Dielectric breakdown of one-dimensional Mott insulators Sr_2CuO_3 and $SrCuO_2$

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Dielectric breakdown phenomena accompanied by characteristic time delay and subsequent negative differential resistance effect have been observed for typical one-dimensional (1D) Mott insulators, Sr_2CuO_3 and $SrCuO_2$. In the both compounds, the threshold electric-field for the breakdown, defined as a field above which the differential resistance becomes negative, has been found to increase exponentially with decreasing temperature. This result is similar to the case of charge-density-wave depinning, and indicates the collective nature of charge dynamics in these 1D systems. The electric-field dependence of the delay time is analyzed in terms of a phenomenological model.

One-dimensional (1D) electron systems with strong interaction have long been of great interest[1–6] because of their unique features different from those of higher-dimensional systems. For instance, if we start from noninteracting 1D electrons and take into account the interaction among them as a perturbation, even an infinitesimal magnitude of interaction turns the nature of the liquid from that of Landau-type to that of Tomonaga-Luttinger-type in the case of noninteger filling. In the Tomonaga-Luttinger liquid, no longer well-defined is the single-particle excitation, instead of which collective charge- and spin-density fluctuation disentangled from each other turns to be the elementary excitation of the system. In the case of integer filling, the system becomes Mott insulator due to the umklapp scattering process. Another important characteristics of the 1D system is the effect of strong fluctuation, which prevents any kind of long-range ordering down to much lower temperatures than that predicted by mean-field theories. Therefore, the long-range-ordered phase, if at all with the help of weak three-dimensional interaction, is expected to be fragile to perturbations or external stimuli.

The Sr_2CuO_3 and $SrCuO_2$ investigated here are typical 1D Mott insulators, composed of single- and double-CuO chain sandwiched by insulating $(SrO)_2$ layers, respectively, as depicted in Figs. 1(a) and 1(b). These compounds with $S=1/2$ have extensively been investigated thus far by spectroscopic[7,8] as well as magnetic[9–11] measurements and have proved to be really ideal 1D systems. Moreover, the single- and double-chain are identical with those of the cuprate superconductors, $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$, respectively, and transport studies on the detwinned crystals of them have shown[12,13] a highly conductive nature of the hole-doped single and double chains. In this paper, we report dielectric breakdown or current-switching phenomena with application of moderately high electric field and the associated negative differential resistance in Sr_2CuO_3 and $SrCuO_2$. The threshold electric field (E_{th}) for the current switching shows an exponential dependence on the temperature (T), analogously to the case of charge-density-wave (CDW) depinning in low-dimensional systems,[14] implying the collective nature of the charge dynamics in these 1D compounds. Such a collapse of Mott insulator under nonequilibrium condition is

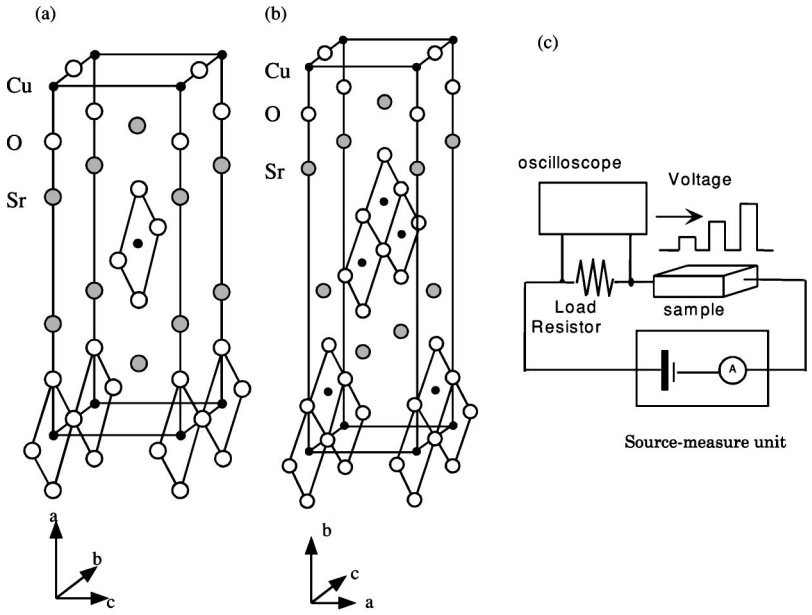

FIG. 1. (a),(b) Schematic crystal structure of Sr_2CuO_3 and $SrCuO_2$, respectively. (c) Schematic diagram of the measurement circuit.

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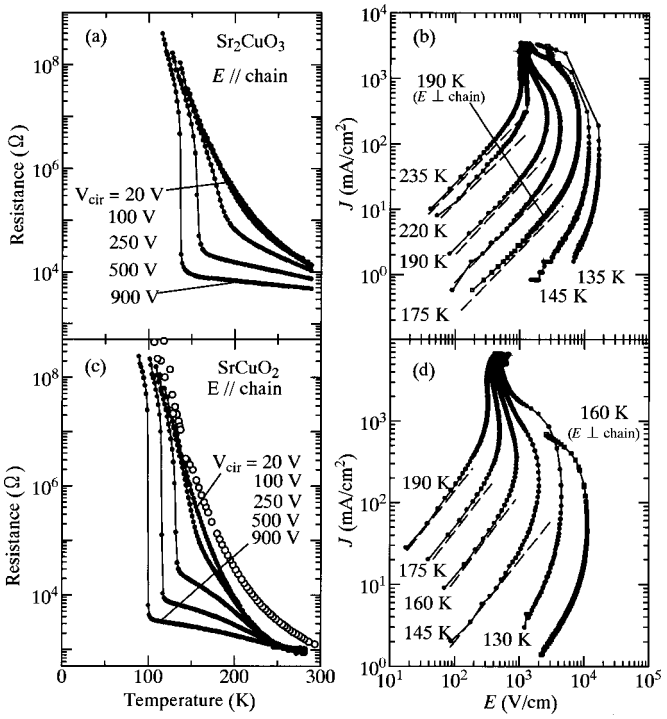

FIG. 2. (a),(c) Temperature variation of the resistance with various applied voltages (V_{cir}) on the whole circuit for Sr_2CuO_3 and $SrCuO_2$, respectively. Open circles in panel (c) are the results with four-probe method for a crystal from the same batch (see text). (b),(d) Electric-field (E)–current-density (J) characteristics at various temperatures for Sr_2CuO_3 and $SrCuO_2$, respectively. The result along the a-axis (b-axis) direction at 190 (160) K is also shown for Sr_2CuO_3 ($SrCuO_2$). 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. The size of Sr_2CuO_3 ($SrCuO_2$) crystal is $0.53\times0.53\times0.53$ mm^3 ($0.53\times0.59\times0.25$ mm^3).

reminiscent of the recently observed breakdown phenomena of charge-ordered state in $Pr_{0.7}Ca_{0.3}MnO_3$ upon the application of high electric-field,[15] laser pulse illumination,[16] and x-ray irradiation,[17] as well as in the stripe-ordered $La_{2-x}Sr_xNiO_4$ system with applying high electric-field.[18]

The single crystals investigated here were grown by the traveling solvent floating zone method. The resistance along the CuO chain direction was measured by two-probe method. Samples were cut out from the rods into a cube with the size of $0.53\times0.53\times0.53$ mm^3 in the case of Sr_2CuO_3 and into a rectangle with the size of 0.53 mm in length (chain direction), 0.59 mm in width, 0.25 mm in thickness in the case of $SrCuO_2$. Electrodes were made of heat-treatment-type silver paste and the typical value of the contact resistance at room temperature was less than 50 Ω for both compounds, which is much lower than the sample resistance itself [see Figs. 2(a) and 2(c)]. Further evidence for the dominant contribution of the sample resistance is provided by the comparison between the results by the two-probe and four-probe methods (*vide infra*). The measurements of current-voltage(I-V) characteristics and temperature dependence of resistance at various applied voltages were performed using a circuit composed of the crystal and a load resistor in series, as schematically shown in Fig. 1(c). The load resistor R_L of 100 kΩ was inserted to protect the circuit against a burst of current upon the switching. A voltage (V_{cir}) up to 1000 V was applied on the whole circuit, namely, the crystal connected in series to the load resistor, and the current (I) was measured by the source-measure unit. The voltage (V) applied on the crystal was obtained via the relation that $V=V_{cir}-R_LI$. Pulse voltage (usually with 200-msec duration) was applied to minimize Joule heating effect. Temporal response of current and hence the sample resistance was measured via the voltage drop at the load resistor with use of a storage oscilloscope.

In Fig. 2(a), we show the temperature variation of resistance (R) of Sr_2CuO_3 at various applied voltages. The R tends to decrease considerably at high voltages, particularly at low temperatures. A discontinuous jump from a low- to high-resistive state is observed at 140 K at a bias voltage (V_{cir}) of 900 V. The results for $SrCuO_2$ crystal are displayed in Fig. 2(c). We have also measured the resistivity, with the four-probe method with small current (1–10 μA), for a crystal from the same batch (but not the exactly same crystal), and have plotted the resistance values (open circles) expected for a sample with the same size as that of the sample measured under high voltages. The resistance value at $V_{cir}=20$ V is even lower than that by the four-probe method, which means that the resistance measured by the two-probe method is dominated not by the contact resistance, but by the sample resistance. A discontinuous jump is also seen in the case of $SrCuO_2$, but the low-resistive state subsists down to 100 K, reflecting the more conductive nature of the double chain than the single-chain in Sr_2CuO_3. At slightly higher temperatures than the discontinuous jump, the R value under 900 V is smaller by more than four orders of magnitude than the R value measured with low voltages.[19]

Such a huge reduction of resistance under high electric field cannot be accounted for in terms of Joule heating effect. For example, the resistance of the Sr_2CuO_3 before the switching is larger than 10^8 Ω just above 140 K. The switching to the low-resistive state is observed with delay time of 100 msec upon the application of 874 V (see the inset to Fig. 4). The total power consumed in the crystal prior to the switching is estimated to be less than 7.6×10^{-4} J. Using the measured value of heat capacity ($\approx$90 J/mol K) of this crystal at 140 K and the sample volume ($\approx2.9\times10^{-6}$ mol), the upper bound of temperature rise due to the Joule heating would be less than 3 K, even if the crystal were thermally isolated completely during this period. Similar estimate of possible temperature rise for the $SrCuO_2$ crystal at about 100 K is even less ($\approx$1 K) than the case of Sr_2CuO_3. Therefore, the observed low-resistive state must be an intrinsic state of strongly correlated electron system under a highly nonequilibrium condition.

Recently, Fukui and Kawakami have theoretically studied the breakdown of 1D Mott insulator[20] with use of asymmetric Hubbard model. They introduced an asymmetric hopping integral to deal with a situation of forced motion of correlated electrons, namely, a situation where electrons are supplied at the first site and dissipate at another boundary. The model was solved exactly by means of the Bethe ansatz method and it was shown that the Hubbard gap may close, or equivalently, the breakdown of Mott insulating state may occur as the asymmetry of hopping is increased. Although the breakdown does occur in that particular model, the quantitative comparison with the present experiment cannot be made[21] because of the lack of direct correspondence between

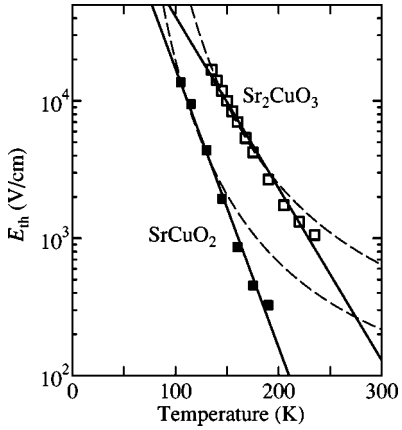

FIG. 3. Temperature dependence of the threshold electric-field E_{th} (measured and defined with use of 200-msec voltage pulses) for Sr_2CuO_3 and $SrCuO_2$. The solid lines are the results of fitting with use of Eq. (1). The dashed lines are the best fit with the use of activation-type temperature dependence.

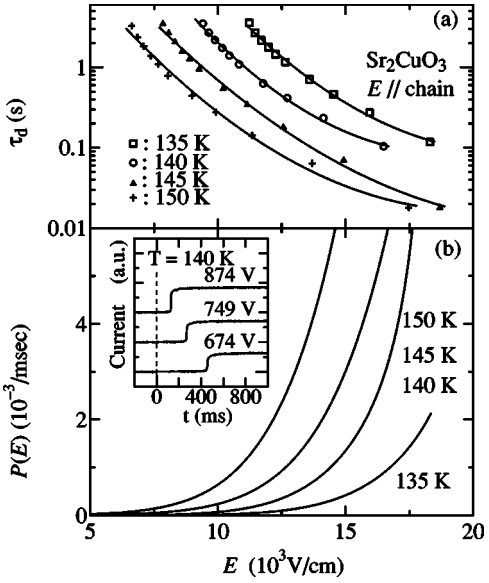

FIG. 4. (a) Electric-field dependence of delay time (τ_d) for the current switching for Sr_2CuO_3 crystal. (b) Electric-field dependence of the generation rate, $P(E)$, of the conductive domains, derived with use of the phenomenological model and the τ_d vs E data shown in (a). The inset exemplifies the temporal evolution of current at various applied voltages on the whole circuit which is composed of 100 kΩ load resistor and the crystal ($0.53\times0.53\times0.53$ mm^3) connected in series.

the asymmetric hopping term in the theory and the applied electric field in the experiment. Nevertheless, the observed low-resistive state may be relevant to such a metallic state as considered theoretically under a highly nonequilibrium condition. The weak interchain interaction as well as the minimal disorder in the chain, which have been confirmed in several experiments,[7–13] seem to be also crucial for the present breakdown phenomena.

In Figs. 2(b) and 2(d), we show the electric-field (E) dependence of current density J for the respective crystals. The values of E were derived from the relation that $E=(V_{cir}-R_L I)/d$, d being the length of the sample. At relatively small values of E, the J-E characteristics show Ohmic relations as indicated by dashed lines, but as the applied electric-field is increased, they exhibit clear deviation from the Ohmic law and eventually show negative differential resistance (NDR) behavior above threshold electric field (E_{th}). Such a NDR behavior means the presence of self-multiplication (avalanche) process of current. For comparison, we show the J-E characteristics for the E vector perpendicular to the CuO chain axis, i.e., along the a axis (b axis) at 190 K (160 K) for Sr_2CuO_3 ($SrCuO_2$). Reflecting the anisotropy of the crystal and electronic structures, the resistance is fairly high compared with that of the chain direction. The current-switching behavior is apparently observed also in this direction, but this may probably be due to the possible misalignment (1°–2°) between the crystallographic axis and the direction of applied field in this two-terminal experiment, and may not be an intrinsic J-E characteristic of this direction.

We show in Fig 3 the obtained E_{th} with use of 200-msec voltage pulses as a function of temperature for the both crystals. As readily seen in the figure, $E_{th}(T)$ for each compound obeys the following temperature dependence:

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

Straight lines in Fig. 3 are the best fit for the experimental data with use of Eq. (1), and the obtained value of T_0 is 34.7 K (21.4 K) for Sr_2CuO_3 ($SrCuO_2$). If we adopt a theory of dielectric breakdown by Fröhlich[22] based upon electron avalanche process, thermal activation-type temperature dependence of $E_{th}(T)\propto\exp(T_0/T)$ would be expected. As shown in Fig. 3 by dashed lines, the activation-type temperature dependence is distinct from the present experimental observation. An exponential T-dependence has often been found in CDW systems[23] and interpreted in terms of weakened pinning potential due to the thermal fluctuation of CDW phase ϕ: Thermal fluctuation of ϕ reduces the pinning potential V that is proportional to E_{th} in such a manner as $V(T)/V(0)=\exp(-\langle\phi^2\rangle/2)$, where $\langle\phi^2\rangle/2=T/T_0$.[24] T_0 is a constant which represents the pinning strength of the CDW phase. Therefore, the observed exponential dependence of E_{th} suggests that the collective motion of carriers present on the 1D chain is responsible for the NDR effect. The values of E_{th} (10^2–10^4 V/cm) for these materials are larger than the typical value of E_{th} (10^{-2}–10^{-1} V/cm) for conventional CDW pinned by impurity,[14] but is much smaller than the typical value of breakdown field ($\sim10^7$ V/cm) in band insulators.[25]

The current switching into the low-resistive state accompanies a characteristic delay time (τ_d) of 10–1000 msec after the voltage is applied on the circuit. The temporal variation of current for the case of Sr_2CuO_3 is exemplified in the inset to Fig. 4(b). The delayed but still steep switching may arise from spatially inhomogeneous formation of conductive domains and subsequent percolation process. The measured τ_d is plotted in Fig. 4(a) as a function of applied electric field at several temperatures. From the E-dependence of τ_d, we derived the generation rate [$P(E)$] of the conductive domain in an electric field E with use of the following model.[26] At time t, 1D chain is composed of metallic domain (fraction y) and insulating domain (fraction $1-y$), and we assume that all the applied voltage is effectively applied only on the insulating domain. Thus, the effective electric field that the insulat-

ing domain feels becomes larger and larger as the fraction of metallic domain is increased. As for the temporal evolution of the fraction y of metallic domain, the following equation holds:

$$dy/dt = P(E)(1-y), \tag{2}$$

where $E = V/d(1-y)$, d being the length of the sample. By integrating Eq. (2), one obtains the relation

$$P(E) = -(Ed\tau_d/dE)^{-1}. \tag{3}$$

Using Eq. (3) and the smoothed data shown in Fig. 4(a) (solid curves), we obtained the electric-field dependence of the generation rate $P(E)$, and plotted them in Fig. 4(b). P depends on E in a strongly nonlinear manner ($P \propto E^6 \sim E^8$), while a subsisting switching to a lower field on a longer time-scale prevents the accurate determination of $E_{\rm th}(\tau \rightarrow \infty)$. These features may reflect a large fluctuation of charge dynamics in 1D system.

In summary, we have observed dielectric breakdown and subsequent NDR with application of moderately high electric-field for typical 1D Mott insulators, Sr_2CuO_3 and $SrCuO_2$. The threshold electric field for the NDR effect shows an exponential increase with lowering temperature, and the similarity between the presently observed result and the results of CDW depinning implies the collective nature of the charge dynamics in these strongly correlated 1D systems. The current switching is accompanied by a characteristic delay time, which is phenomenologically interpreted by a simple model that takes into account the nucleation of conductive domains and the effective enhancement of electric-field resting on the remaining insulating domains.

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