Critical sheet resistance observed in high-$T_c$ oxide-superconductor Nd$_2-x$Ce$_x$CuO$_4$ thin films

Satoshi Tanda, Minoru Honma, and Tsuneyoshi Nakayama

Department of Applied Physics, Hokkaido University, Sapporo 060, Japan
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High-$T_c$ oxide-superconductor Nd$_2-x$Ce$_x$CuO$_4$ single-crystal thin films were used to observe the critical sheet resistance at the insulator-superconductor transition. The films were epitaxially grown on SrTiO$_3$(100) by the method of molecular-beam epitaxy. The electron transport properties of these films are drastically influenced by oxygen-impurity concentration and show clearly the evidence of Anderson localization attributed to two dimensionality. It is found that the critical sheet resistance $R_{\Omega}$ at the onset of superconductivity takes a value within a range from 6 to 8 k$\Omega$, which is close to the value $h/4e^2=6.45$ k$\Omega$. Our results provide clear evidence that Anderson localization induces the universal behavior observed at the superconductor-insulator transition.

Recently there has been considerable interest in the physical origin of the insulator-superconductor transition in disordered thin films at very low temperatures. One of the reasons for this interest is the observation that the onset of superconductivity appears to occur when the normal-state sheet resistance $R_{\Omega}$ falls below a critical value close to $h/4e^2=6.45$ k$\Omega$, between superconductivity and electron localization in a disordered two-dimensional (2D) system; this critical value may be universal, independent of materials and microscopic structures. Granular thin films or homogenous ultrathin films have been used to observe the critical sheet resistance $R_{\Omega}$ since the superconducting transition temperature $T_c$ can be suppressed to very low values by changing the sizes of the grains or film thickness. The ultrathin films of high-temperature oxide superconductors have been also used to observe the critical sheet resistance.

In this paper, we report measurements of the critical normal sheet resistance $R_{\Omega}$ using oxide-superconductor Nd$_2-x$Ce$_x$CuO$_4$ single-crystal thin films prepared by the method of molecular-beam epitaxy (MBE). It is found that the critical sheet resistance $R_{\Omega}$ takes a value between 6 and 8 k$\Omega$. Our measurements were made by varying gradually the oxygen contamination in single-crystal thin films by heat treatment. This is quite different from earlier reports for ultrathin amorphous films or granular thin films. We discuss the relation between earlier reports and our results for the thin films of oxide superconductor Nd$_2-x$Ce$_x$CuO$_4$.

Nd$_2-x$Ce$_x$CuO$_4$ single-crystal films (5 mm x 5 mm x 1000 Å, $x=0.15$--0.20) were grown on SrTiO$_3$(100) by the method of MBE using Knudsen cell sources for Nd, Cu, and Ce. Oxygen pressure partial was $10^{-4}$ Torr in the processing chamber. The details of our MBE method will be reported elsewhere. The crystal structures of the films were characterized by x-ray-diffraction analysis. The appearance of only (002)$n$ sharp peaks indicates that the (001) plane is highly oriented parallel to the film surface, i.e., that the films were grown epitaxially on SrTiO$_3$(100). The lattice constant $c$ was derived from the reflection signals: $c=12.07$ Å ($x=0.15$) and $c=12.06$ Å ($x=0.18$), close to those of Nd$_2-x$Ce$_x$CuO$_4$ ceramics. The film thickness, determined from the cross section of the films by the electron microscope, was 1000 Å within an accuracy of a few percent.

In order to obtain excessively oxidized samples, the films were heated up to 950°C in air and kept at that temperature for 120 min, and quenched down to room temperature. These oxidized films were reduced in Ar (pressure 0.4 Torr, with O$_2$ pressure of less than $10^{-6}$ Torr) at temperatures ranging from 500 to 700°C for 20 min. This annealing in vacuum plays a role in reducing the oxygen concentration in Nd$_2-x$Ce$_x$CuO$_4$ films, which is required for the appearance of the superconductivity. We have measured the resistivities down to liquid-He temperature at various stages of oxygen reduction, and find that the films show the insulator-superconductor transition under some conditions of oxygen contamination.

The temperature dependences of resistivities $\rho(T)$ were measured by the standard four-terminal method with evaporated gold electrodes. The current terminals were covered with gold electrodes along the edge of the films in order to eliminate the ambiguity due to inhomogeneous current flow arising from strong anisotropy. The current density was taken as 10 A/cm$^2$ throughout our measurements. The sheet resistances $R_{\Omega}$ per CuO$_2$ layer were obtained using the relation $R_{\Omega} = \rho/d$, where $d=(6.03$ Å) is the lattice spacing between CuO$_2$ layers in Nd$_2-x$Ce$_x$CuO$_4$ film ($x=0.18$). This relation is based on the assumption that each CuO$_2$ layer forms a twodimensional superconducting sheet. The assumption is reasonable because the resistivities along the basal plane $\rho_{ab}$ and along the $c$ axis $\rho_c$ are highly anisotropic, with a ratio that becomes $\sim$1000. In addition, the validity of the assumption will become clear from our confirmation that the normal-states transport properties of the films show the typical characteristics of weak localization associated with two dimensionality (see Figs. 2 and 3). The sheet resistances $R_{\Omega}$ for Nd$_2-x$Ce$_x$CuO$_4$ films are plotted in Fig. 1 as a function of temperature for two samples ($x=0.18$, $A-E$; $0.16$, $F$) in which oxygen impurities were reduced by the procedure mentioned above. We emphasize here that the curves $A-E$ are the data taken from the same sample but with different oxygen contamination. The point is that the insulator-superconductor transition was observed using the same single-crystal film. Detailed discussion of the data will be given later.

Figure 2 shows the measurements of magnetoresis-
FIG. 1. Temperature dependence of the sheet resistance $R_{\square}$ for Nd$_{2-x}$Ce$_x$CuO$_{4+y}$, single-crystal films. The curves A E are for $x = 0.18$, and F for $x = 0.16$. From A to E, excess oxygen concentration $y$ is varied by changing the annealing conditions: $A$ at 500°C, $B$ at 550°C, $C$ at 600°C, $D$ at 650°C, and $E$ at 700°C, respectively, for 20 min in a vacuum ambient (Ar pressure of 0.4 Torr and O$_2$ pressure of less than 10$^{-6}$ Torr). Curve F shows the data with high transition temperature $T_c$ in a series of our experiments.

FIG. 2. Magnetic-field dependence of resistance $\Delta R(H)/R(H=0)$ at $T = 4.2$ K, in which magnetic fields $H$ are applied normally to the film surface. These data are obtained from the sample corresponding to curve $B$ in Fig. 1. The experimental points are fitted to expression (1) using the values of parameters given in the text.

\[ \Delta R(H)/R(H=0) = A H^2 + B H^4 \]

\[ A = -2 \alpha e^2/\pi^2 \hbar \left( \psi \left( \frac{1}{2} + \frac{1}{a \tau} \right) - \psi \left( \frac{1}{2} + \frac{1}{a \tau_e} \right) - \ln \left( \frac{\tau_e}{\tau} \right) \right) \]

Here $\alpha$ is the constant prefactor, $\tau$ the relaxation time due to normal impurity scattering, $\tau_e$ the inelastic scattering time, and $a = 4DeH/h$ where $D$ is the diffusion coefficient. This formula was derived under the condition $a \tau < 1$, where $a = 4DeH/h$. The solid curve in Fig. 2 was determined using Eq. (1) from least-squares fitting. Best fitting were obtained by choosing $\tau/\tau_e = 0.05$ and $a \tau_e = 0.15$, respectively. Thus, we see that formula (1) is applicable for our case because $a \tau = 0.75$.

In Fig. 1, the change from the insulating state ($R_\square$ increase as $T \to 0$) to the superconducting state ($R_\square$ decrease as $T \to 0$) occurs between 6 and 8 kΩ, close to a value $h/4e^2 = 6.45$ kΩ. Whether or not the $R_\square(T)$ data fall to zero just after this transition is not possible to resolve by the present measurements which were limited to a range of temperatures above 4.2 K.

Our observations of the superconductor-insulator transition naturally indicate that the phenomenon is related to the competition between electron localization and superconductivity. There are two length scales characterizing
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FIG. 3. Angular dependence of the transverse magnetoresistance for the normalized negative part $\Delta R(H)/R(H=0)$ for curve B in Fig. 1 at $H=60$ kOe and $T=2.2$ K, where $\theta=0$ at $H \perp$ surface. The solid curve is only a guide for the eye.

our systems. These are the superconducting coherence length $\xi$ and the electron localization length $l_e$ due to the random distribution of impurities (oxygen in our case): $l_e = l \exp(\pi k_F l/2)$, where $l$ is the mean free path. In our system the latter can be regarded as the control parameter for the onset of superconductivity. We can estimate the order of the mean free path $l$ as follows: It is believed that oxygen impurities are a key element for electron localization for Nd$_{2-x}$Ce$_x$CuO$_4$, in which excess oxygen impurities are reduced by heat treatment. These excess oxygen impurities occupy different sites from those at the superconducting CuO$_2$ layer and play a role as impurity potentials. Though the definite position of excess oxygen impurities is not crucial in the present work, the reasonable positions seem to be those as close as possible to CuO$_2$ layers. It is known that the largest value of excess oxygen concentration is around $y = 0.04$ (Nd$_{2-x}$Ce$_x$CuO$_{4+y}$). From this value, we can estimate the smallest mean free path due to elastic scattering as $l \sim 20$ Å, where use is made of the relation $a^*/l^* = 0.04$, and the lattice constant is taken as $a = 3.95$ Å. As oxygen impurities are reduced by heat treatment, the mean free path $l$ increases, i.e., the localization length $l_e$ increases exponentially and crosses over the coherence length $\xi(T)$ at some value of oxygen concentration. It is natural to claim that, when $l_e$ is smaller than the average size of a Cooper pair ($\xi$), the electrons exist as localized states and the pairs are broken due to the effect of Coulomb repulsion and vice versa. Thus the criterion for the onset of superconducting state is $l_e = \xi$. It should be noted that the effect of disorder on superconductivity has been intensively discussed in connection with Anderson localization by Maekawa and Fukuyama.

In the case of ultrathin films, the length scale $l$ is the average size of the superconducting grains. When the size of grains is larger than $\xi_0/(k_F l)^{1/2}$, the system is viewed as consisting of randomly coupled Josephson junctions, where each grain is described by a single order parameter. Most experiments for granular thin films reported so far$^{1-4}$ utilize the fact that the grain sizes can be artificially varied by choosing the appropriate deposition condition.

To summarize, we have observed the critical sheet resistance using oxide-superconductor Nd$_2$-Ce$_x$CuO$_4$ films, which were epitaxially grown on SrTiO$_3$(100) by the MBE method. Thermal treatment of films gradually reduced the oxygen-impurity concentration and resulted in the appearance of the superconducting state. It is remarkable that our system with high impurity density shows clear evidence of Anderson localization originating in two dimensionality. Oxide-superconductor Nd$_{2-x}$Ce$_x$CuO$_4$ thin films are particularly advantageous for the observation of the critical sheet resistance, because these have two-dimensional CuO$_2$ superconducting layers and the impurity (oxygen) density can be continuously varied by heat treatment. This implies that one can set up the situation where the localization length $l_e$ crosses over the coherence length $\xi$, i.e., the control parameter for the onset of superconductivity is the oxygen concentration. There are a variety of theories to explain the critical sheet resistance observed at the onset of superconductivity.$^{18-24}$ Among them, the theories assuming Josephson coupling between superconducting islands will be ruled out because such a picture is not appropriate for our system. It seems to be necessary to develop the theory from very general points of view incorporating explicitly the aspect of Anderson localization to explain our observation.$^{21-23,25}$

We conclude that our findings for oxide-superconductor Nd$_{2-x}$Ce$_x$CuO$_4$ films provide an interesting system for revealing the mechanism for the superconductor-insulator transition. In particular, we have presented clear evidence that Anderson localization induces the universal behavior observed at the insulator-superconductor transition. Another important feature is that our results play a role in investigating the nature of electron pairing or the normal-state properties of high-$T_c$ oxide superconductors.

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