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To cite this article: H Deguchi *et al* 2018 *J. Phys.: Conf. Ser.* **1054** 012026

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# Paramagnetic and Glass States of a YBCO Film Containing Nanorods at Low Magnetic Fields

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**Abstract.** The magnetic properties of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> (YBCO) multilayered film containing BaHfO<sub>3</sub> (BHO) nanorods at zero and small fields have been investigated. The dc magnetization and the ac susceptibility were measured with a SQUID magnetometer. The paramagnetic magnetization is shown after the field-cool-warming below the transition temperature  $T_c = 87$  K. The nonlinear susceptibility had a large peak around  $T_c$ , which reflects a glass-transition. An aging effect occurred in the relaxation of zero-field-cooled magnetization below the  $T_c$ . Paramagnetic behavior and magnetic-glass properties were observed in the YBCO multilayered film containing BHO nanorods. The results suggest that the YBCO multilayered film is a novel magnetic-glass system at low fields.

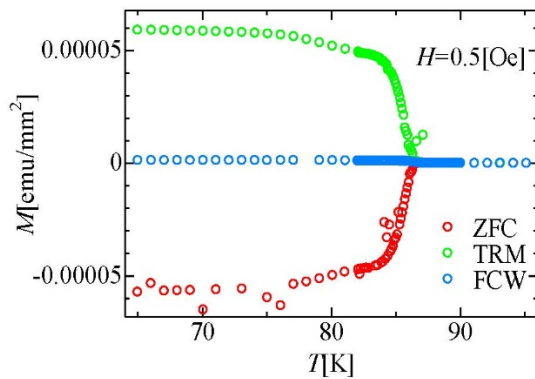
## 1. Introduction

Recently, multilayered films comprising YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> (YBCO) with nanorods as artificial pinning centers have been extensively studied for their flux pinning properties in strong magnetic fields. The transport properties of YBCO films are strongly enhanced by columnar pins, which are produced by the introduction of compound defects, e.g., BaHfO<sub>3</sub>(BHO), BaZrO<sub>3</sub>, and BaSnO<sub>3</sub> nanorods.[1,2] YBCO films with these nanorods effectively improve the critical current density ( $J_c$ ) in strong magnetic fields. High-temperature superconductors exhibit rich phase diagrams with vortex glass, and Bragg and Bose glass phases. They are actively studied to further understand the behavior of the glass-liquid transition.[3] However, there are few experimental studies concerning the magnetic properties of YBCO films that have investigated the low field, which corresponds to the Meissner state of the bulk YBCO. Even at such low field, the magnetic flux can be trapped in the nanorods of the YBCO films, and the vortex state of a small number of vortices confined in the nanorods would be different from that of the bulk samples. In addition, the geometrical disorder due to the random location of the nanorods in the films corresponds to random flux in the glass system such as the chiral-glass one in the ceramic cuprate superconductors.[4-7] In this work, we have investigated the magnetic properties of a YBCO multilayered film containing BHO nanorods at zero and small fields to elucidate the magnetic states of random vortices in the system. Paramagnetic and glass states were observed at low fields below the  $T_c$ .

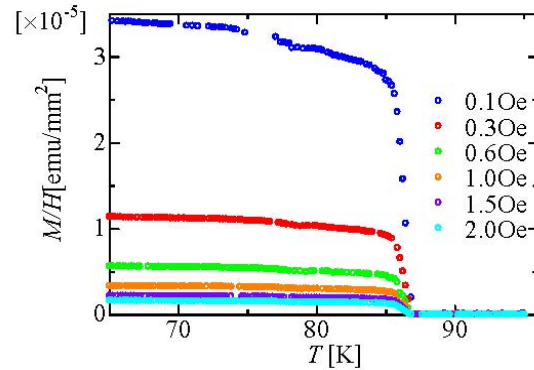
## 2. Experimental

Multilayered films comprising three YBCO layers containing BHO and four pure YBCO layers were prepared on a SrTiO<sub>3</sub> substrate via alternating the ablation of the YBCO stoichiometric target and YBCO-BHO mixed target.[1] The concentration of BHO in the premixed targets is 6 wt%. The thickness





**Figure 1.** Temperature dependence of the ZFC, FCW, and TR magnetization at  $H = 0.5$  Oe.



**Figure 2.** Temperature dependence of the FCW susceptibility at different dc applied fields.

of each layer was 30 nm, and the total film thickness was 220 nm. Transmission electron microscopy was used to characterize the microstructure of the film. The BHO nanorods with 7 nm diameter grew almost parallel to the  $c$ -axis direction of the film.

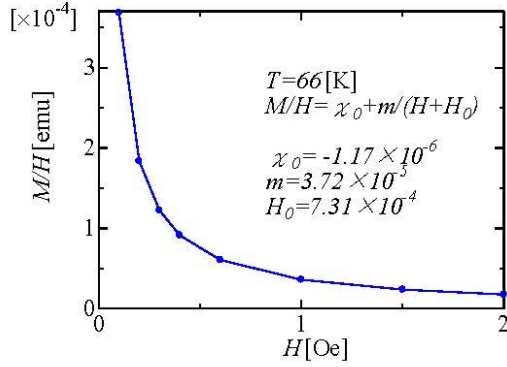
The dc magnetization and the ac magnetic response were measured with a SQUID magnetometer (Quantum Design MPMS-7) using the ultra-low-field option at the temperature sweep mode. The sample space of the magnetometer was shielded with  $\mu$ -metal. As a result, the residual field was reduced to less than 10 mOe. The nonlinear susceptibility was derived from the harmonics in-phase Fourier component for the ac-field response. An external field was applied perpendicular to the film plane. The magnetizations and the susceptibilities were normalized with the in-plane area of the film.

### 3. Results and Discussion

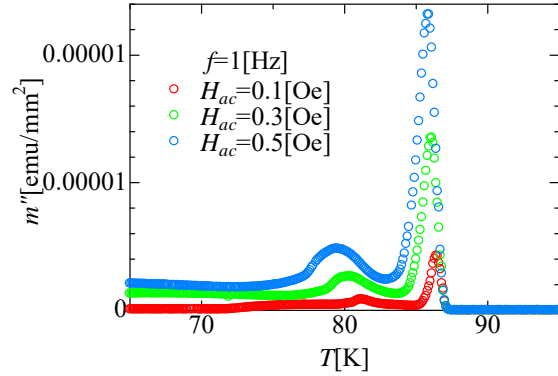
The temperature dependence of the dc magnetization and ac susceptibility was measured to investigate the glass behavior at low fields. Figure 1 shows the temperature dependences of the zero-field-cooled (ZFC), field-cool-warming (FCW), and thermoremanent (TR) magnetizations at  $H = 0.5$  Oe. The transition at  $T_c = 87$  K was identified as the superconducting ordering, in which diamagnetism due to the Meissner effect appears in ZFC magnetizations. Moreover, the FCW magnetization was much lower than the ZFC one and was positive below the  $T_c$ . The paramagnetic signal of the FCW magnetization is known as the paramagnetic Meissner effect (PME) or Wohleben effect.[8] Magnetic-glass behaviors such as the presence of TR magnetization and the discrepancy between the FCW and ZFC magnetization at the  $T_c$  were observed at low field. Below the  $T_c$ ,  $M_{ZFC} - M_{FCW} = M_{TRM}$  is adequately satisfied for all temperatures.

In Figure 2, the FCW susceptibilities ( $M_{FCW}/H$ ) are plotted as a function of temperature at dc magnetic fields between 0.1 and 2.0 Oe. Below the  $T_c$ , the FCW susceptibility increased with decreasing field and decreasing temperature, and was nearly constant at low temperatures. The field dependence of the FCW susceptibilities at 66 K is shown in Figure 3. The FCW susceptibilities at 66 K could be fitted using the equation  $M_{FCW}/H = \chi_0 + m/(H+H_0)$ , where  $\chi_0 = -1.17 \times 10^{-6}$  emu,  $m = 3.72 \times 10^{-5}$  emu and  $H_0 = 7.31 \times 10^{-4}$  Oe. Such field dependence is observed in ceramic samples of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Bi2212) superconductors, and suggests a paramagnetic Meissner behavior.[9]

Next, the temperature dependence of the in-phase ( $m'$ ) and of out-of-phase ( $m''$ ) ac magnetic response was measured at a frequency of 1 Hz and an ac-field amplitude of 0.1, 0.3, and 0.5 Oe under zero external field. The in-phase response  $m'$  abruptly changed at the  $T_c$  and showed diamagnetism below it. This behavior was similar to that of the ZFC dc-magnetization shown in Figure 1. The diamagnetism of  $m'$  saturated below *ca.* 70 K, and the saturated value linearly increased with the ac-



**Figure 3.** Field dependence of the FCW susceptibility  $M_{\text{FCW}}/H$  at 66 K. The line represents the fit using the equation given in the text.



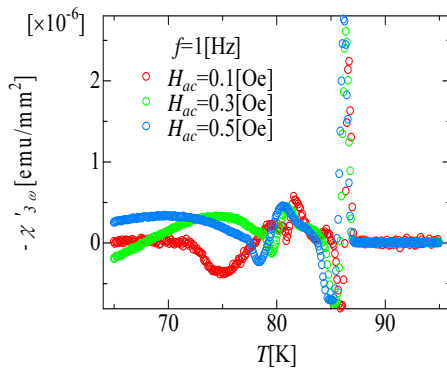
**Figure 4.** Temperature dependence of the out-of-phase ac magnetic responses  $m''$  at a frequency of 1 Hz with an ac-field amplitude of 0.1, 0.3, and 0.5 Oe at  $H = 0$  Oe.

field amplitude, which is due to the Meissner effect resulting from the superconductivity of the film. Figure 4 shows the temperature dependence of the out-of-phase response  $m''$ . The large peak corresponding to  $m''$  was observed at the  $T_c$ .  $m''$  decreased at about 85 K. Then,  $m''$  formed a double maximum. The magnitude of the peak depended on the ac-field amplitude, and the results exhibit dissipation even in the low-frequency region. The nonlinear susceptibilities estimated from the first term of the series of in-phase odd-harmonic responses at 1 Hz with ac-field amplitudes of 0.1, 0.3, and 0.5 Oe are shown in Figure 5. A positive and a negative peak of nonlinear susceptibility were respectively observed around the  $T_c$  and 85 K; the intensity did not depend on the ac-field amplitude. The former peak corresponded to the superconducting transition and the latter to the glass-transition.[4,7]

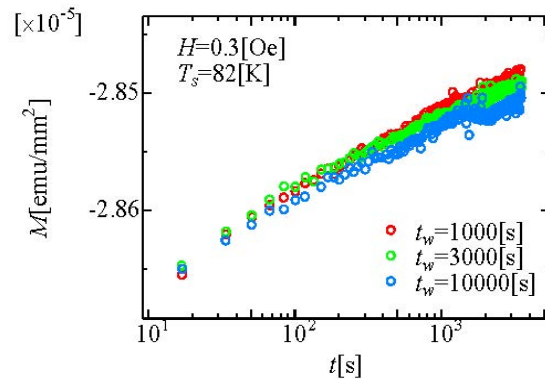
The relaxation of the ZFC magnetization was observed via the following procedures. The sample was cooled down from 100 K above the  $T_c$  to the measuring temperature ( $T_s = 82$  K) under zero field at a cooling rate of 4 K/min; then, it was maintained at the  $T_s$  for a certain time  $t_w$ . The ZFC magnetization was then measured as a function of time under a 0.3 Oe field. The relaxation was logarithmically slow in time and the ZFC magnetization towards the value of the FC one. In Figure 6, the time dependence of the ZFC magnetization measured at  $T_s = 82$  K is shown. Three different waiting times,  $t_w = 1000$ , 3000, and 10000 s, were used. The relaxation curves depended on the waiting time  $t_w$ . The three curves were different, and the relaxation rate decreased with increasing  $t_w$ . Such influence of the waiting time  $t_w$  on the dynamics is called the aging effect, which has been observed for frustrated systems such as spin-glasses [10] and the granular Bi2212 superconductor [11] with the PME. The aging effect of the granular Bi2212 was understood by modeling the polydomain microstructure of the superconducting grains as Josephson-junction networks.

Luzhbin *et al.*[12] investigated the paramagnetic magnetization in field-cooled YBCO thin films. The experimental results of the PME are different from the results of this work. They observed a PME over a wide magnetic-field range ( $5 < H < 10^4$  Oe), and the paramagnetic value of magnetization steadily increased with decreasing temperature, indicating the absence of saturation. However, the magnetic-glass behavior was not observed. They concluded that the origin of the PME was in good agreement with the model of vortex compression proposed by Koshelev and Larkin.[13] Koblishka *et al.*[14] observed the PME in an artificially granular YBCO thin film which was patterned into a hexagonal close-packed lattice of disks with a diameter of 50  $\mu\text{m}$ . The field and temperature dependence of the PME is similar to our results. The authors concluded that the PME response was due to flux trapping effects in the spaces between the superconducting disks.

Li [15] reviewed the experimental results, and the simulations and theories related to the PME and



**Figure 5.** Temperature dependence of the nonlinear susceptibility at a frequency of 1.0 Hz with an ac-field amplitude of 0.1, 0.3, and 0.5 Oe at  $H = 0$  Oe.



**Figure 6.** Observation time dependence of the ZFC magnetization for  $H = 0.3$  Oe at  $T = 82$  K with waiting times of 1000, 3000, and 10000 s.

dynamical phenomena such as the glass behavior and aging effect of ceramic cuprate superconductors at zero and low fields. Our results were consistent with those of said superconductors. Recently, Granato [16] studied the magnetic flux disorder in nanohole thin films via numerical simulation of a Josephson-junction array model. He considered the geometrical disorder owing to the random location of nanoholes in the film as the random flux in the model. Therefore, the vortex due to the random locations of nanorods in the YBCO film may correspond to the random flux in the cuprate ceramic superconductors.

#### 4. Summary

The PME was observed in the FCW magnetization of a YBCO film containing BHO nanorods below the  $T_c$ . The presence of TR magnetization and the discrepancy between the FCW and ZFC magnetization at  $T_c$  are shown at low field. The large peaks of the out-of-phase response  $m''$  and nonlinear susceptibility were observed around the  $T_c$  at zero-field. The slow relaxation and aging phenomena of the ZFC magnetization were observed at 82 K. All these observations suggest that the YBCO multilayered film has a novel paramagnetic and glass state at low field, which is different from the vortex phases existing above the Meissner state.

#### Acknowledgements

The SQUID measurement was conducted at the Center for Instrumental Analysis, Kyushu Institute of Technology.

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