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Variable-range-hopping-type transport near the superconductor–insulator transition observed in $Nd_{2-x}Ce_xCuO_4$ thin films

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ABSTRACT

Electrical transport properties of high-T_c $Nd_{2-x}Ce_xCuO_4$ single-crystal thin films were investigated to reveal the conduction mechanism near the superconductor–insulator transition. We find that two different types of localized state exist in the insulator side: one shows typical behaviour or weak localization and the other is the variable-range-hopping (VRH) conduction type. We claim this to be a new type of VRH conduction process with interlayer hopping by Cooper pairs.

The experimental and theoretical study of charged boson transport in two-dimensional (2D) disordered systems has become a rapidly developing field of contemporary solid state physics, particularly since the discovery of high-temperature (high-T_c) superconductors. An important issue is the influence of disorder on boson transport. Variable-range hopping (VRH) conduction due to strongly localized electrons has been studied theoretically by Mott (1968, 1978, 1993), Efros and Shklovskii (1975) and Shklovskii and Efros (1984). The VRH is characterized by an activation-type conductivity, which yields Mott's famous law

$$\sigma(T) = \sigma_0 \exp\left[-\left(\frac{T_0}{T}\right)^{\alpha}\right], \quad \alpha < 1, \tag{1}$$

where the exponent α takes different values depending on the dimensionality and the density of states $g(E_F)$ at the Fermi level E_F. For 2D systems, one expects $\alpha = \frac{1}{3}$ when $g(E_F)$ is almost constant or a slowly varying function and $\alpha = \frac{1}{2}$ when there is a parabolic gap at the Fermi level (Efros and Shklovskii 1975). However, the behaviour for localized bosons has not been revealed either experimentally and theoretically. Here we report evidence of VRH for charged bosons, observed near the superconductor–insulator (SI) transition using high-T_c $Nd_{2-x}Ce_xCuO_4$ single-crystal thin films. We find that two different types of localized state exist in the insulator side: one shows typical behaviour of weak localization ($\sigma \propto \ln T$) and the other is the VRH conduction type due to the formation of Bose-glass states. We claim this to be a new type of VRH conduction with an interlayer hopping process by Cooper pairs in the Bose-glass states.

It is well known that the high-T_c oxide superconductor $Nd_{2-x}Ce_xCuO_4$ is peculiar among Cu-based oxide superconductors with perovskite structures. Ordinary copper oxide high-T_c superconductors have CuO_2 networks with pyramidal or octahedral arrangements, and, in addition, the charge carriers in the normal state are holes. The $Nd_{2-x}Ce_xCuO_4$ system consists of 2D CuO_2 layers with no apical O atoms. The CuO_2 layers form an ideal 2D conducting sheet as verified by the observation that the normal-state transport properties of the single-crystal films show the typical

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characteristics of weak localization associated with two-dimensionality (Anderson 1958, Abrahams, Anderson, Licciardello and Ramakrishnan 1979, Tanda, Honma and Nakayama 1991).

$Nd_{2-x}Ce_xCuO_4$ single-crystal films were grown on $SrTiO_3(100)$ single crystals by molecular beam epitaxy using Knudsen cell sources for Nd, Cu and Ce. After deposition, the O_2 flow to the film surface was stopped immediately and the substrate was cooled from 800°C to room temperature with a background pressure of 10^{-4} Torr. The appearance of only ($002n$) peaks indicates that the (001) plane is oriented parallel to the film surface. These films were reduced with a background Ar pressure of 0·4 Torr with the temperature ranging from 450 to 750°C over 20 min. This reduction in a partial vacuum permits a reduction in the O concentration in the $Nd_{2-x}Ce_xCuO_4$ films, which is required for the appearance of superconductivity. The film thickness, determined from the cross-section of the films by electron microscopy, was 1000 Å within an accuracy of a few per cent.

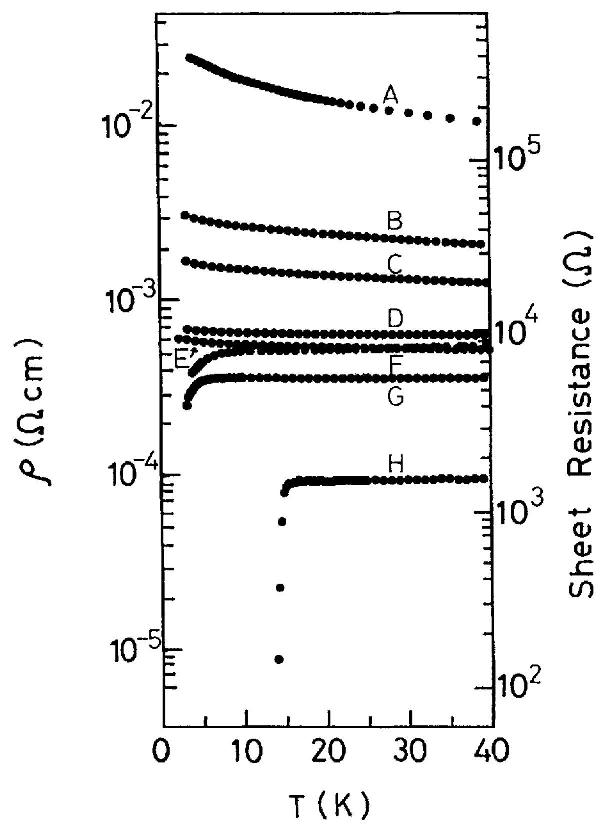
The temperature dependence of the resistivities $\rho(T)$ was measured by the standard four-terminal method with evaporated Au electrodes. The current terminals were covered with Au along the edge of the films in order to eliminate the ambiguity due to inhomogeneous current flow arising from strong anisotropy. The current density was taken as $10\,\mathrm{A\,cm^{-2}}$ throughout our measurement. A sensitive voltmeter (Keithley 182) was used to measure all resistances and magnetoresistances, and 10^{-9} resolution was achieved. The temperature dependences of the resistivities $\rho(T)$ for the $Nd_{2-x}Ce_xCuO_4$ films at various stages of oxygen reduction are shown in fig. 1. The residual resistivity was $75\,\mu\Omega\,\mathrm{cm}$ for sample H ($x = 0{\cdot}16$). This value is small compared with that of bulk single crystals (Hidaka and Suzuki 1989, Hagen *et al* 1992). This is due to the easier reduction of O impurities in thin films compared with bulk samples. These vacuum-annealed films, with optimum reduction, are characterized by a sharp superconducting transition with $T_c(\rho = 0) = 15$ K and a transition width of less that 0·9 K. The curve of sample H ($k_F l = 19$) agrees fairly well with the theory by Azlamazov and Larkin 1968, indicating that 2D superconducting fluctuations are relevant for this less-disordered sample.

O-rich non-superconducting samples (A–C) clearly show behaviour typical of weak Anderson localization in two dimensions (Tanda *et al.* 1991). We find that the temperature dependence of the sheet resistance changes from the $\ln T$ form of weak localization to that for the 2D VRH-type conduction. As seen from the curves E and F in fig. 1, the separation between the insulating state ($R_\square$ increases as $T \rightarrow 0$) and the superconducting state ($R_\square$ decreases as $T \rightarrow 0$) occurs in the range between 8 and 9 kΩ, which is close to the value $h/4e^2$. The sheet resistances (curves C–E) shown in fig. 1 are replotted in fig. 2. One sees from fig. 2 (a) that only curve C follows a $\ln T$ behaviour indicative of 2D fermion weak localization. A remarkable feature is that curves D and E close to the SI transition take the functional form $\exp[-(T_0/T)^\alpha]$ of the 2D VRH type, as seen from fig. 2 (b). This implies that a new type of localized state exists when R_α approaches a value close to $h/4e^2$. The precise value of α was obtained by transforming the expression for the sheet resistance $R_\square = R_0 \exp[(T_0/T)^\alpha]$ into

$$\log\left(\frac{\mathrm{d}(\log R)}{\mathrm{d}(1/T)}\right) = (\alpha - 1)\log\left(\frac{1}{T}\right) + \log(\alpha T_0^\alpha), \quad (2)$$

where $\mathrm{d}(\log R)/\mathrm{d}(1/T)$ corresponds to the characteristic hopping energy or generalized activation energy. In fig. 3 we have plotted $\mathrm{d}(\log R)/\mathrm{d}(1/T)$ as a function of $1/T$ on a log–log scale for sample D. The straight line represents a least-squares fit to the data.

Fig. 1

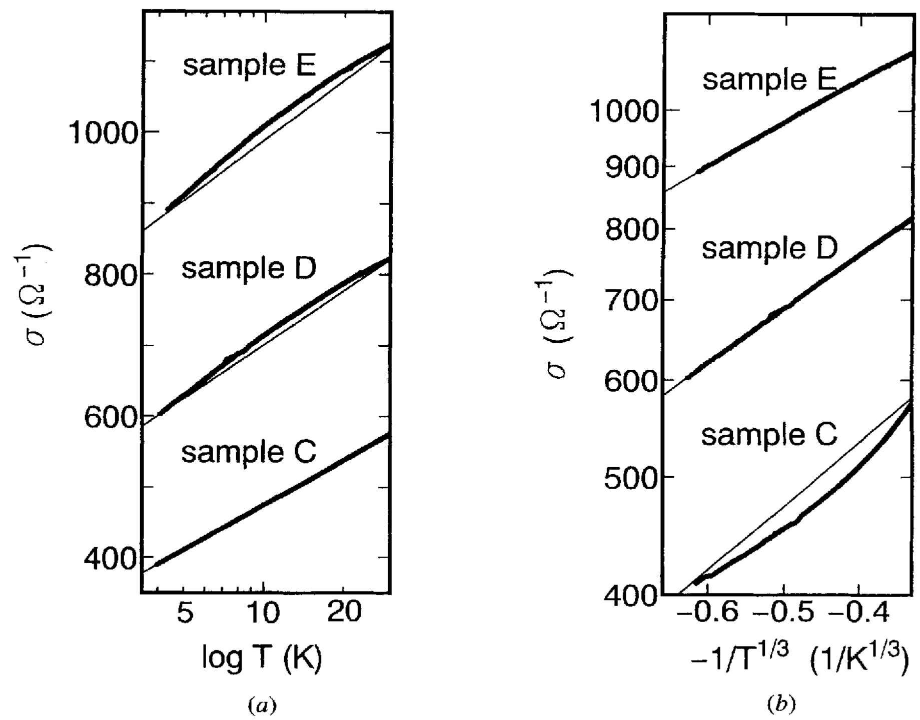


Temperature dependence of resistivities (left-hand scale) for $Nd_{2-x}Ce_xCuO_4$ single-crystal films at various stages of disorder controlled by the annealing process. Curves (samples) A–G correspond to $x = 0{\cdot}18$, and curve (sample) H to $x = 0{\cdot}16$. The temperatures for a 20 min anneal in a partial vacuum were as follows: curve A, 450°C; curve B, 500°C; curve C, 550°C; curve D, 600°C; curve E, 650°C; curve F, 700°C, curve G, 750°C; curve H, 750°C. The right-hand scale refers to the sheet resistance per CuO_2 layer (see text).

The slope m is related to α through the relation $m = \alpha - 1$. The observed value of m gives $\alpha = 0{\cdot}31 \pm 0{\cdot}05$. We have obtained $\alpha = 0{\cdot}33 \pm 0{\cdot}05$ for sample E. Electrical transport in the strongly disordered regime is attributable to VRH. In VRH transport, the value of α depends on both the dimensionality d of the system and the density of states at the Fermi level ($\alpha = \frac{1}{3}$ in two dimensions (Mott 1968)). In addition, α is related to the electron correlation. Altshuler, Lee, Kheml'nitzkii and Larkin (1979) have shown that the density of states has a minimum (but finite) value at the Fermi level in disordered metals. If the disorder increases, the electronic states at the Fermi level are strongly localized. Efros and Shklovskii (1975) have claimed that the minimum becomes a gap (the Coulomb gap) and that the conductivity is described by VRH with $\alpha = \frac{1}{2}$, independent of dimensionality. Our results for samples D and E ($\alpha \approx \frac{1}{3}$) show the characteristics of 2D localization arising from induced disorder and not from the Coulomb correlation effect. We claim in the following that the charged carriers are bosons.

To determine the nature of the charge carriers we investigated the field-tuned

Fig. 2



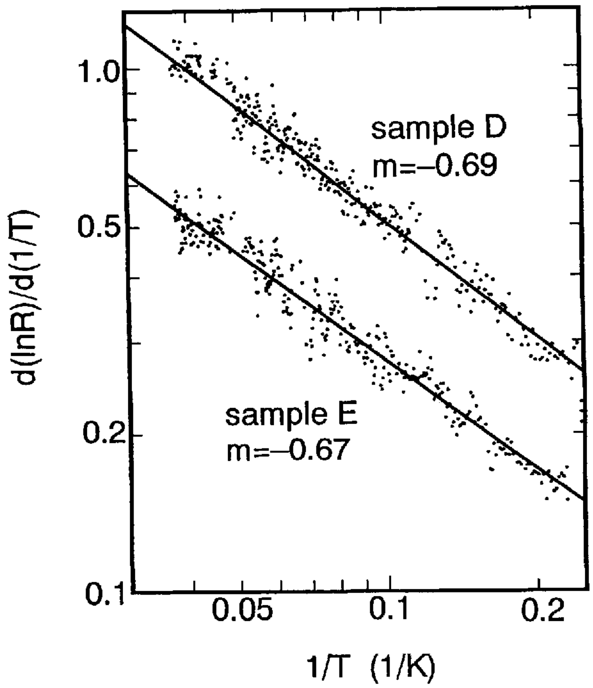
The temperature dependence of conductances σ for sample C, D and E. (*a*) σ against $\log T$ (weak localization); (*b*) σ on a logarithmic scale against $-T^{-1/3}$ (VRH conduction).

transport properties for the superconducting sample F. Figure 4 shows the resistance of sample F measured as a function of magnetic field at various temperatures. Below the critical field $B_c = 2{\cdot}9\,\mathrm{T}$, the resistance decreases with decreasing temperature whereas, above B_c, the resistance increases with increasing magnetic field B. The resistance $R_\square$ at B_c is $8{\cdot}5\,\mathrm{k}\Omega$. According to Fisher's scaling theory, the slopes of the $R_\square(B)$ curves at B_c should follow a $T^{-1/z_B\nu_B}$ power-law dependence. z_B and ν_B are the dynamical critical exponent and the static critical exponent respectively for the superconducting correlation length ξ_B. Scaling theory predicts that the resistances should scale, under a magnetic field B, as (Fisher 1989, 1990)

$$R(B,T) = \frac{h}{4e^2} f\left(\frac{c_0(B-B_c)}{T^{1/z_B\nu_B}}\right), \tag{3}$$

where c_0 is a non-universal constant, where $\xi_B \propto (B-B_c)^{-\nu_B}$ and $\Omega_B \propto \xi_B^{-z_B}$, Ω_B being a characteristic frequency. Near the SI transition, one expects that the length scale ξ_B diverges, which sets ξ_B as the length scale characterizing the system. With increasing magnetic field, the remarkable possibility arises that vortices can become delocalized and undergo a Bose condensation at some critical field B_c. When the slopes of the $R_\square(B)$ curves at B_c are plotted against $1/T$ on a double-logarithmic plot, we find a straight line in the range $1{\cdot}6\,\mathrm{K} \leqslant T \leqslant 8\,\mathrm{K}$. From the reciprocal of the slope of this straight line, we determine $z_B\nu_B = 1{\cdot}2 \pm 0{\cdot}1$ (Tanda, Ohzeki and Nakayama 1992). This value is consistent with theoretical expectations $z_B\nu_B \geqslant 1$. The observed resistivities near the field-tuned SI transition thus provide evidence for a Bose-glass–vortex-glass phase

Fig. 3



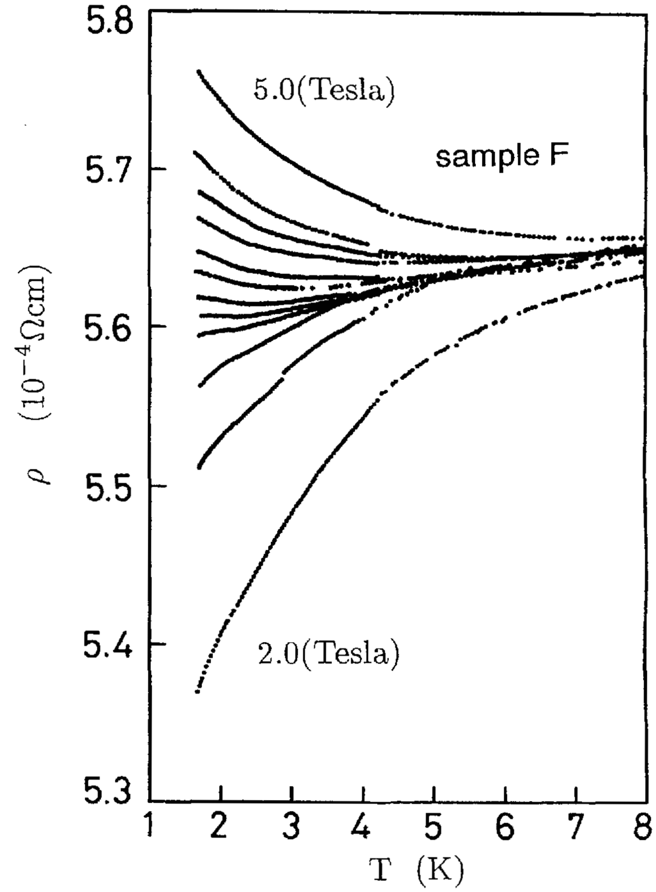
Logarithmic plot of d(log R)/d($1/T$) as a function of $1/T$. The exponents ($\alpha = m + 1$) were extracted from the slope m as $\alpha = 0{\cdot}31 \pm 0{\cdot}05$ (sample D) and $\alpha = 0{\cdot}33 \pm 0{\cdot}05$ (sample E).

transition, indicating that the boson phase is formed on the insulator side of the SI transition.

Resistivities on the insulating side of fig. 4 show a conductance varying as $\exp[-(T_0/T)^\alpha]$, indicating that VRH is the dominant process for transport. In fig. 5 we have plotted d(log R)/d($1/T$) against $1/T$ on a log–log scale for the field-tuned sample F (a field $B = 5$ T was applied perpendicular to the film surface). The straight line represents a least-squares fit to the data. The observed value of m gives rise to $\alpha = 0{\cdot}49 \pm 0{\cdot}05$ and not $\alpha = \frac{1}{3}$ as obtained for samples D and E. This suggests that under high magnetic fields the boson transport occurs along a quasi-one-dimensional path (in which case $\alpha = \frac{1}{2}$).

From the data on samples D and E we estimate $T_0 = 5{\cdot}0$ and $4{\cdot}5$ K respectively. These values are much smaller than those observed for disordered fermion systems such as doped semiconductor materials. According to the expression for the Mott 2D VRH, one has $k_B T_0 = 27/\pi g(E_F)\xi^2$, where ξ is the localization length. By taking into account the experimental value of $\gamma \approx 30\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$ in specific-heat measurements (Ghamaty *et al.* 1989, Tigheza *et al.* 1990), one can estimate $\xi \approx 2000$ Å from the observed T_0. The value of ξ is the same order as that for the weakly localized regime. It is natural that this localization length ξ does not represent the size of Cooper pairs (the coherence length is about 70 Å), but rather the correlation length of the superconducting region, that is the size of the local superconductor. Scaling theory predicts that the characteristic energy T_0 represents the deviation of the parameter characterizing the SI transition from the right of the SI transition, namely $T_0 \propto \delta - \delta_c$, where δ is the parameter characterizing the disorder (Fisher 1990). This suggests that T_0 takes a

Fig. 4



The temperature dependence of the resistivities of sample F under magnetic fields ranging from 2·0 to 5·0 T. The magnetic fields are 2·0, 2·4, 2·6, 2·8, 2·9, 3·0, 3·1, 3·2, 3·4, 3·6, 4·0 and 5·0 T in order of increasing resistivity and are applied perpendicular to the film surface. The critical field B_c of the SI transition is 2·9 T.

sufficiently small value near the SI transition. The above arguments are, however, based on the assumption of ideal 2D VRH. Recently, Chakraverty, Sudbo, Anderson and Strong (1993) suggested that the interlayer tunnelling of Cooper pairs is important for high-T_c superconducting cuprate. If ξ diverges over the sample size, the interlayer tunnelling coupling (Josephson coupling) becomes perfect, and the system has global coherence. When ξ is finite, interlayer hopping between local coherence regions of each layer take place. In this respect, our findings favour the quasi-2D VRH of Cooper pairs.

Acknowledgments

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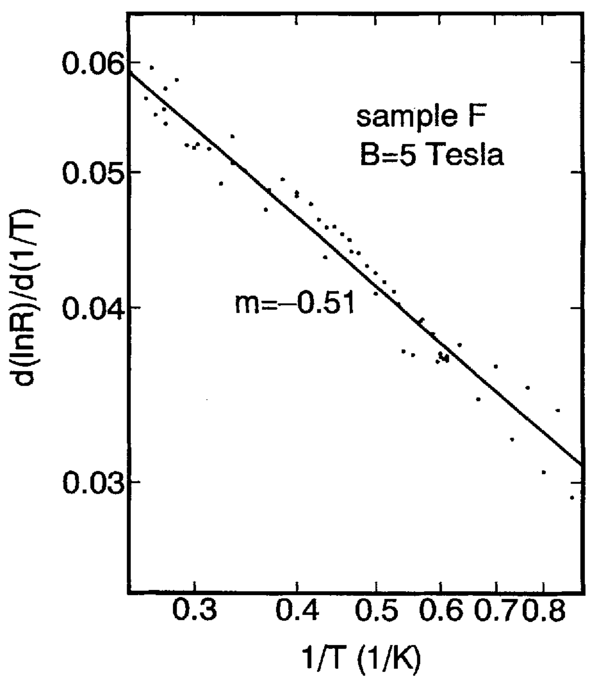
Fig. 5



Logarithmic plot of d(log R)/d(1/T) as a function of 1/T. Th exponent ($\alpha = m + 1$) was extracted from the slope m as $\alpha = 0{\cdot}49 \pm 0{\cdot}05$ (for field-tuned sample F).

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