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Evidence for the existence of the inherent periodicity in the switched state at low temperatures in $K_{0.3}MoO_3$

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The current response was measured in $K_{0.3}MoO_3$ in the highly conducting state after the switching at low temperatures, both in time domain and in frequency domain. Clear evidence was found which shows that the switching or the strong nonlinear conduction in this temperature range is due to the sliding motion of charge-density waves. Furthermore, the dynamical behavior of the charge-density waves very near the threshold voltage was analyzed quantitatively, which is unique to the low-temperature switching in $K_{0.3}MoO_3$.

It has been well established that many quasi-one-dimensional materials undergo a Peierls transition at a finite temperature T_P, below which the macroscopic quantum condensate, i.e., charge-density wave (CDW) is formed. Furthermore, some of them show nonlinear conduction and accompanying interesting phenomena due to the collective motion of the CDW's. These phenomena have provided a great deal of interest for ten years in solid-state physics.[1] Potassium molybdenum oxide, $K_{0.3}MoO_3$, has a quasi-one-dimensional electronic structure,[2] and undergoes a Peierls transition at $T_P = 180$ K,[3] below which it behaves as a semiconductor.[4] In the semiconducting state, the collective transport due to the CDW is observed in this material, as was in $NbSe_3$ and TaS_3.[5] Two years ago, we found that it shows a drastic switching with a threshold field of about 100 V/cm.[6] By this switching, conductivity of the sample becomes more than 10^3 times. In this highly conducting state, a large variety of phenomena were observed in the time-domain current response including quantized voltage jumps,[7] a finite time delay,[6] impulsive relaxation-type oscillation (RTO), intermittency,[8] etc. Since some of these were observed in the materials like $NbSe_3$ and TaS_3,[9-13] and since the breakdown phenomena by the uncondensed electrons seem impossible because of their low mobility, we considered that the switching in $K_{0.3}MoO_3$ in this temperature region is due to the sliding motion of the CDW's. In particular, the RTO seems to correspond to the periodic modulation of the current response due to the inherent periodicity in the CDW; the so-called narrow-band noise.[14] However, this RTO is affected strongly by the external circuit elements. For instance, the externally attached capacitor C_{ext}, parallel to the sample, decreases the frequency of the oscillation without changing its amplitude. In other words, C_{ext} increases the charge carried by each impulse.[7] On the other hand, the frequency of the narrow-band noise in $NbSe_3$ and TaS_3 is not affected by the external capacitance. This kind of oscillation is also possible simply in terms of the relation between the switching I-V characteristic of the sample and the load line of the circuit. Thus, there still remains a possibility that the low-temperature switching in $K_{0.3}MoO_3$ is not due to the sliding motion of the CDW's. In this Rapid Communication, we show that the oscillation which is insensitive to the external circuit does exist in the low-temperature switched state in $K_{0.3}MoO_3$, which clearly shows that the switching at low temperatures in $K_{0.3}MoO_3$ is due to the sliding motion of the CDW's. Furthermore, the first quantitative analysis is given on the behavior of the CDW very near the threshold voltage.

Samples were prepared by the electrolytic reduction method.[15] Electrical contacts were made by ultrasonically soldered indium. The current response of the sample in two-probe configuration was recorded in the digital memory as the voltage drop across an Ohmic resistance R_0, which is much lower than the resistance of the sample, under the constant voltage V_0 applied across the sample and the Ohmic resistance (Fig. 1). Frequency-domain analysis of the response was performed both by the fast-Fourier transform of the response and by analyzing the signal directly by an analog spectrum analyzer after preamplification.

Figure 2 shows an I-V characteristic of a sample. A switching phenomenon with a hysteresis was observed above the voltage of about 17.3 V. For the applied voltage larger than V_T, the current response had large fluctuations. As shown below, the oscillatory response was clearly seen in the time domain as the response to the dc continuous voltage. Figure 3 shows examples of the responses

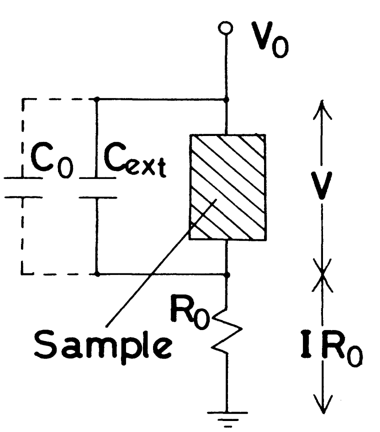

FIG. 1. Configuration of the circuit including the positioning of a specimen. C_0 is the stray capacitor. See the text for detail.

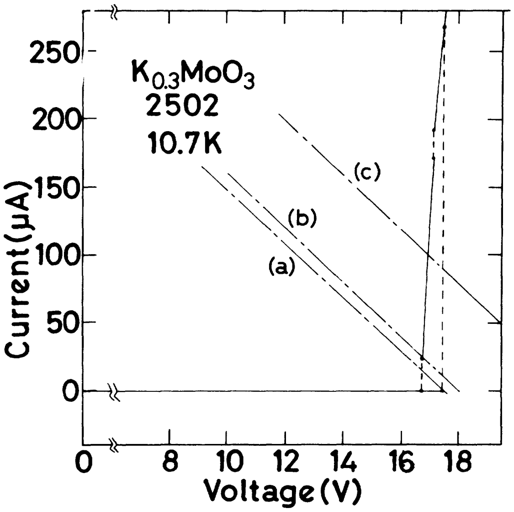

FIG. 2. I-V characteristic of a sample which displays the switching. The dashed-dotted lines show the load lines which correspond to the measuring conditions for the time-domain traces shown in Fig. 3.

for three values of applied voltage which are shown in the form of a load line in Fig. 2. For the load line (a), the circuit-sensitive RTO mentioned above was observed [Fig. 3(a)]. With increasing applied voltage, an oscillatory response with a slightly different waveform was observed [Fig. 3(b)]. The frequency of this new oscillation increases with increasing current. It is approximately proportional to the average excess current. The ratio of the excess current density to the frequency of the oscillation, J/f is 3 A/MHz cm^2. With further increasing current, the oscillation could not be observed in time domain [Fig. 3(c)]. The frequency-domain analysis, however, shows that the oscillating components still exist. As the origin of the oscillations shown in Figs. 3(b) and 3(c), an interplay between the nonlinear I-V characteristic and the load line does not exist because they have a stable crossing point in the I-V plane. In fact, as shown in Fig. 4, the frequency-domain analysis by a spectrum analyzer shows that the frequency of the oscillation does not change when the externally attached capacitance C_{ext} was increased. This behavior is the same as that observed in $NbSe_3$ and TaS_3 as mentioned above. Thus, it becomes clear that the oscillations shown in Figs. 3(b) and 3(c) are due to the sliding of the CDW's. The small value of J/f is possibly due to the inhomogeneity in the sample.

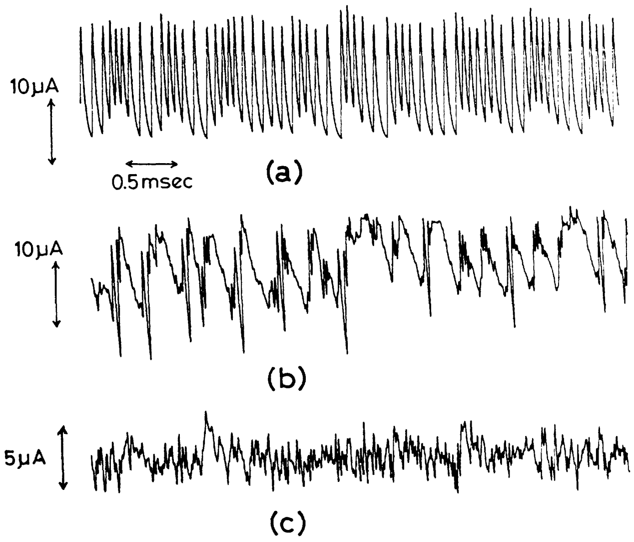

FIG. 3. Oscillating current responses of the same sample as in Fig. 2 observed in the time domain for a continuous dc applied voltage. The average current were (a) 10, (b) 20, and (c) 100 μA, each of which corresponds to the load lines shown in Fig. 2.

From the result shown above, it is reasonable to assume that the current in the form of the RTO is also carried, at least in part, by the sliding CDW which has the inherent periodicity. Then it seems quite natural to associate each period of the RTO with the periodicity of a CDW even

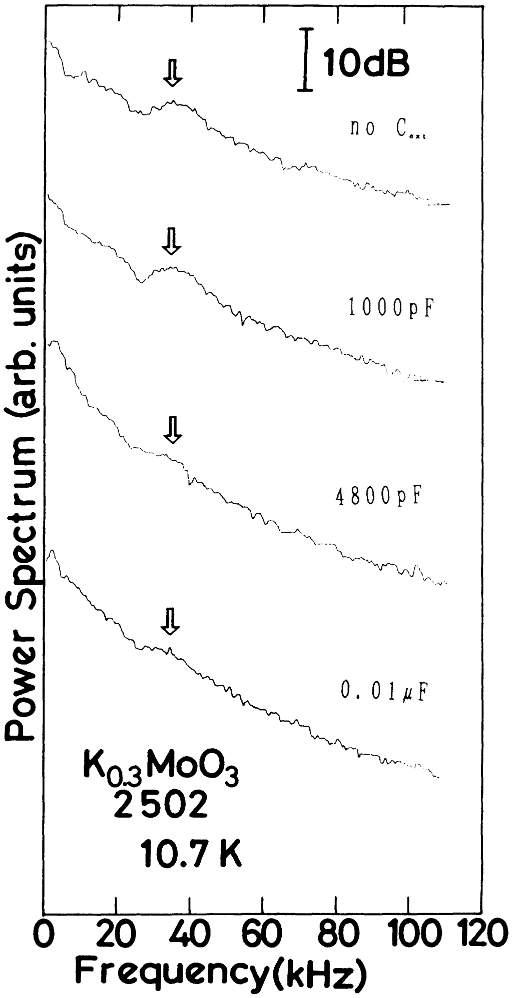

FIG. 4. Dependence of the power spectrum of the oscillating current response on the externally attached capacitance, C_{ext}. The average current was 100 μA. Arrows indicate the frequency of the oscillating component.

though this RTO is surely caused partly by the coupling between the shape of the I-V curve and the load line of the circuit. However, the fact that the charge Q under each cycle (or J/f) increases with increasing C_{ext} is puzzling from this point of view. One possible explanation is the inclusion of the single-particle contribution for the excess current.[16] Alternatively, the increase in Q (or J/f) is due to the increase of the coherent cross section of the moving condensate. In fact, the value of J/f of the RTO is, for example, 0.7 A/MHz cm^2 for Fig. 3(a), which is much smaller than that of Figs. 3(b) and 3(c). If the two kinds of oscillation are related in the same way to the periodicity of the CDW, the RTO of Fig. 3(a) must involve current in a smaller cross section. Thus, the value of the cross section is variable. So, the suggestion that the C_{ext} increases the coherent cross section is possible. These problems will be discussed in more detail in a later publication.

The switching phenomenon in this temperature range in $K_{0.3}MoO_3$ is novel in the sense that the bare response of the CDW can be observed very near the threshold field because of the absence of uncondensed carriers. Because of the existence of the bistability, however, samples with switching seem inappropriate for the quantitative analysis. Fortunately, sometimes samples were found which did not show switching, but still had a definite threshold voltage. The circuit-insensitive oscillation reported above was also observed in these nonswitching samples. For these samples, the behavior in the nonlinear conduction very near the threshold voltage can be discussed quantitatively. Figure 5(a) shows the I-V characteristic of a nonswitching specimen. The threshold voltage V_T, which is indicated by an arrow, was easily determined by analyzing the fluctuating component of the current response. Even below V_T, weak nonlinearity was observed. However, it seems to be the phenomenon different from the strong nonlinearity above V_T. The field-induced depolarization current may be the origin.[17] Figure 5(b) shows the frequency of the oscillation f as a function of the reduced field Φ which is defined as

$$\Phi = (V - V_T)/V_T \ . \qquad (1)$$

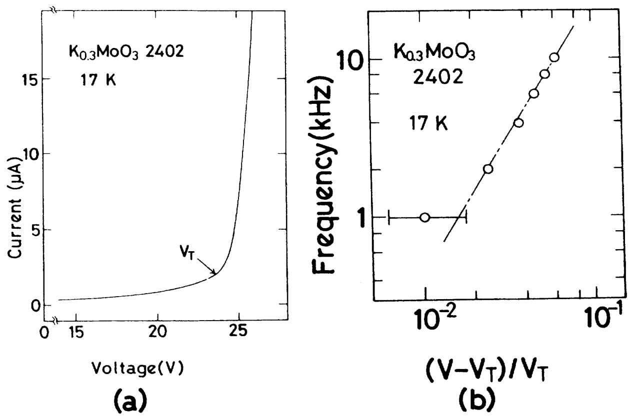

FIG. 5. (a) I-V characteristic of a sample which does not show the switching. The threshold voltage is indicated by an arrow. (b) Frequency of the oscillations as a function of the reduced field defined in Eq. (1) in the text.

As shown in this figure, the power law is roughly valid, that is,

$$f \propto \Phi^{\alpha} \ , \qquad (2)$$

with the exponent of $\alpha = 1.7$. It should be noted that $10^{-2} < \Phi < 10^{-1}$. So far as we know, it is the first quantitative analysis of the nonlinear current in such a small region of Φ. This behavior can be compared with the theory related to the dynamical critical phenomenon.[18] However, further development in this field is needed for detailed discussion.

In summary, evidence was found which clearly shows that the switching or strong nonlinearity at low temperatures in $K_{0.3}MoO_3$ is due to the sliding of the CDW's. Furthermore, as a result, it becomes possible to discuss the behavior of the nonlinear current very near V_T quantitatively. This is characteristic of the low-temperature switching in $K_{0.3}MoO_3$.

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[1] For a review of recent developments, see, for example, *Proceedings of the Yamada Conference XV on Physics and Chemistry of Quasi One-Dimensional Conductors, Lake Kawaguchi, 1986,* edited by S. Tanaka and K. Uchinokura [Physica B **143** (1986)].

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