ordering state. We have measured $S(T)$ for samples with different $\delta$ that correspond to the key-points in the diagram $T_c(\delta)$ shown in Fig. 1. These points are marked by closed circles. The obtained results have indicated that the oxygen content does not have an appreciable effect on the topology of the curve $S(T)$ at $\delta < \delta_{th}$. It contains a single maximum observed previously in many experiments. However, at $\delta > \delta_{th}$, the curve $S(T)$ undergoes qualitative changes.

**Experimental details**— We have studied three YBCO single crystals, S1, S2, and S2, and the melt-textured single domain sample, MTG1. All single crystals were initially saturated with oxygen. They have the critical temperature $T_c \approx 92.5$ K and the transition width of about 0.5 K. One of these samples, S2, with sizes $2 \times 1.5 \times 0.02$ mm$^3$ was subjected to several heat treatments with the aim to reduce the oxygen content $(7 - \delta)$. The heat treatments were performed in gaseous mixtures of oxygen and helium at the annealing temperature 615°C during 72 hours. This process was completed by fast cooling down to room temperature in the same gaseous mixture. The value of $\delta$ was evaluated on the basis of measured $T_c$ and the data from Fig. 1. The melt-textured YBCO plate-like sample MTG1 with sizes $14.5 \times 8.5 \times 0.5$ mm$^3$ was cut from the homogeneous part of a textured monolith. The $c$-axis was perpendicular to the largest face of the sample. The critical temperature for MTG1 was about 91 K.
We studied the relaxation of the static magnetization $M = (B - H_e)/4\pi$ of our samples. To discriminate a small signal from a superconducting sample and to increase the resolution of measurement, it is suitable to measure separately the normal component of the magnetic induction $B$ in the central zone of a sample surface and the field $H_e$. We used for this aim two Hall probes. One of them was placed directly onto a sample and the second one, which measured the external field $H_e$, was shifted away from a sample at a distance of about 2 cm. Both Hall probes were supplied by ac current from the same ac current source. This allowed one to increase the signal-noise ratio. To attain good stability of the sample temperature, a special gas-flow cryostat was designed. As low as 0.01 K temperature stability was reached.

Experimental results— The relaxation was studied for different magnetic prehistory of the sample. The main results were obtained for samples in the remanent magnetic state. The external magnetic field $H_e$ was switched on and then it increased up to the value of about 1 T and decreased to zero. The external magnetic field changed linearly in time with the rate of about 0.2 T/min. The measurement of relaxation of magnetization $M$ was carried out for 1500 s beginning from 10 s after switching off the field $H_e$. We found that the dependence $\ln M(\ln t)$ was almost linear (with a deviation of less than 2%) for all types of samples at any temperatures. The relaxation rate $S$ vs temperature $T$ for oxygen-saturated single crystals S1, S2 and the melt-textured sample MTG1 is shown in Fig. 3. The curves demonstrate pronounced maxima of $S(T)$. The maximum positions for different single crystals are near $T = 40$ K, whereas the maximum for the melted textured sample is close to $T = 30$ K.

The most interesting results were obtained for sample S2 with different oxygen contents. The results are presented in Fig. 3 for reduced temperature. One can see that the curves $S(T)$ for $T_c > 51$ K have the maxima similar to the curves in Fig. 3. All maxima correspond to approximately the same reduced temperature $T/T_c \sim 0.4$. This means that the oxygen contents does not have an essential influence upon the distribution of the pinning-center in samples with $T_c > 51$ K. However, the curve $S(T)$ undergoes the striking topological change for the sample with $T_c = 51$ K. The maximum at $T/T_c \sim 0.4$ transforms to a plateau and a new sharp maximum appears at $T/T_c \sim 0.1$ ($T = 5.5$ K). Such a maximum at low temperature does not exist for single crystal S3 having $T_c = 92.5$ K.

![Figure 2](image1.png)  
**Figure 2.** The dependence of $T_c$ vs the oxygen deficiency, $\delta$, for YBa$_2$Cu$_3$O$_{7-\delta}$ samples (data are taken from Ref. [12]). Closed circles correspond to the oxygen contents used in our measurements.

![Figure 3](image2.png)  
**Figure 3.** The relaxation rate for sample S2 with different oxygen contents. Lines are guide for the eye.

At first glance, such behavior of relaxation rate is very surprising. Indeed, the relaxation rate significantly increases in the helium temperature range (the maximum position is observed at
$T = 5.6 \text{ K}$). We think that this observation reflects the essential rearrangement in the distribution of the oxygen vacancies. A remarkable thing is that just for $\delta < \delta_{\text{th}} \approx 0.37$ (or $T_c < 60 \text{ K}$), where the new maximum is observed, the YBa$_2$Cu$_3$O$_{7-\delta}$ samples transit from the disordered oxygen state to the ordered one. Based upon the interpretation of the maximum in the dependence $S(T)$ given in Ref. [1], we conclude that the appearance of the new maximum demonstrates an existence of a new kind of weak pinning centers in the sample. The role of such centers can be played, say, the stripes or other extended long-range correlated objects widely discussed in the literature [13]. Indeed, the extended defects, e.g. twins, produce a pronounced guiding effect. The pinning, preventing the vortex movement across the extended defect, is strong, but the activation energy for the movement along the defect can be very small.

To ensure it is the extended-defects (super)lattice that provide the new maximum in the relaxation rate, we have ground sample S2 and obtained a powder with the same oxygen content. Ipso facto, we have eliminated the extended pinning centers from the superconductor. As a result, the new maximum at $T/T_c \sim 0.1$ in the dependence $S(T)$ disappeared and the “old” maximum at $T/T_c \sim 0.4$ was recovered (see the curve with open squares in Fig. 3). Our results correlate well with data of recent paper [14].

Thus, we consider the change in the topology of curve $S(T)$ as a macroscopic manifestation of this transition.

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