Switching of $K_{0.3}MoO_3$ at low temperatures. II. ac conductivity in the highly conducting state

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The ac conductivity was measured as a function of the dc bias, the ac amplitude, and the frequency in the low-temperature highly conducting state of $K_{0.3}MoO_3$. As a result, the following new features were observed. (1) The Drude-like conductivity was observed with the relaxation time equal to the period of the one-wavelength motion of the charge-density wave (CDW). For frequencies above 5 MHz the conductivity increases again with a further increase in frequency. (2) The clear resonantlike interference structure was observed at the bias voltage where the frequency of the current oscillation of the CDW becomes just equal to the frequency of the external ac field. Almost all of these features were well explained by the so-called Fukuyama-Lee-Rice model, in which only classical dynamics of the phase of the CDW is taken into account. Thus, the amplitude mode was found not to play any important role in the ac response in the radio-frequency region.

I. INTRODUCTION

The dynamics of the collective motion of the charge-density wave (CDW) in quasi-one-dimensional materials has been one of the most interesting topics in solid-state physics during this decade.[1] Although common features of the collective motion of the CDW have been clarified by extensive studies, several topics are still open to question. In contrast to what is initially expected, the conductivity due to the sliding CDW stays finite even in the high-field limit, which clearly demonstrates that the CDW in the sliding state suffers from finite damping. Several microscopic models of the mechanisms of the damping of the CDW have been presented.[2,3] Experimentally, the conductivity due to the sliding CDW often scales with that of the normal carriers,[4–6] and the microscopic damping is masked. This is partly because of the existence of the normal carriers. In many cases of sliding, the motion of the CDW is associated with the screening effect of the normal electrons; this screening prevents the observation of the true features of the sliding CDW's.[7]

The same problems lie in the observation of the dynamical conductivity, namely the conductivity of the CDW for an alternating electric field, in the sliding state. Detailed experiments were performed in $NbSe_3$ (Refs. 8–10) and TaS_3 (Refs. 10 and 11) under the presence of finite dc bias voltage, and many of the experimental results are in good agreement with the theoretical models based on the classical description of the sliding CDW.[12–15] In $NbSe_3$ and TaS_3,[10,11] both the real part and imaginary part of the conductivity σ are almost independent of the frequency in the low-frequency region, and $\mathrm{Re}\sigma$ monotonically increases with increasing dc bias voltage, whereas $\mathrm{Im}\sigma$ decreases with increasing dc bias voltage. In the higher-frequency region, both the real part and the imaginary part of the conductivity increase with increasing frequency. There is also the structure due to the interference effect between the intrinsic periodicity of the sliding motion and the external ac field. From these experiments, however, it is not clear if the finer structure exists or not because of the large value of the conductivity due to the normal electrons. Here again, the presence of the normal carriers hinders the detailed inspection of the dynamics of the CDW.

In this sense, the study of the switching in $K_{0.3}MoO_3$ observed only below about 30 K (Refs. 16 and 17) is valuable because in this temperature region there are almost no normal carriers. At the switching threshold, conductivity suddenly increases by several orders of magnitude. It has been clarified that this phenomenon is also due to the sliding motion of the CDW.[18] The detailed investigation of the response of the CDW to the dc electric field shows the following characteristic aspects.[19] The conductivity in the highly conducting state increases with decreasing temperature, this is in sharp contrast to the decrease of conductivity in the low-conducting state. This is anomalous when compared with the ordinary scaling behavior between the conductivity due to the CDW (σ_{CDW}) and that due to normal carriers σ_0 mentioned above.[4–6] Furthermore, the current response observed in the time domain shows various behaviors. In the preceding paper (paper I),[19] we have shown that many aspects of these phenomena are explained by a model proposed by Littlewood.[20] That is, at low temperatures, because of the absence of normal carriers, screening of the deformation of the CDW associated with the sliding motion becomes insufficient. This leads to the transition from the screened motion to the unscreened quasirigid motion of the CDW when the velocity of the CDW increases. The anomalous temperature dependence of the highly conducting state conductivity is consistent with this model. The finite time delay, together with various other phenomena observed in the time domain, is also consistent with this model.

In the same paper,[19] it was inferred that there are three types of unit motions of the CDW, and that the effect of the amplitude mode should be taken into account for a full description of the sliding motion of the CDW's.

Most of these conclusions could be derived because the variation in conductivity due to the switching is enormously large. From the same reason, we can also expect that the dynamical conductivity due to the sliding CDW can be extracted in the highly conducting state of $K_{0.3}MoO_3$ at low temperatures. In this paper, the detailed investigation of the frequency-dependent conductivity under the presence of both the small ac field and the dc bias voltage above the switching-threshold field will be presented. Several new features were found. The meaning of these results will be discussed. A part of the present results was very briefly reported in Ref. 21.

II. EXPERIMENT

The sample was prepared as reported in Ref. 22.

Conductivity measurement was performed by the two-probe method. Electrical contact was made by ultrasonically soldered indium. We confirmed that the magnitude of the contract resistance was much smaller than the resistance of the sample itself.

The ac electric conductivity was measured by a Hewlett-Packard HP4194A impedance analyzer. This can supply the dc bias voltage up to 40 V with the resolution of 10 mV, which makes the combined ac-dc experiment easy. Four coaxial cables from the HP4194A, with each length of 1 m, were connected to the sample. The frequency range is between 100 Hz and 15 MHz in the present configuration. The amplitude of the ac voltage was varied from 10 mV to 40 V.

III. EXPERIMENTAL RESULTS

Figure 1 shows the dynamical conductivity of a $K_{0.3}MoO_3$ sample which shows the switching as a function of frequency at various dc bias voltages above the switching-threshold voltage. Several structures are observed. In order to see these structures more clearly, these results are schematically drawn in Fig. 2. In the highest-frequency region, as is denoted by "A," both parts of conductivity, Reσ and Imσ, increase with increasing frequency. It was sometimes possible to measure the frequency dependence of the conductivity below the switching-threshold field at around 30 K when the samples with relatively low resistance were obtained. In this case, we also observed the structure "A" even at low dc bias voltage.

The second remarkable structure denoted by "B" in Fig. 2 is a definite peak in Reσ and the corresponding structure in Imσ. This structure moves to higher frequencies with increasing dc bias voltage. This behavior strongly resembles that of the inherent current oscillation observed only in the dc response.[1,18] Figure 3 shows the comparison of the frequency of the structure "B" and that of the current oscillation, f_{osc}. Both frequencies are almost the same, and increase with increasing current. Thus, it is concluded that the structure "B" comes from the interference effect between the applied ac signal and

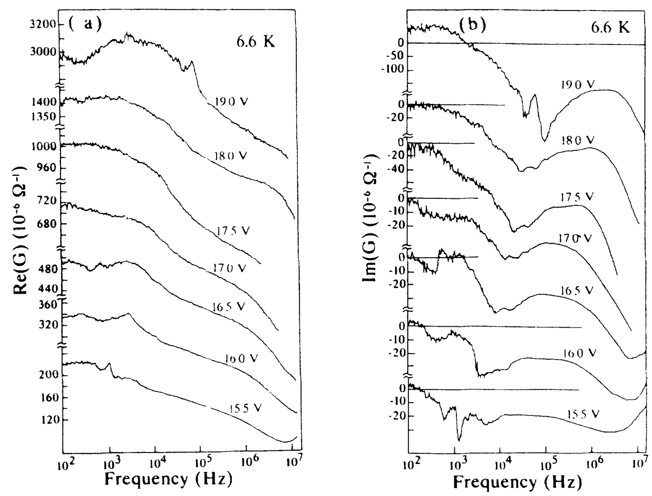

FIG. 1. The frequency dependence of the ac conductance of a $K_{0.3}MoO_3$ sample which shows switching at low temperature under various dc biases at 6.6 K. The threshold voltages for this sample are 17.0 and 15.25 V for increasing and decreasing dc bias, respectively. (a) The real part and (b) the imaginary part.

the inherent periodicity of the CDW. As is observed in Fig. 1, the amplitude (or the sharpness) of this structure strongly depends on the dc bias voltage, but does not change monotonically. Anyway, the existence of this kind of the interference effect is a clear evidence of the existence of the intrinsic periodicity in the highly conducting state of $K_{0.3}MoO_3$.

In Fig. 1, another structure exists around the structure "B." It may be said that this structure is a part of the structure "B." However, as shown in Fig. 4, this structure does not depend on the amplitude of the alternating field, whereas the structure "B" strongly depends on the amplitude of the alternating field. Thus, below we distinguish this structure from the structure "B," and call this

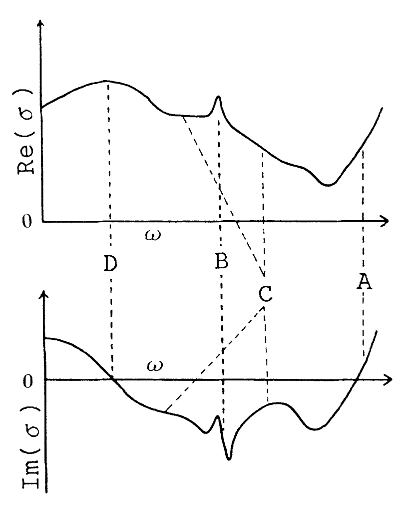

FIG. 2. Schematic drawing of the ac conductivity under the dc bias above the switching-threshold voltage V_T' as was observed in Fig. 1.

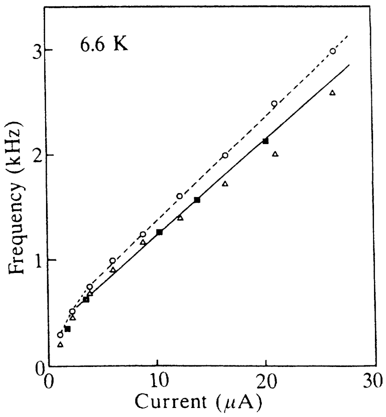

FIG. 3. The comparison of the frequency of the current oscillation f_{osc} observed under the stationary dc voltage (solid squares) with that of the structure "B" shown in Fig. 2 (open circles and open triangles) as a function of the dc current at 6.6 K. Triangles and circles represent the maximum of Reσ and the minimum of the Imσ, respectively. The amplitude or the ac signal is 0.5 V. Dashed and straight lines are a guide for the eye.

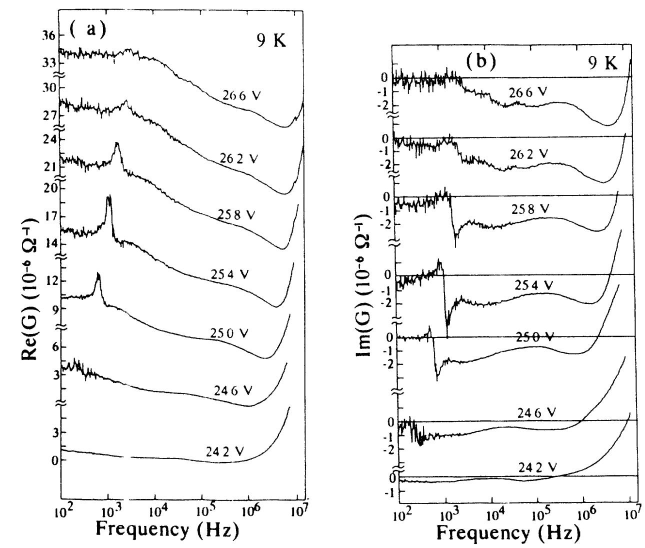

FIG. 5. The frequency dependence of the ac conductance of a nonswitching $K_{0.3}MoO_3$ sample under the various dc biases at 9 K. (a) The real part and (b) the imaginary part.

the structure "C."

The "C" resembles a Drude-type conductivity expressed as

$$\sigma(\omega)=\sigma_0/(1+i\omega\tau) \tag{1}$$

with the relaxation time $1/\tau$, which is almost equal to the peak frequency in Reσ ("B"). It is not puzzling that the Drude-type conductivity is observed in the sliding state, because the Drude-type conductivity is a conductivity of the freely moving carrier. However, what is puzzling is the fact that the relaxation time is almost equal to the so-called "washboard" frequency f_{osc}.[12] This problem will be discussed in Sec. IV. This structure was not observed in $NbSe_3$ nor TaS_3.

There is another broad hump "D" in the lowest-frequency region, which becomes remarkable when the bias voltage is relatively large (for instance, the data at 19 V in Fig. 1). As will be shown below, this structure was observed only in the samples which show the switching.

Sometimes we obtained the sample which does not show the discrete jump even below 30 K. We called it the "nonswitching" sample in the preceding paper.[19] Figure 5 shows the frequency dependence of the conductivity at various dc bias voltages in a nonswitching sample. All structures observed in the switching sample other than the structure "D" were obtained also in nonswitching samples.

In the next section, the origin of these structures will be discussed.

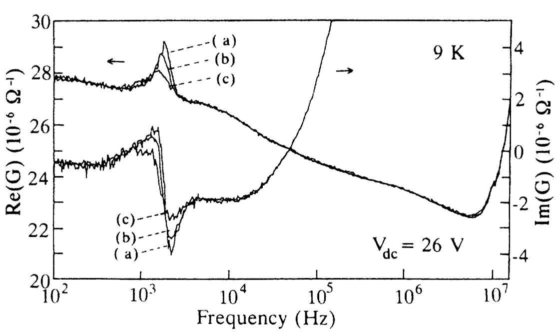

FIG. 4. The dependence of the ac conductivity of a switching sample on the amplitude of the ac voltage. (a), (b), and (c) are the data for the amplitude of 0.1, 0.5, and 1 V, respectively.

IV. DISCUSSION

A. The structures "A" and "C"

As was mentioned above, the structure "A" is observed even in the pinned state at high temperatures where the switching is not observable. Thus, this structure can be considered as a tail of the pinned phase mode which is observed both by the microwave and millimeter-wave[23–26] and far-infrared optical measurements.[27] The fact that the contribution of the "pinned" phase mode is observable even when the CDW slides above the switching threshold contradicts the idea of the rigid motion of the CDW. In the rigid-particle picture, the conductivity in the sliding state should be the Drude type with the relaxation time which is determined by the microscopic mechanism of the damping. The most trivial possible reason is that a part of the sample is in the highly conducting state, and the remaining part of the sample is in the pinned state. However, this kind of response was also observed in $NbSe_3$ and TaS_3 in the ordinary sliding state.[11] Thus, it seems that in this frequency region, CDW behaves intrinsically "pinned" even in the sliding state, and at some frequency, the crossover from the high conductivity of the sliding state to the low conductivity of the pinned state should occur. This crossover just seems to correspond to the structure "C"—that is, the Drude-type conductivity with the relaxation time which is almost equal to f_{osc}.

As was mentioned above, these behaviors cannot be explained in the rigid-particle picture, and the deformation associated with the sliding motion should be taken into account. In the deformable CDW picture, which is called the Fukuyama-Lee-Rice (FLR) model,[13] the phase of the CDW is treated as a dynamical classical valuable, as follows:

$$d^2\phi/dt^2+\gamma\, d\phi/dt+\kappa\, d^2\phi/dx^2 + \sum_i (V_0/m^*)\sin(Qr_i+\phi)=(eE/m^*)e^{i\omega t}\,, \quad (2)$$

where ϕ and m^* are the phase and the mass of the CDW, respectively, γ is the phenomenological damping constant, V_0 is the pinning potential, r_i is the position of the impurities, $Q=2k_F$ is the wave number of the CDW. Based on this model, many numerical calculations have been performed in detail.[28–33] As a result, the motion of the CDW was found to be quite different from what we expect from the term "sliding." That is, the sliding motion is basically the repetition of the creation annihilation of the phase solitons, and during most of the time of the sliding, the CDW is pinned by impurities.[29,30]

Based on this picture, the observed structure "C" could be understood as follows. For frequencies well below f_{osc}, the sliding CDW must collide against the pinning centers many times during a period of the external ac field. Thus, the change of the external field is small during the one-wavelength motion of the CDW and, therefore, the ac response of the CDW is essentially the same as that under the dc applied field. On the other hand, for the ac applied field with the frequency well above f_{osc}, the ac field changes its magnitude and sign during the time shorter than the period with which the CDW collides against the pinning centers. In this case, the ac response of the CDW is essentially that of the pinned phase mode. Thus, we can observe the crossover from the highly conductive dc response to the pinned response of the CDW near f_{osc}. The fact that the Drude-like contribution to the ac conductivity was observed in the sliding state is characteristic of the low-temperature switching state in this material. In fact, numerical calculation based on the FLR model reproduces these frequency dependences of the conductivity very well.[33]

We do not know whether this contribution exists commonly in other materials like $NbSe_3$ or TaS_3. However, we think that it is impossible to observe this structure in other materials, because of the following two reasons. First, in $NbSe_3$ and TaS_3, the pinning frequency is much smaller and therefore the low-frequency tail of the pinned phase mode is in the same range of frequency as f_{osc} (typically 10^7 Hz in $NbSe_3$). Second, the conductivity increase due to the CDW motion above the threshold voltage V_T is relatively small because of the large conductivity due to the normal carriers.

B. The interference structure

The interference structure observed as the structure "B" in Fig. 1 clearly demonstrates that the inherent periodicity exists in the highly conducting state of $K_{0.3}MoO_3$ at low temperatures, as does in the ordinary sliding conduction in this material at higher temperatures and in other materials. However, the detailed feature of the interference is slightly different from those observed in other cases.[10,11] Figure 6 shows the schematic representation of the interference structure. In Fig. 6(a), where the resonancelike structure observed in the low-temperature highly conducting state is shown, Re(σ) takes its local maximum at the washboard frequency, and Im(σ) has a corresponding structure. On the other hand, in Fig. 6(b), which is almost common to the interference structure of other case like in $NbSe_3$ (Ref. 10) and TaS_3 (Ref. 11), Im(σ) takes the minimum at the washboard frequency. At present, the origin of this difference is not known. However, one possible origin is the difference in the experimental condition. In the present case, the data were taken under the nearly constant-voltage condition, and the amplitude of the alternating current is much smaller than the dc bias voltage. On the other hand, in the case of the interference in the ordinary sliding, most of the measurements were performed under the nearly constant-current condition, and the amplitude of the alternating current is a few times larger than the dc bias current, namely a few times larger than the threshold current of the sliding. Although this large difference in experimental conditions makes the problem complicated, the characteristic feature of the low-temperature switching in $K_{0.3}MoO_3$ should be also considered as another origin of the difference. That is, in the ordinary sliding state, the presence of the normal carriers leads to the screening of the polarization of the CDW. This may lead to the dissipative structure in the interference effect. On the other hand, in the low-temperature highly conducting state, the absence of the screening of the random deformation of the CDW may change the nature of the interference effect. This may lead to the resonancelike

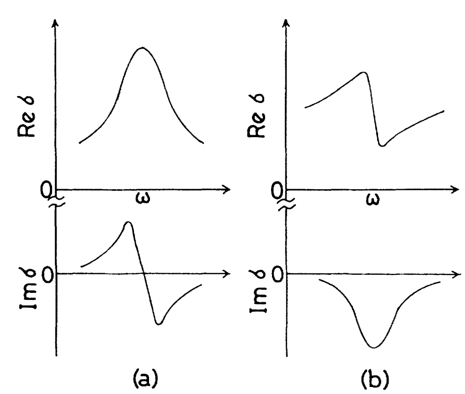

FIG. 6. Schematic drawings of the interference structure observed in the frequency dependence of the ac conductivity. (a) In the switching in $K_{0.3}MoO_3$ at low temperatures, and (b) for the ordinary sliding in $NbSe_3$ and TaS_3.

structure observed in Fig. 1. This resonant structure in the interference is also obtained in the numerical simulation based on the FLR model without taking the contribution of the normal carriers.[33]

C. The low-frequency structure "D"

The low-frequency structure "D" is broad and very noisy. It was observed only in the samples which show the switching, whereas the structures "A"–"C" were observed also in samples which do not show the switching. Thus, we consider that this structure is strongly correlated with the low-frequency fluctuation which was observed as an intermittency in the time domain.[19] Under the presence of a rf field, the intermittent response may become quasiperiodic.

V. CONCLUSION

The ac conductivity was measured as a function of the dc bias, the ac amplitude, and the frequency in the low-temperature highly conducting state of $K_{0.3}MoO_3$. The Drude-like conductivity was observed in the radio-frequency region. The relaxation time was found to be equal to the so-called "washboard" frequency. In the higher-frequency region, the conductivity increases again with further increasing frequency. Furthermore, the clear resonancelike interference structure was observed at the bias where the washboard frequency of the CDW becomes equal to the frequency of the external ac field.

Almost all of these features cannot be explained by the classical rigid-particle model of the sliding of the CDW, but is well understood by the so-called FLR model, in which only the classical dynamics of the phase of the CDW is taken into account. In paper I, we showed that the relaxation of the random deformation of the CDW is absent in the low-temperature highly conducting stater of $K_{0.3}MoO_3$, based on the temperature dependence of the dc conductivity in the highly conducting state. On the other hand, the present result strongly suggests that the collective coherent deformation is still present. Furthermore, the above results show that the amplitude mode does not play any important role in the ac response in the radio-frequency region.

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