Dielectric Breakdown of the Insulating Charge-Ordered State in $La_{2-x}Sr_xNiO_4$

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(Received 26 July 1999)

Dielectic breakdown associated with characteristic delay time and subsequent negative differential resistance (NDR) have been observed in the charge-stripe phase of the $La_{2-x}Sr_xNiO_{4+\delta}$ crystal, whose hole doping level n_h $(= x + 2\delta)$ is around 1/3. Threshold voltage ($E_{\rm th}$), above which the NDR emerges, varies with n_h and reaches a maximum at the commensurate doping of $n_h \sim 1/3$. The $E_{\rm th}$ increases exponentially with decreasing T, but tends to saturate at low temperatures perhaps due to the quantum fluctuation of the stripes. The results indicate that the dielectric breakdown is related to depinning and collective transport of the ordered holes on the stripe.

PACS numbers: 71.27.+a

Phenomena of charge ordering in transition metal oxides have been of renewed interest in the light of metal-insulator transitions in correlated electron systems [1]. In particular, the charge ordering in layered structures of cuprates and nickelates shows the vertical or diagonal stripe patterns on the CuO_2 or NiO_2 square lattice, respectively, in which the interplay between magnetic correlation and charge dynamics is of current interest in relevance to the mechanism of high-temperature superconductivity [2]. The diagonal charge and spin stripes in $La_{2-x}Sr_xNiO_{4+\delta}$ and their effects on the magnetic, transport, and optical properties have been investigated extensively in recent years [3–11]. The charge-ordering wave vector for the $x < 0.5$ ground state is approximately proportional with the hole doping level $n_h = x + 2\delta$, although there is observed a clear commensurability effect as locked-in at $n_h = 1/3$ [11]. The ground state remains insulating in all the stripe phases ($x < 0.5$) of the nickelates in contrast to a metallic feature of the vertical stripes in the layered cuprates.

The purpose of this paper is to report on the dielectric breakdown of the insulating charge stripe phase and the subsequent negative differential resistance (NDR) effect in single crystals of $La_{2-x}Sr_xNiO_{4+\delta}$ $(0.25 < x < 0.50)$. We have observed the current switching phenomena associated with depinning of the charge stripe or with collapse of the charge ordering, as well as the current-induced metallic state subsisting down to the lowest temperature. The observed switching phenomena bear some analogy to the field-induced depinning of the charge density wave (CDW) in the low-dimensional system [12], and in fact some classical and quantum hallmarks in the collective carrier motion are observed in the present system. Here, we investigate systematically the hole-doping dependence of the dielectric breakdown and NDR effect in terms of the controlled pinning potential relevant to the aforementioned $n_h = 1/3$ commensurability.

All the samples of $La_{2-x}Sr_xNiO_{4+\delta}$ investigated here are single crystals grown by a floating zone method. The details of the crystal growth procedure as well as the neutron diffraction characterization of the spin/charge order are described elsewhere [7,10,11]. By the iodometric analysis, the hole doping level $(n_h = x + 2\delta)$ of each sample was determined precisely as follows: $n_h(x) = 0.289(0.27), 0.301(0.30), 0.339(0.36), 0.398(0.39), 0.502(0.50)$. The measurements of current-voltage (I-V) characteristics and temperature dependence of resistivity at various applied voltages were performed using the circuit composed of the crystal and the load resistor in series, as shown in the inset to Fig. 1. All the samples are rectangular with a common size: 0.66 mm in length (d), 0.6 mm in width, and 0.3 mm in thickness. Electrodes for a two-probe method were made at each end plane of the specimen using silver paint and hence

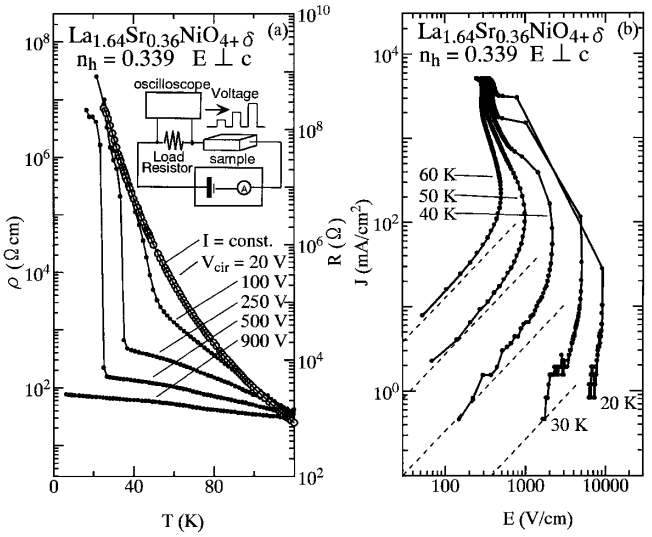

FIG. 1. (a) Temperature dependence of resistivity of a $La_{2-x}Sr_xNiO_{4+\delta}$ $(x = 0.36, n_h = x + 2\delta = 0.339)$ crystal at low constant current (5–100 μA, open circles) and with various applied voltages (closed circles) on the whole circuit, that is shown in the inset. (b) Electric field (E), current density (J) characteristics for an $n_h = 0.339$ crystal at various temperatures. The electric field of the crystal is obtained by removing the voltage biased on the load resistor (see text). Dashed lines represent the Ohmic relation.

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the electrode distance was always $d = 0.66$ mm. The direction of applied field and current was parallel to the NiO_2 plane. A load resistor R_L of 100 $k\Omega$ was inserted to protect the crystal against a burst of current upon the switching. Pulse voltage (usually with 200 msec duration) was applied to minimize the Joule heating effect. Through a voltage drop at the load resistor, temporal response of current and hence the sample resistance were measured by a storage oscilloscope.

Temperature dependence of resistivity ρ (left ordinate) or bare resistance R (right ordinate) is shown in Fig. 1(a). Open circles indicate results by a conventional four-probe measurement with small (5–100 μA) constant current. The resistivity measured with a relatively low (<20 V) voltage is in accord with that of the constant-current measurement. At higher voltages, however, R tends to decrease remarkably, in particular at lower temperatures. At a bias voltage ($V_{\rm cir}$) of 500 V, a discontinuous jump from a low- to a high-resistance state is observed at 25 K. At $V_{\rm cir} = 900$ V, the resistance jump disappears and the low-resistive state subsists down to the lowest temperature, 5 K. The measured ρ or R value at the lowest temperature is observed to be reduced by more than 5 orders of magnitude from the low-voltage R value. Such a large reduction in resistance upon the application of high voltage cannot be ascribed to simple Joule heating. For example, the sample resistance before the switching exceeds 10^9 Ω at 20 K. Application of 900 V on this insulating crystal causes the sudden switching of the resistive state with the delay time less than 10 ms [cf. the inset to Fig. 4(a)], and hence the total power consumed in the crystal prior to the switching is less than 10^{-5} J. Considering a typical value of the specific heat ($\approx$5 J/K mol) at this temperature and the sample volume ($\approx 2 \times 10^{-6}$ mol), the increase of temperature by Joule heating would be less than 1 K, even provided that the sample were thermally isolated within this period ($\sim$10 ms).

Figure 1(b) shows the current density J against the electric field E on the sample with $n_h = 0.339$. E is obtained by the relation that $E = (V_{\rm cir} - R_L I)/d$. The J-E curve shows a remarkable deviation from the Ohmic relation (indicated by dashed lines) and eventually shows a NDR behavior above the threshold electric field $E_{\rm th}$ [13]. With decreasing temperature, the $E_{\rm th}$ increases and the NDR behavior becomes more conspicuous. Such highly nonlinear conduction is observed for other compounds with $n_h = 0.289$–0.502. Phenomenologically, the J-E characteristics for $n_h = 0.339$ can be described using the expression of current-dependent conductance: $\sigma(J) = \sigma_0 + \sigma_1' f(J)$, where $f(J) = J^n$ ($n \sim 1.5$) and σ_0 and σ_1' are temperature-dependent quantities. This formula implies the presence of the self-multiplication (avalanche) process of current or carrier number.

Figure 2(a) shows the hole doping (n_h) dependence of $E_{\rm th}$. Remarkably, $E_{\rm th}$ shows a distinct maximum at $n_h \sim 1/3$, where the charge ordered state is most stabilized. This result indicates that the NDR effect in $La_{2-x}Sr_xNiO_{4+\delta}$ is related to the stability of the charge-ordered state. It is likely that ordered holes are pinned by the commensurate potential derived from the electron-lattice or electron-electron interaction at $E < E_{\rm th}$, while depinned at $E > E_{\rm th}$, resulting in the NDR effect. The $E_{\rm th}$ in $La_{2-x}Sr_xNiO_{4+\delta}$ (10^3–10^4 V/cm) is much larger than a typical value of $E_{\rm th}$ (10^{-2}–10^{-1} V/cm) for conventional CDW pinned by impurity [12], but is still by far smaller than a typical breakdown field ($\sim 10^7$ V/cm) in band insulators [14].

The idea of electric-field depinning of stripes is confirmed by the temperature dependence of $E_{\rm th}$ as shown in Fig. 3. Except at low temperatures, $E_{\rm th}(T)$ obeys the following temperature (T) dependence:

$$E_{\rm th}(T)/E_{\rm th}(0) = \exp(-T/T_0). \qquad (1)$$

The behavior described by Eq. (1) is often observed for CDW systems [15], and indicates that the pinning potential V proportional to $E_{\rm th}$ is reduced with increasing T in such a manner that $V(T)/V(0) = e^{-\langle\phi^2\rangle/2}$, by the thermal fluctuation of the CDW phase ϕ. In a classical model, $\langle\phi^2\rangle = T/T_0$, where T_0 is a constant [16]. Thus, the observed temperature dependence of $E_{\rm th}$ implies that the NDR effect in $La_{2-x}Sr_xNiO_{4+\delta}$ happens when the electric-field induced motion of the stripes overcomes the pinning potential. Namely, the moment the depinning occurs, the holes are in collective motion in analogy to the sliding of CDW.

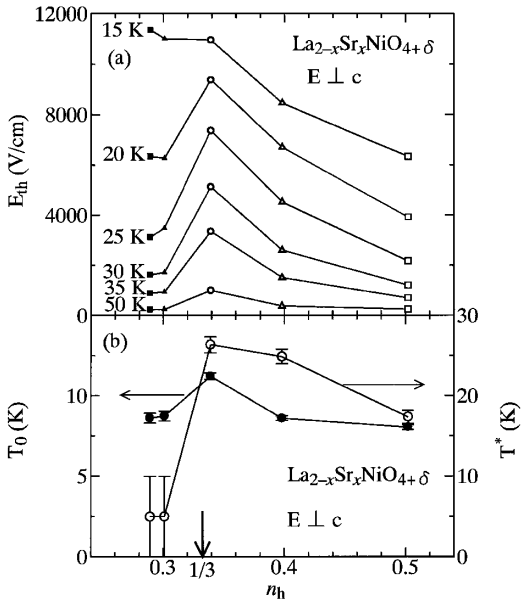

FIG. 2. Hole doping (n_h) dependence of (a) the threshold electric field $E_{\rm th}$ for $La_{2-x}Sr_xNiO_{4+\delta}$ crystals at various temperatures, and (b) parameters appearing in Eq. (2). T_0 (closed circles) and T^* (open circles) correspond to the strength of the pinning potential and the quantum fluctuation for the collective carrier motion, respectively (see text). Errors in panel (a) are less than the size of the respective symbols.

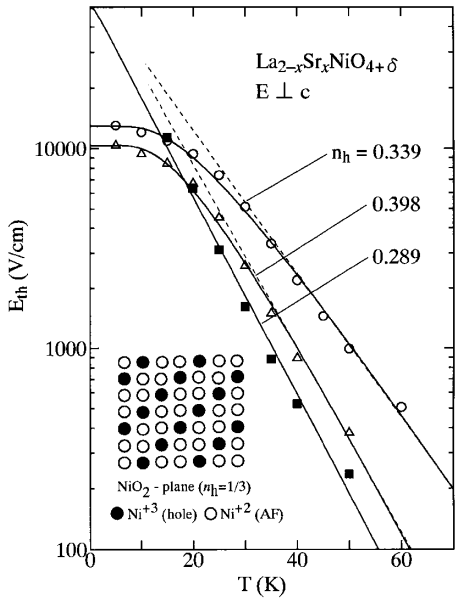

FIG. 3. Temperature dependence of the threshold electric field $E_{\rm th}$ at various doping levels, $n_h = 0.289$ (closed squares), 0.339 (open circles), and 0.398 (open triangles). Dashed lines are the results of fitting with use of Eq. (1) which is a classical approximation, while solid lines with use of Eq. (2) which takes account of the quantum fluctuation as well (see text). The inset represents schematically the striped pattern of the ordered holes ($n_h = 1/3$) in the NiO_2 plane.

Upon the depinning, there are two possible directions of the collective (sliding) motion of the charge stripe (see the inset to Fig. 3). The one is a vertical (100), and the other is a diagonal (110) direction which is parallel to the stripes. In the former case that would be favored by a larger hopping interaction, diagonal stripes themselves may immediately collapse as observed for the case of dielectric breakdown in charge-ordered $Pr_{1-x}Ca_xMnO_3$ [17,18]. In the latter case, the holes would flow within the stripe. At the moment, we cannot determine which is the case, since the charge-ordered crystal is composed of fine (<100 nm) multidomains and the in-plane anisotropy for the NDR effect is hard to be detected. Considering the conspicuous NDR effect, that is rarely encountered in the conventional CDW systems, we speculate that depinned holes initially in a collective or sliding motion may eventually destroy the charge-ordered state, generating such a metallic region subsisting to the lowest temperature.

At low temperatures, $E_{\rm th}$ cannot be fit with the equation (1), as seen in Fig. 3. To interpret this, the quantum fluctuation of ϕ may have to be taken into consideration [19]. Let us assume that the phase fluctuation is represented as $\phi = g(b + b^\dagger)$ by using boson (*e.g.*, phonon) operators $b^\dagger$ and b, and $g^2 = 1/M\omega_0^2$ where M and ω_0 are mass of collective carrier motion and pinning frequency, respectively. Then, the mean squared amplitude of fluctuation is given as $\langle\phi^2\rangle = g^2 \coth(\omega_0/2T)$, and accordingly the expression of the threshold field is modified from the classical expression Eq. (1) to [19]

$$E_{\rm th}(T)/E_{\rm th}(0) = \exp\{-(T^*/T_0)\coth(T^*/T)\}. \quad (2)$$

Here, $T_0 = 2M\omega_0^2$ and $T^* = \omega_0/2$, corresponding to the elastic constant and the pinning frequency, respectively. T_0 is related with pinning strength of the CDW phase and T^* with the quantum fluctuation, or " zero-point vibration" of the charge stripe. Experimentally, we can determine the T_0 from a slope of $\log E_{\rm th}$ vs T plot in a high-temperature region, and T^* from a temperature where the deviation of the experimental data from Eq. (1) is observed. As shown in Fig. 3, experimental data of $E_{\rm th}(T)$ can be well fit to the Eq. (2), representing the classical to quantum crossover with T^* as the crossover temperature.

In Fig. 2(b), the hole concentration dependence of T_0 and T^* is shown. The T_0 shows a maximum at $n_h \sim 1/3$, indicating that the pinning strength of the charge stripe is maximized by the $n_h = 1/3$ commensurability. The T^* is appreciable at $n_h > 1/3$, but steeply decreases below $n_h = 1/3$. This means that the quantum fluctuation is minimal and the mass (M) for the collective charge motion tends to diverge for $n_h < 1/3$. Such an asymmetric behavior with respect to the commensurate point ($n_h = 1/3$) is reminiscent of the anomalous chemical potential shift with n_h, as probed by photoemission spectroscopy [20]. Only for the $n_h < 1/3$ side, the chemical potential shift is completely suppressed, implying a segregation of doped holes on a microscopic scale. The $n_h = 1/3$ charge stripe phase can be viewed as a Mott insulator [11], as in the case of the $n_h = 0$ insulator. Therefore, the present results imply that the microscopically segregated stripe phases into the $n_h = 0$ and $1/3$ states show no low-lying collective modes like the Mott insulators.

The current switching into the NDR regime accompanies a characteristic delay of 10–1000 msec after the voltage step is applied. In the inset to Fig. 4(a), the temporal feature of current is exemplified with varying applied voltage ($V_{\rm cir}$). Compared with the typical CDW system $K_{0.3}MoO_3$ whose delay time τ_d in switching is 1–1000 μsec [21], the τ_d in $La_{2-x}Sr_xNiO_{4+\delta}$ is considerably longer. Strong pinning potential in $La_{2-x}Sr_xNiO_{4+\delta}$ perhaps causes such a long delay time. In Fig. 4(a), the electric field dependence of τ_d is shown for $n_h = 0.339$. The τ_d strongly depends on the electric-field strength, while it is much less dependent on temperature and hole doping level. The τ_d is quite reproducible when measured under the same condition. Phenomenologically, the observed feature of the delayed but still very steep switching is ascribed to spatially inhomogeneous formation of the conduction domains and the subsequent avalanche breakdown process. From the electric field dependence of τ_d, we can crudely estimate the generation rate [$P(E)$] of the conductive domain in an electric field E exceeding the threshold $E_{\rm th}$

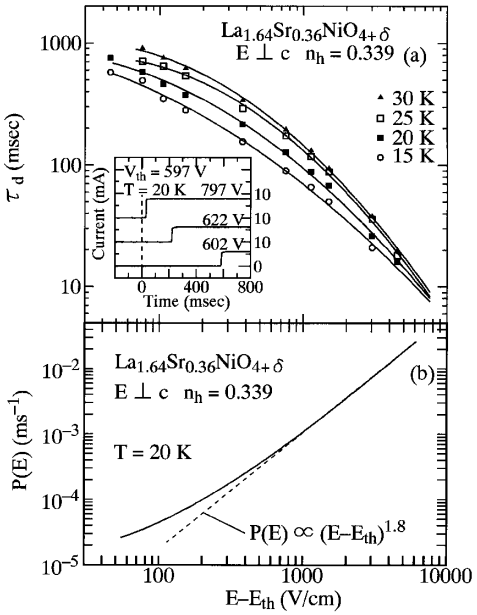

FIG. 4. (a) Electric-field dependence of the delay time τ_d of the switching for an $n_h = 0.339$ crystal. The inset exemplifies the time dependence of current at various applied voltages on the whole circuit which is composed of 100 kΩ load resister and the crystal ($0.66 \times 0.60 \times 0.30$ mm^3) connected in series. (b) Electric-field dependence of the generation rate, $P(E)$, of the conductive domains, derived with use of the phenomenological one-dimensional model and the τ_d vs E data shown in (a) (see text).

[19]. To evaluate $P(E)$ in a semiquantitative manner, we have adopted the simplest model: At time t, the one-dimensional sample is composed of the metallic (fraction y) and insulating (fraction $1 - y$) domains, only on the latter of which all the applied voltage is effectively applied. Then, the fraction (y) of the conductive path in the sample obeys the following equation:

$$dy/dt = P(E)(1 - y), \tag{3}$$

where $E = V/d(1 - y)$. By integrating Eq. (3), one obtains the relation

$$P(E) = -(Ed\tau_d/dE)^{-1}. \tag{4}$$

The electric field dependence of $P(E)$, determined from Eq. (4) and the smoothed data shown in Fig. 4(a) (solid curves), is shown in Fig. 4(b). The P depends nonlinearly on E in such a manner that $P \propto (E - E_{\rm th})^{1.8}$ except for a low-field region where the experimental uncertainty and error is large. The present quantity $P(E)$ may bear some analogy to the critical behavior of the average velocity v of the CDW near the threshold of depinning. According to the mean-field theory [22], $v \propto (E - E_{\rm th})^{3/2}$, whose exponent ($\nu = \frac{3}{2}$) may be well compared with the present case ($\nu \approx 1.8$).

In conclusion, the dielectric breakdown and conspicuous NDR effects are observed by application of a moderately high electric field for the charge-ordered state of $La_{2-x}Sr_xNiO_{4+\delta}$. The onset of the NDR effect in $La_{2-x}Sr_xNiO_{4+\delta}$ is induced by the collective motion of holes on the charge stripes, in analogy to the sliding of the CDW. This is partly evidenced by the exponential temperature dependence of the threshold electric field. At low temperatures, the quantum fluctuation of the further hole-doped stripe phase ($n_h > 1/3$) seems to reduce the threshold electric field. In the switching effect, the prolonged delay time exists near the threshold voltage, indicating the avalanche process limited by the field-dependent nucleation of the conductive regions.

We would like to thank N. Nagaosa for many enlightening discussions, and R. Kumai and Y. Okimoto for helpful advice on our experiment. This work was supported in part by Grant-in-Aids for Scientific Research from the Ministry of Education, Science, Sports, and Culture, Japan, Nissan Science Foundation, and the New Energy and Industrial Technology Development Organization of Japan (NEDO).

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