Meissner state of high-T_c oxide thin films observed by scanning superconducting quantum interference device microscopy

I. Iguchi[a)] and T. Takeda
Department of Physics, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro, Tokyo 152-8551, Japan

A. Sugimoto
National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan

T. Imaizumi, H. Haibara, and T. Kawai
Department of Physics, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro, Tokyo 152-8551, Japan

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The magnetic level of the Meissner state in high-T_c oxide thin films is investigated using scanning superconducting quantum interference device microscopy. We find that the Meissner level observed is not uniquely determined and shifts with the temperature and depends on the doping level of individual oxide films. The result at higher temperature may be interpreted by a grain-coupled model which reflects the granular nature of high-T_c oxides and is useful for evaluation of high-T_c film quality. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1599624]

It is well known that the Meissner state in a superconductor is uniquely determined and the magnetic flux penetrates it in terms of a quantized vortex above the lower critical field H_{c1}.[1] There has been little research on detailed study on the magnetic level of the Meissner state in high-T_c oxide superconductors. The scanning superconducting quantum interference device microscope (SSM) is a powerful tool for spatially detecting small magnetic flux and provides direct information on the magnetic property of materials. The usefulness of the SSM has been demonstrated for both the basic study of high-T_c superconductors[2–7] as well as for their application.[8–10]

In this letter, we report the measurement of the spatial distribution of the magnetic level of the Meissner state for patterned high-T_c thin films by SSM. The magnetic level was found to depend on the carrier doping level and shift with the temperature. This may be interpreted by the granular nature of high-T_c oxide thin films and provides evaluation of them.

The samples were fabricated by depositing $YBa_2Cu_3O_{7-y}$ (YBCO), $Bi_2Sr_2CaCu_2O_y$ (Bi-2212), and $La_{2-x}Sr_xCuO_4$ (LSCO) thin films on $SrTiO_3$ and $LaSrAlO_4$ substrates, respectively, using the pulsed laser deposition technique and patterning them into square or hexagonal shapes of a few 100 μm by the conventional photolithography technique. Nb and Pb films were also deposited using conventional evaporation and sputtering chambers. All high-T_c oxide films were c axis oriented. The film thickness was 150–200 nm for YBCO, Bi-2212, and LSCO, 100 nm for Nb, and 200 nm for Pb films. The YBCO films were near optimally doped, while the Bi2212 and LSCO films were slightly overdoped or underdoped. The observed critical current density was 3×10^6 A/cm^2 for YBCO, 1×10^6 A/cm^2 for Bi2212, and 5×10^5 A/cm^2 for LSCO at 4.2 K. In most samples, two different oxide films were deposited and patterned on the same substrate[11] so that the comparison of the Meissner magnetic levels between them could be made directly. The SSM made use of a Nb superconducting quantum interference device (SQUID) and had a spatial resolution of 5–10 μm limited by an input coil diameter. The SQUID resolution was less than $5\times10^{-6}\Phi_0$ Hz$^{-1/2}$. Details are described elsewhere.[5,6,12] A small magnetic field normal to the film surface could be generated by a coil wound around the sample.

Figure 1(a) shows an example of the observed three-dimensional magnetic image of the patterned LSCO (T_c $=30$ K) and YBCO ($T_c=90$ K) film domains fabricated on the same $LaSrAlO_4$ substrate. It was recorded at 3 K under an external magnetic field of 1 μT in a window of 400×200 μm^2, in which two flat domains [on the right LSCO film (part of the hexagonal shape is seen), on the left YBCO film] with several quantized vortices trapped during the cooling process are Meissner domains. The region in-between the two domains where no film exists corresponds to the external field level. The enhanced magnetic field around the patterned edge regions shows that the Meissner state really expelled the external magnetic field at the film edge and the magnetic level inside the film was quite flat within the spatial resolution of the SSM. Although the observed vortices appeared to be quite large due to the spread of vortex flux in free space, their size in the film is about an order of London penetration depth. In fact, the simulation yielded almost the right size vortex measured at pickup coil height of 3–4 μm.

Figure 1(b) shows three-dimensional SSM images of the patterned LSCO ($T_c=24$ K, right) and YBCO ($T_c=90$ K, left) film domains at 3 K under an external field of 2 μT in a window of 400×300 μm^2, in which the difference in Meissner level height between the two oxides appeared remarkable. The LSCO film here was less underdoped compared with that in Fig. 1(a). Note that the degree of doping level for LSCO films was estimated by the T_c, c axis length and the

a)Electronic mail: i-iguchi@ap.titech.ac.jp

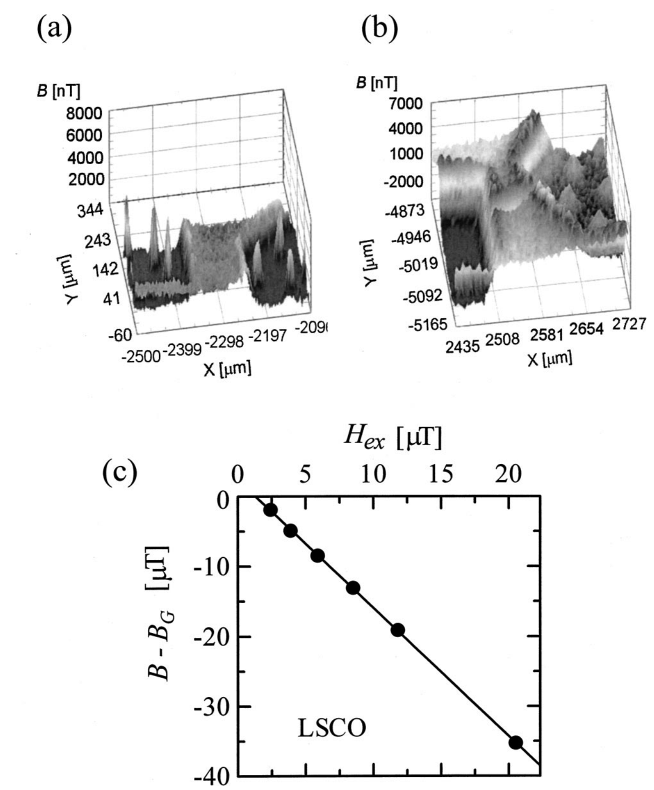

FIG. 1. (a) Three-dimensional magnetic image of two Meissner domains with trapped quantized vortices for optimally doped YBCO (left, $T_c=90$ K) and slightly underdoped LSCO (right, $T_c=30$ K) film domains at 11 K in a window of 400×200 μm^2. The local magnetic field at the patterned edges was enhanced due to expulsion of the magnetic flux. (b) Three-dimensional magnetic image of two Meissner domains for optimally doped YBCO (left, $T_c=90$ K) and underdoped LSCO (right, $T_c=24$ K) films at 3 K in a window of 300×300 μm^2. In this sample, the two Meissner levels were different even at the lowest temperature. (c) Meissner level shift as a function of the external magnetic field at 3 K for the LSCO film. The linear relation exhibits clear diamagnetic behavior up to at least 20 μT.

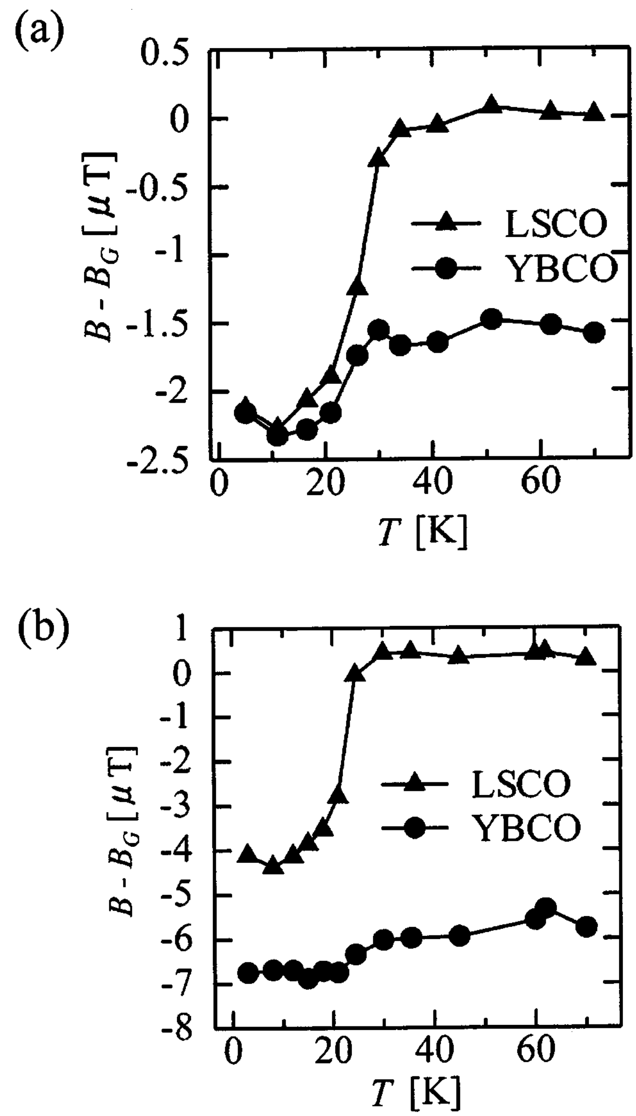

FIG. 2. Temperature dependence of two Meissner levels measured from the external field level of the two YBCO/LSCO samples in Fig. 1.

resistivity versus temperature curve. Four vortices were visible in the LSCO film. When the external magnetic field was varied, the Meissner level changed without any change in the vortex state in these domains. Figure 1(c) shows such an example for the LSCO film. With an increase in the magnetic field from 2 to 20 μT at 3 K, the Meissner level measured from the external field level became deeper, indicating stronger magnetic expulsion. This process was quite reversible. In this experiment, H_{c1} was found to be greater than at least 20 μT.

Figures 2(a) and 2(b) show plots of the Meissner level height measured from the external field level obtained by averaging the data in the 30×30 μm^2 area as a function of the temperature for the two samples in Fig. 1. The standard deviation of these data points was about 10%. Surprisingly, both Meissner levels shifted continually as the temperature was increased. However, the trapped vortices remained present up to very close to T_c. We have also measured the temperature-dependent Meissner shift for Bi2212 and YBCO films fabricated on the same substrate. The observed results were qualitatively similar, and the shift of the Meissner level with the temperature is a common phenomenon among high-T_c oxide superconductors. It is emphasized here that it occurred uniformly in space, not in terms of penetration of quantized vortices above H_{c1}.

In the conventional sense, the Meissner level would not shift while the film is in the superconducting state. We note that, in raising the temperature, the external field was kept at constant value, and the sample–pickup coil distance was kept unchanged. The possible thermal expansion effect of the SQUID apparatus was investigated by independent measurement of trapped vortices in a single YBCO film of good quality and it was found that the sample–pickup coil distance was unchanged up to 70 K.

Figure 3 shows the Meissner level height measured from the external field level as a function of the normalized temperature T/T_c for various kinds of superconductors. For YBCO oxide films with optimal doping, the shift of the Meissner level with the temperature was rather small but quite evident. On the other hand, for the Nb ($T_c=8.5$ K) metal film, it was not recognized up to very close to the vicinity of T_c, consistent with perfect diamagnetism. Similar behavior was also observed for the Pb films.

Now we try to interpret the observed results. First, because of high critical current density J_c, the argument based on the pinning force in the grain boundary and the formation of a certain critical state[13] under very low field is not likely

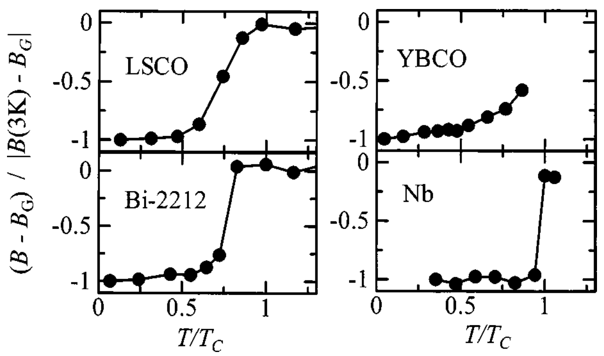

FIG. 3. Temperature dependence of the Meissner level measured from the external field level normalized by the value at 3 K for various cuprate oxides (YBCO, Bi-2212, and LSCO) and Nb metal. For all high-T_c oxide superconductors, the Meissner levels were found to shift with the temperature, while that of Nb was almost constant except very close to the vicinity T_c.

since it gives a negligibly small magnetic penetration depth[14] and a perfect Meissner state almost everywhere.

The other possibility relies on the granular network model in which the film consists of the aggregation of many small grains. The magnetic field penetrates the grain boundaries and this field will be shielded by distributed superconducting grains. The grain size would be at least smaller than a few μm, possibly around 1 μm by considering the flatness of the Meissner plane observed. Because of high J_c, the scaling parameter in the grain boundary would be characterized by the London penetration depth λ rather than by the Josephson penetration depth. Inside the film, the small shielding current loops in the grains cancel each other out, leaving a shielding current around the film edge which yields the magnetic expulsion behavior shown in Fig. 1, just like in the case where the aggregation of many small magnetic dipoles only yields a circulating current around the fringe of a magnetic body. The observation of strong Meissner repulsion signal indicates grain size significantly greater than λ. In this model, the Meissner shift at higher temperature may be interpreted by the change of λ with the temperature. To support this idea, the temperature dependence of λ based on the measurement of the magnetic image of a single vortex in the YBCO film ($T_c = 90$ K) using the SSM is shown in Fig. 4. Below 50 K, it was almost impossible to deduce the λ value from the vortex image observed, but it is considered to be almost independent of the temperature.[4] The rapid increase of λ above 60 K is recognizable. The result qualitatively agrees with that in a previous report.[4] The result above $T/T_c \approx 0.6$ seems to correspond to the temperature dependent behavior of the Meissner shift in Figs. 2 and 3, however, the gradual Meissner shift observed below $T/T_c \approx 0.5$ cannot be

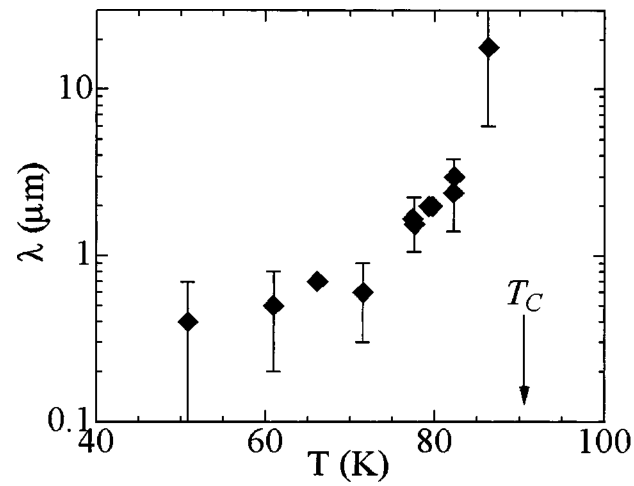

FIG. 4. Temperature dependence of the London penetration depth λ obtained by magnetic imaging of a single vortex in the YBCO film observed by the scanning SQUID microscope.

interpreted simply by this argument. The quantity $[d(B - B_G)/dT]$ depended on the film quality. For the films of rather poor quality, it became large, whereas for the films of good quality, it became small. In the other words, measurement of the Meissner shift provides a method for evaluating the quality of high-T_c oxide thin films.

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