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Low-temperature neutron irradiation effects on superconducting Y-Ba-Cu oxides

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The results of in-pile measurements of low-temperature neutron irradiation (20 K, fast neutron dose 5.9×10^{16} n/cm^2) for two different high-T_c Y-Ba-Cu oxide superconductors are reported. A broadening of the 60- and 90-K superconducting transitions and a drastic increase in resistivity near 200 K are observed.

Since Bednorz and Müller[1] reported the existence of oxide compounds with a high superconducting transition temperature T_c, extensive work has been done on high-T_c oxides. The finding of Y-Ba-Cu oxides having a T_c above 90 K by Wu *et al.*[2] offered the possibility of operating superconducting devices at liquid-nitrogen temperatures. At present, our knowledge of the high-T_c oxides has been rapidly expanded and it is known that the characteristics of the superconducting properties of the Y-Ba-Cu-O system vary depending on the composition and the heat treatment.[3,4] The structure showing the high T_c has been determined to be an oxygen-defect perovskite with an orthorhombic structure containing oxygen-poor layers.[5,6] The oxygen-poor layer structure is considered to be related to superconductivity of the oxide compounds. It is important to study the sensitivity of the Y-Ba-Cu oxides to radiation if we expect to use the oxides as high-T_c superconductors in high-field magnets in fusion reactors. Our particular interest is due to the behavior of the superconductivity of the oxides under neutron irradiation at low temperature and during recovery of defects induced by neutron irradiation. The aim of this communication is (i) to show our recent results about neutron irradiation effects on the superconducting properties of the Y-Ba-Cu oxide, and (ii) to discuss tentatively the possibility of a higher-T_c component in this system.

For the present experiment, two kinds of Y-Ba-Cu oxide samples were prepared from sintered bulk materials. The different superconducting characteristics were obtained by heat treatments under different conditions. The sinterings of a mixture of Y_2O_3, $BaCO_3$, and CuO (cation ratio Y:Ba:Cu = 1:2:3) were carried out by solid reacting at 850 °C (3 h) and 950 °C (3.5 h) for sample 1 (S1) and at 950 °C (20 h) for sample 2 (S2), respectively. From the x-ray diffraction data, the surface layer of S1 and the bulk of S2 were determined to be of the $Y_1Ba_2Cu_3O_x$ orthorhombic structure. However, the inside of S1 was tetragonal structure.

The neutron irradiation experiment was carried out using the low-temperature irradiation loop facility (LTL) at the Kyoto University Reactor (KUR). The two specimens simultaneously set in an inductance coil (1000 turns) were mounted on a boat and irradiated at the top of the horizontal loop. During the low-temperature irradiation the specimens were kept at about 20 K by cooled helium gas circulated from a refrigerator. In-pile measurements of resistivity and susceptibility were performed before and after the irradiation, during the cooling down of the loop, and also during the slow warming up by controlling the gas circulation. The resistance of each sample was independently measured by the four-terminal method using a stabilized dc source and a digital voltmeter, however the inductance was simultaneously measured at an ac frequency of 4 kHz using a LCR meter.

Figures 1 and 2 show the results of resistivity measurements for samples 1 and 2, respectively. S1 had a relatively high resistance (1.5 Ω at 300 K) and clearly showed two stages in the superconducting transition before the irradiation, i.e., a small drop at 85 K and a large drop at 57 K in the resistivity-temperature curve. No detectable change in susceptibility was observed because of the small volume fraction of the surface-layer superconductor in S1. After the low-temperature irradiation [fast neutron dose $=5.9\times10^{16}$ n/cm^2 ($E > 0.1$ MeV), thermal-neutron dose $=7.0\times10^{17}$ n/cm^2, and γ-ray dose $=1.3\times10^9$ R], the transition of S1 became broad, i.e., T_c(onset) appeared at a slightly higher temperature (88 K) and T_c(offset) at a lower temperature (37 K), as seen in Fig. 1. In the voltage-current curve measured at 16.5 K after the low-temperature irradiation, no change was detected until 550 A/cm^2 but a gradual increase appeared over 710 A/cm^2. Such a low critical current I_c may be caused by thin and unstable superconducting components formed near the surface of S1 resulting in high resistance. After the irradiation, its onset resistivity was reduced to two thirds of that before the irradiation, and its resistance increased monotonously, the same as before the irradiation with increasing temperatures until it reached 1.2 Ω at 300 K.

In Fig. 2 the behavior of S2, which had a very low room-temperature resistance (3.8×10^{-2} Ω at 300 K), is shown. The small closed circles indicate the resistivity-temperature curve obtained by out-of-pile measurements for S2. The unirradiated sample shows a sharp transition at 93 K in resistivity and at 92 K in susceptibility. There is some instability of conductivity in the temperature range between 190 and 270 K, although no peak was observed in susceptibility. After the low-temperature irradiation the 93-K transition appeared to be very gradual and obscure. The voltage-current curves obtained at 22 and 130 K showed similar characteristics and indicated $I_c=1430$ A/cm^2 at 22 K and 1070 A/cm^2 at 130 K. However, the resistivity increased rapidly at 195 K and showed a large peak in the current at 10 mA. Below 195 K (higher than 130 K) an instability appeared showing

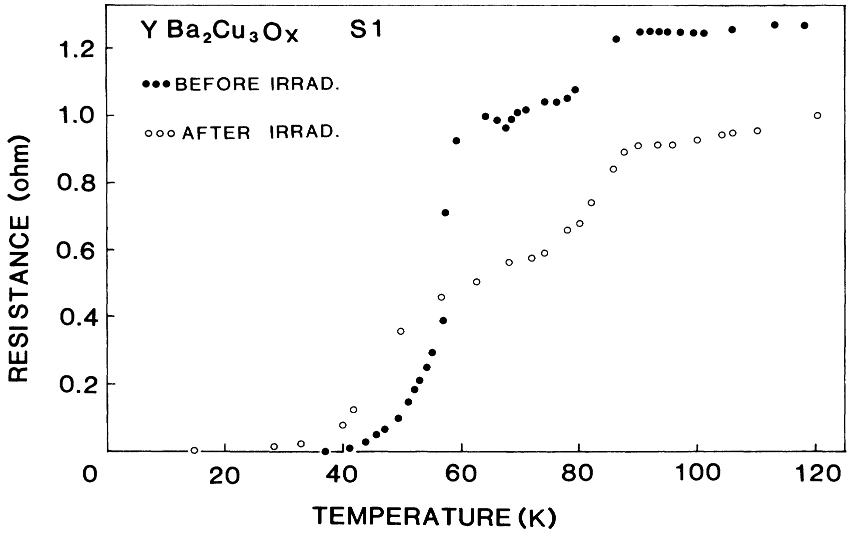

FIG. 1. Superconducting transition curves of Y-Ba-Cu oxide (S1) obtained by in-pile resistivity measurements before and after the low-temperature neutron irradiation (fast neutron dose $=5.9\times10^{16}$ n/cm^2, thermal-neutron dose $=7.0\times10^{17}$ n/cm^2, and γ-ray dose $=1.3\times10^9$ R).

higher values of resistivity when the measuring current was higher than 100 mA. The drastic change was confirmed by repeating the resistivity measurement, however, no large change was observed in the susceptibility because of its smaller sensitivity to a small volume fraction. This fact suggests that the drastic change in resistivity is related to the formation and breakdown of local paths rather than arising out of the bulk structure.

The broadening of the superconducting transition can be understood if we consider the oxygen-poor layers to be locally damaged in atomic-collision sequences. A similar tendency has been reported by Clark, Marwick, Koch, and Laibowitz[7] in the case of ion implantation at room temperature into thin films of Y-Ba-Cu oxides, where the electrical properties rapidly changed under radiation damage induced by 500-keV oxygen ions and the T_c(offset) rapidly decreased at a much higher rate than the T_c(onset). The T_c(offset) was almost destroyed at a dose of 12×10^{13} ions/cm^2 in their results. According to an analogy from results[8] for *A*15 superconductors, the

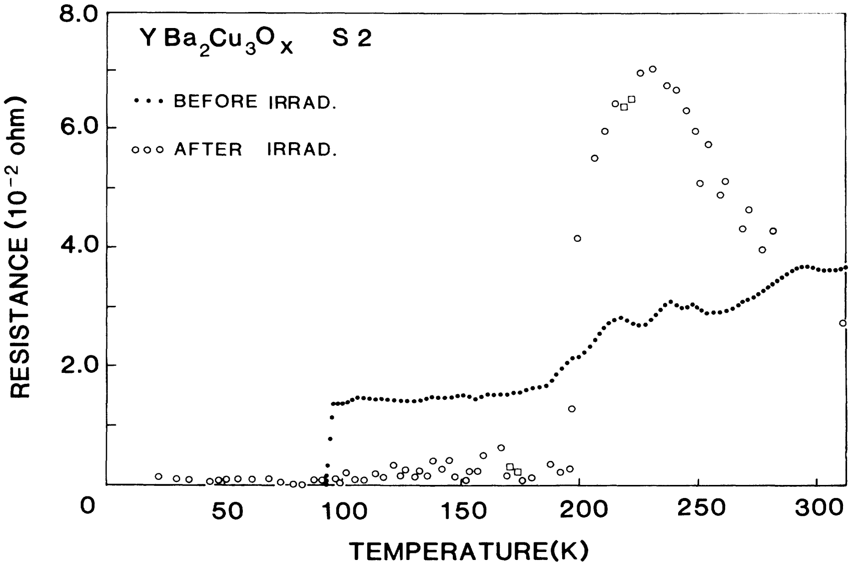

FIG. 2. Comparison of resistivity-temperature curve obtained for Y-Ba-Cu oxide (S2) by in-pile measurement after irradiation with that before irradiation. The irradiation conditions are the same as Fig. 1. Data points by repeated measurement are indicated by open square (□).

broadening of a superconducting transition due to neutron irradiation is understandable because the radiation-induced damages produce local difference in structure and composition. In the case of the oxides, the oxygen vacancies may be preferentially produced by neutron irradiation and the high mobility of radiation-induced defects may be the origin of the sensitivity of the oxide superconductors to radiation. Although we do not yet have a good explanation for the drastic change in resistivity appearing in S2 after the low-temperature irradiation (Fig. 2), it seems to be related to the instability of conductivity in the unirradiated state, and may be caused by the excitation of some intrinsic unstable phase or by the increment of oxygen defects due to low-temperature neutron irradiation.

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