Paramagnetic moments and time effects in melt-textured NdBaCuO system with Nd422 inclusions
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Abstract. We have performed magnetic measurements in two melt-textured NdBaCuO₆.₈ samples with Nd422 inclusions under magnetic fields from 0.05 up to 14 T, applied parallel to the ab planes. The measurements were made with a superconducting quantum interference device (SQUID) and a vibrating sample magnetometer (VSM). Paramagnetic moments could be observed during FCC (field-cooled cooling) and FCW (field-cooled warming) experiments. This effect, known as Paramagnetic Meissner Effect (PME), persisted up to 14 T and strong irreversibilities were observed among FCC and FCW experiments, revealing the presence of time effects. These time effects were confirmed by specific magnetic relaxation experiments in different cooling rates and temperatures, showing an anomalous and curious paramagnetic behavior. We explain our results based on the flux-compressed state generated within non-superconducting regions of the sample, such as the Nd422 inclusions dispersed into the superconducting matrix. These inclusions may produce a strong vortex pinning that stabilize the paramagnetic state, allowing the admission of extra vortices into the sample responsible for the positive moments during the relaxation experiments.

1. Introduction

The paramagnetic Meissner effect, or simply PME, has been characterized by a paramagnetic moment exhibited when a superconducting material is cooled below the critical temperature in the presence of a magnetic field (field-cooled procedure), contrary to conventional diamagnetism. The first observation
of the PME occurred with the Bi$_2$Sr$_2$CaCu$_2$O$_x$ superconductor (Bi-2212) [1,2]. Since then the effect has been widely observed in different superconducting systems, such as Nb [3], pure Pb [4], YBa$_2$Cu$_3$O$_{7-\delta}$ [5,6], multiphase compounds [7], organic systems [8], superlattices [9] and recently in iron arsenide systems [10]. Together with the observation in different superconducting systems, the PME is also reported in different type of samples, since sintered or melt-textured polycrystalline materials [5,11,12], thin films [13,14,15] and even in single crystals [16,17].

Due to different characteristics of the PME observed and the differences between the superconducting materials, several models have emerged in order to explain the origin of the effect. Some models attribute the origin of the PME as a consequence of $\pi$ junctions randomly distributed in a granular array of superconducting grains [18,19,20,21], or a compressed flux state modulated by pinning effects [22,23,24]. On the other hand there are models based on surface and geometry effects [3,6,25,26], chiral pairing [10] and strong pinning effects [12].

In this work we present the experimental observation of the paramagnetic Meissner effect (PME) in melt-textured NdBa$_2$Cu$_3$O$_{7-\delta}$ samples in field-cooled (FC) experiments with magnetic fields up to 14 T applied along the $ab$ planes. Our results show a PME with a time dependence confirmed by specific magnetic relaxation experiments. We believe that our results can be explained in terms of the flux compression state modulated by pinning effects due to Nd422 particles embedded into the superconducting matrix, similar to experimental results observed in YBa$_2$Cu$_3$O$_{7-\delta}$ superconductor [12].

2. Sample preparation and magnetic measurements

Two melt-textured NdBa$_2$Cu$_3$O$_{7-\delta}$ samples, labeled Nd1 and Nd2, were prepared by Bridgman technique with Nd422 inclusions in a vertical furnace employing a thermal gradient of 4 °C/m and using a constant dragging speed of 1 mm/h. The samples were cut out from the melt-processed materials into the form of small cylinders with 2 mm height and 2 mm in diameter. The c-axis was oriented along the cylinder height.

The magnetic measurements were performed either in a superconducting quantum interference device (SQUID) and in a vibrating sample magnetometer (VSM), both by Quantum Design. Magnetic fields from 0.05 up to 14 T were applied parallel to the $ab$ planes according to the zero field cooling (ZFC), field-cooled cooling (FCC) and field-cooled warming (FCW) procedures. In the ZFC procedure the sample is first cooled at zero field and after the magnetic field is turned on and the magnetic moment is measured during the increasing of the temperature. In the FCC procedure, the magnetic field is first turned on and the magnetic moment is measured while slowly cooling the sample in the constant field. In the FCW procedure the magnetic field is also first turned on and the sample is cooled, then the magnetic moment is measured while slowly warming the sample in the same magnetic field. Although the PME would be observed only by FCC and FCW procedures, the ZFC measurements were always performed in order to verify the superconducting character of our samples for all magnetic fields employed. The Figure 1 shows representative ZFC and FCC results for Nd1 sample in a magnetic field of 1 T. In order to study the time dependence among FCC and FCW magnetic moments, specific FC magnetic relaxation measurements were performed in a fixed magnetic field and temperature. Details will be presented and discussed in the next section.
3. Results and discussion

The Figure 2 (a) and (b) shows FCC and FCW results for samples Nd1 and Nd2, respectively. For the sample Nd1 were applied magnetic fields from 0.05 (see inset) up to 5 T, while for the Nd2 sample were applied magnetic fields from 0.5 T up to 14 T. A strong paramagnetic moment may be observed for both samples during FCC and FCW procedures. The PME exhibited by our samples clearly increases monotonically as the applied magnetic field is raised up to 5 T for Nd1 sample as well as up to 14 T for Nd2 sample. A more detailed inspection of the Figure 2 reveals irreversibilities effects, where the magnitude of the FCW magnetic moment exceeds the FCC magnetic moment. This feature can be observed for magnetic fields above 2 T for both samples and is an indication of time effects, since FCC and FCW experiments were performed in temporal sequence, starting from FCC followed by FCW procedures. Similar results were observed in YBa$_2$Cu$_3$O$_{7-\delta}$ superconductors [12,16,17,27]. The inset in the Figure 2 (a) shows a FCW procedure performed in a magnetic field of 0.05 T, where can be observed a conventional diamagnetic response, reinforcing the idea that our PME is dependent of the magnetic field range. For magnetic fields above 0.05 T the superconducting material exhibits a paramagnetic moment, overcoming the usual diamagnetic response. This curious behavior may be found in other works [12,17].

In order to investigate the time effects observed by the irreversibilities FCC-FCW showed in the Figure 2, we have performed specific field-cooled (FC) magnetic relaxation experiments in both samples. In this case the magnetic field was applied above the critical temperature (field-cooled procedure) and the sample was cooled in a predetermined cooling rate to a fixed temperature, then the magnetic moment was measured as a function of time in the constant field.

The Figure 3 (a) and (b) shows FC magnetic relaxation results, observed in the Nd1 and Nd2 samples, respectively. In the Figure (a) a magnetic field of 1 T, a cooling rate of 0.5 K/min and a temperature of 50 K were the parameters employed, whereas in the Figure (b) the same magnetic field was applied, but a 20-fold higher cooling rate was used and the experiment was performed in a fixed temperature of 70 K.
Figure 2. FCC and FCW results for samples (a) Nd1 and (b) Nd2 showing the occurrence of paramagnetic moments. The inset in the Figure (a) shows the diamagnetic response exhibited by Nd1 sample when a low magnetic field was applied.

The results of the Figure 3 are represented in terms of the normalized FC magnetic moment, where $M_0$ is the magnetic moment when $t = 0$ s. For Nd1 sample the paramagnetic moment raises by 15% after 50,000 s, while for the Nd2 sample the paramagnetic response raises by 6% after almost 18,000 s. Note that in both samples and for different parameters the magnetic relaxation is toward higher paramagnetic moments.

Several magnetic relaxation studies have been reported in the literature [5,12,14,27], including a theoretical prediction for paramagnetic relaxation in samples exhibiting the PME [28], and the...
relaxation results showed in the Figure 3 suggest that time effects constitute a relevant characteristic of our samples.

The paramagnetic Meissner effect (PME) can be observed either in low or high magnetic fields with different characteristics. In the low field range, generally few mT, the PME is known as Wohlleben effect. In this case the paramagnetic effect first increases as the magnetic field is raised, but decreases and even vanishes as the magnetic field is raised above few mT [2,18], and then the usual diamagnetism is established. On the other hand, sometimes the PME is observed on from some tenths of Tesla increasing as the magnetic field is increased [12,17,27], exactly as presented by our results. According to some authors [12,13,14] in this case the effect exhibits time effects in which the field-cooled magnetization relaxes toward an increasing value of the FC magnetic moment. However, in spite of the several studies, there isn't a definitive model to explain the PME for high magnetic fields.

The PME for high magnetic fields, as presented here, in our view can be interpreted in terms of the flux compression phenomenon originally proposed by Koshelev and Larkin [22]. According to the models based on magnetic flux compression [22,23,24] a non-equilibrium compressed flux state is generated due to inhomogeneous temperature or inhomogeneous transition temperature.

In our samples an inhomogeneous scenario can be developed during the sample cooling, where the sample boundary cools first and becomes superconducting, therefore trapping the magnetic field inside the sample. Due to the Meissner effect, the magnetic flux can be guided out from the superconducting boundary layer toward both sides of the sample. A part can be expelled out from the sample and another part is push inward toward the internal non-superconducting regions. As the cooling process goes on, the sample cools toward the interior and the magnetic flux gradually will be swept inward and compressed within the superconducting regions. The magnetic flux compressed depresses the local superconductivity forming a non-superconducting or weaker superconducting region in the bulk and giving rise to a paramagnetic moment. This process of magnetic flux sweeping and compression lets behind a low magnetic flux density region, opening space for new vortices from outside the sample. The strong flux pinning is a necessary condition for the flux compression increase into the sample, which can be provided by the Nd422 particles. The time dependence of the PME verified in our samples by FCC/FCW irreversibilities and FC magnetic relaxation measurements can be explained by the fact that the system acquires a non-equilibrium state. The FC magnetic relaxation results show that additional magnetic flux penetrates into the sample producing a non-equilibrium state.

In summary, we have performed magnetic measurements in melt-textured NdBa$_2$Cu$_3$O$_7$ samples grown by Bridgman technique in order to study the paramagnetic Meissner effect. The measurements were performed for magnetic fields up to 14 T parallel to the $ab$ plane following ZFC, FCC and FCW procedures and the time dependence was investigated by FC magnetic relaxation experiments. Our results can be explained in terms of the flux compression mechanism, such as proposed by Koshelev and Larkin [22], and modulated by the pinning effect due to the Nd422 inclusions.

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