Photocontrol of Dynamic Phase Transition in the Charge-Density Wave Material $K_{0.3}MoO_3$

N. Ogawa,[1] A. Shiraga,[1] R. Kondo,[2] S. Kagoshima,[2] and K. Miyano[3]

[1]*Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan*

[2]*Department of Pure and Applied Science, College of Arts and Sciences, The University of Tokyo, Tokyo 153-8902, Japan*

[3]*Research Center for Advanced Science and Technology, The University of Tokyo, Tokyo 153-8904, Japan*

(Received 26 July 2001; published 29 November 2001)

A photoinduced effect has been studied in the quasi-one-dimensional conductor $K_{0.3}MoO_3$. A weak photoexcitation of the sample can drastically affect the dynamic phase transition of the charge-density wave (CDW) motion from slide to creep. The origin of the photoeffect is attributed to a local destruction of the CDW which leads to the photoinduced phase slip and the redistribution of the CDW phase.

DOI: 10.1103/PhysRevLett.87.256401 PACS numbers: 71.45.Lr, 74.60.Ge, 78.20.–e

The problem of the dynamic response of a periodic elastic medium under the influence of random pinning sites is a topic of continued interest spanning a wide area of condensed matter physics, e.g., charge-density waves (CDW), spin-density waves [1], and a vortex lattice in a superconductor [2]. The response is governed by the interplay between the deformation energy and the pinning potential that favors a particular phase of the periodic medium at a certain point in space [3,4]. Neglecting details, such as the difference in the correlation length along the motion and perpendicular to it [5], the overall evolution of the dynamic response can be qualitatively enumerated in the increasing order of the driving force as follows [6]: (i) deformed solid at rest, (ii) plastic flow or creep, and (iii) sliding solid. The transitions between these phases are called dynamic phase transition. Both the experimental and theoretical studies of the dynamic phase transition have been concentrated on the construction of the phase diagram and on the identification of the symmetry of each phase [7], whereas the dynamic aspect has been studied almost exclusively through the modulation of the applied field [8,9]. In this Letter, we report what we believe to be the first observation of the photoinduced modulation of the effective pinning strength, which results in a slide to creep phase transition. The introduction of light in the field of dynamic phase transition study adds a new dimension to the field with its superb spatiotemporal resolution and wide energy scale.

The experiment has been performed on a CDW material $K_{0.3}MoO_3$. Sample crystals are synthesized by means of the electrolysis method [10]. Typical sample size is $1.3 \times 1 \times 0.2\ mm^3$. Samples are mounted in a continuous-flow-type He cryostat or in a closed cycle refrigerator. Two electrodes of Ag or In were evaporated onto a freshly cleaved surface.

An I-V response of a sample with an electrode gap of 1.17 mm is shown in Fig. 1. As is often observed [1] in $K_{0.3}MoO_3$, a clear switching behavior (at V_s) is seen. A current limiter is set at 500 μA in order to prevent damage to the sample in the sliding state above V_s. In $K_{0.3}MoO_3$, the creeping motion starts at a rather low field [11], and the I-V response at low voltage shown in the figure is dominated by the creep rather than the thermally excited normal current across the CDW gap (for the analysis, see below). Under a weak light illumination (photon energy = 2.33 eV), two things happen; the increase of the creep flow and the shift of the sliding voltage V_s. On reducing the applied voltage, a clear hysteresis is also seen [12]. In contrast to the shift of V_s, the switch-off voltage (V_s') of the hysteresis is not much affected by the light illumination. The temperature variation of the I-V curves in the dark is shown in the inset of Fig. 1. It is clear that the photoeffect is opposite to what the laser heating might have caused.

Considering the penetration depth of the visible light ($\sim$0.1 μm [13]), the effect of light does not reach very deep. Although the electrodes are evaporated on the surface of the crystal, the major portion of the current flows deep in the bulk without being affected by the photoillumination. In order to see the photoeffects more clearly, we repeated the experiment with a gap separation of 50 μm. From a simple geometrical consideration, the current contribution near the sample surface should be proportionately

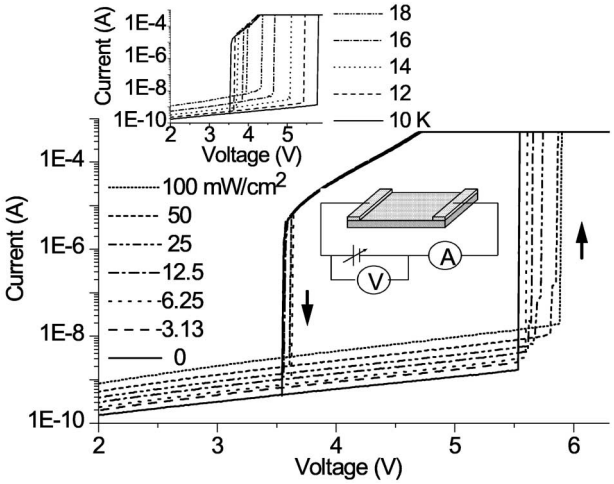

FIG. 1. Change of I-V characteristics of a sample with a large electrode gap (1.17 mm) by light illumination measured at 12 K. The slide voltage and the creep current increase with illumination intensity. Inset shows the temperature dependence of the I-V curves and the cross-sectional view of the contact arrangement.

enhanced in the narrower gap. The data shown below were all taken with these samples with enhanced surface sensitivity.

Figure 2 shows the evolution of the I-V curves under laser illumination of varying intensity. In contrast to the sample with a larger gap, a clear switching behavior is absent for the I-V curve in the dark. A possible cause for the difference will be discussed later. On illumination with the laser light, however, the switching behavior is recovered. As is the case with the sample with a wider gap, the increase of the creep current and pronounced shift of V_s as a function of the light intensity are observed. However, the hysteresis is rather small and of the order of 10 mV for 100 mW/cm^2 excitation (not shown in the figure for clarity).

The effect of the light illumination is analyzed quantitatively as follows. The current below V_s can be fitted well with thermal creep [14],

$$j = \sigma_0(V - V_0)\exp\left(\alpha \frac{V}{T}\right), \qquad (1)$$

as shown in Fig. 3. The prefactor σ_0 depends linearly on the light intensity as presented in the upper inset of Fig. 3, while other parameters in Eq. (1) remain essentially unchanged. The sliding voltage V_s [defined as the voltage at which the I-V curve deviates from the creeping behavior, Eq. (1)] increases with the light intensity as shown in the lower inset of Fig. 3 [15].

The obvious question to ask first is if all these phenomena are due to laser heating. The temperature rise of the sample surface is estimated to be 0.01 K for the light intensity of 100 mW/cm^2 using the known thermal conductivity [16]. This is in good agreement with the temperature rise of 5 to 10 K encountered in Raman scattering measurements in which the intensity is 3 orders of magnitude higher (typically 50 mW focused into a 0.1 mm diameter spot). As is illustrated in the inset of Fig. 2, the creep current increases with temperature since it is activated thermally. However, in order to cause the creep current seen under 100 mW/cm^2 illumination, the sample temperature rise must be 6.6 K, which is highly unlikely. The change of the hysteresis curves for the sample with a larger gap under the light illumination is also qualitatively different from the temperature effect as noted before. The increase of V_s (lower inset of Fig. 3) is opposite to the temperature effect (Fig. 2, inset) and its nearly vertical rise at the low light level is by no means explainable by the temperature rise. Therefore, the laser heating scenario is safely rejected.

The seemingly contradictory effects, the increase of the creep and V_s at the same time, can be understood only as a photoinduced modification of the dynamic transition from slide to creep. Because the photoexcitation is so weak, it is unlikely that the light affects the overall amplitude of CDW; at 100 mW/cm^2 only 16 photons are absorbed per second per unit cell. Using the fast (<10 ps [17]) relaxation time of the photogenerated quasiparticles back to the condensate, the average change of the amplitude due to the light illumination is estimated to be less than 10^{-9}. However, there is one mechanism by which the slow arrival of photons can affect the overall transport. This is by acting on the pinning centers. Let us imagine that a photon happens to destroy the condensate in the highly distorted region near a pinning center. The local CDW amplitude recovers quickly as stated above but the quasiparticles should recondensate into a less deformed phase configuration bypassing the pinning potential because it is

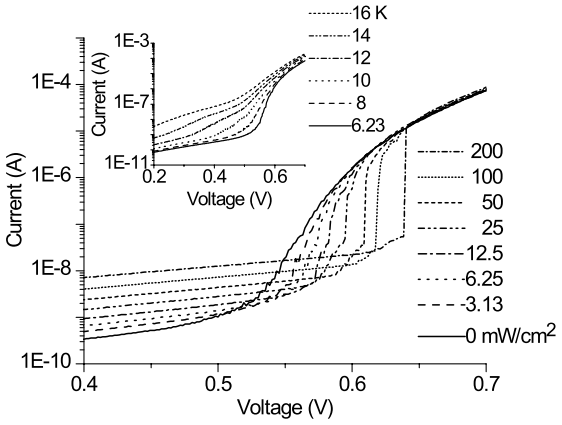

FIG. 2. Change of I-V characteristics of a sample with a small electrode gap (50 μm) by light illumination measured at 7.1 K. Inset shows the temperature dependence of the I-V curves.

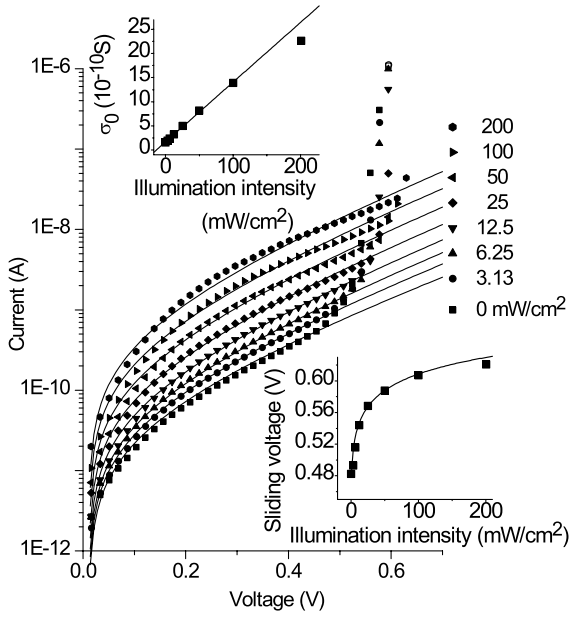

FIG. 3. I-V curves in the creeping regime shown in Fig. 2. The solid lines are fit to Eq. (1). The upper inset is the light intensity dependence of the prefactor, σ_0. The lower inset is the light intensity dependence of the slide voltage [defined as the voltage at which the I-V curve departs from the creep motion, Eq. (1)]. The slope seems to diverge at low light intensity. The curve, proportional to the logarithm of the light intensity, is a guide to the eye.

energetically more favorable. The phase slip is thus efficiently promoted. Because the photoinduced phase slip effectively stabilizes the creep regime, the dynamic phase transition into the sliding motion is postponed to a higher driving force. Since the rate of creep should be proportional to the rate of phase slip, the linear dependence of the prefactor σ_0 to the light intensity (upper inset of Fig. 3) is quite reasonable. The extremely nonlinear rise of V_s as a function of the light intensity (lower inset of Fig. 3) warrants further investigation because it may indicate a singularity in the I/V/light intensity space in which the dynamic phase transition is described. The data points below 100 mW/cm^2 seem to be well represented by the logarithm of the light intensity (soid curve) but we have no explanation for it. In order to augment this scenario, pulse experiments are performed.

Figure 4 presents the current response to a laser pulse (pulse duration = 5 ns, photon energy = 2 eV, 72 nJ/pulse on a 300 μm diameter spot). At low voltage in the creep regime (not shown), the immediate response to the light pulse is the current spike. The highly stressed CDW can instantaneously phase slip past the pinning center. The relaxation causes the displacement current, i.e., a current spike in the forward direction. The spike height is proportional to the voltage as one might expect. As the CDW starts to slide above V_s, the spike is gradually taken over by a subsequent dip. A photoinduced dynamic transition from the sliding state to the creeping state occurs. At this stage, the stress relaxation of the local deformation and braking of the CDW sliding motion occur simultaneously, hence, the turnover forms a spike to a dip. The current recovery to the sliding state is independent of the pulse duration (see also Fig. 5 inset) and is much slower than the duration of the light pulse. The behavior is identical to the response of the same sample to the voltage step in the dark, implying that it is the stress buildup prior to the slip that determines the recovery dynamics [18].

The efficiency of the photoinduced dynamic transition is monitored as a function of the photon energy as shown in Fig. 5. The light source in this case is a difference frequency generated by an OPG-OPA (optical parametric generator and optical parametric amplifier). The short pulse width of 200 fs does not affect the response as is expected from the discussion thus far. The efficiency is defined as the depth of the current dip in response to a laser pulse of a fixed pulse energy relative to the slide current in the dark (Fig. 5, inset). A gap is clearly seen, which nicely coincides with the optical gap of $K_{0.3}MoO_3$ [19]. The edge is broadened partly because of the energy spread in the short laser pulse (200 fs = 20 meV). Because one needs to destroy the condensate for the photoinduced effect, the energy dependence in Fig. 5 is quite natural.

The remaining question is the source of the stress and nature of the pinning centers. There are several mechanisms that can cause the stress buildup, such as the proximity effect of the electrodes. The more "frictional" appearance of the I-V curves for the narrower gap samples may simply be due to the smearing of the dynamic phase transition due to the small sample volume or could be due to the influence of the stress near the electrodes, extensively studied by Brill *et al.* [20]. In fact, the 50 μm gap may be completely covered by the "contact related stress." The space resolved experiment is one way to answer this question. This is in progress and will be reported elsewhere [21]. The asymmetric effect of photons on V_s and V_s' for the wider gap sample is understandable because, once the major portion of the sample undergoes the transition into the sliding state, small perturbation

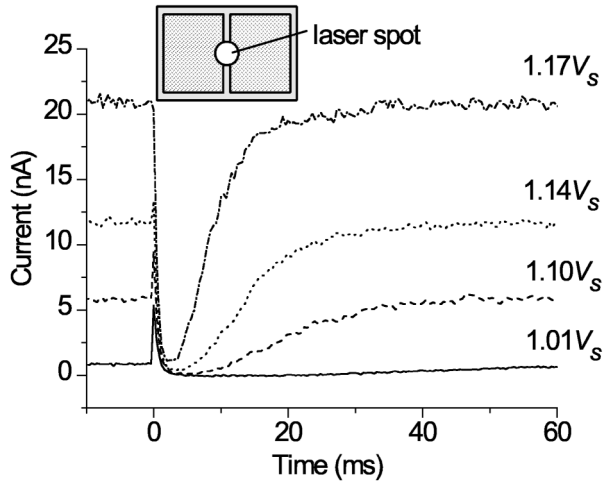

FIG. 4. Transient response of the sliding current by pulsed laser irradiation measured at 11 K. Just above V_s, both spike and dip can be seen. The applied bias voltages are 1.01V_s, 1.10V_s, 1.14V_s, and 1.17V_s, respectively. Inset shows the relative position of the laser spot and the electrodes.

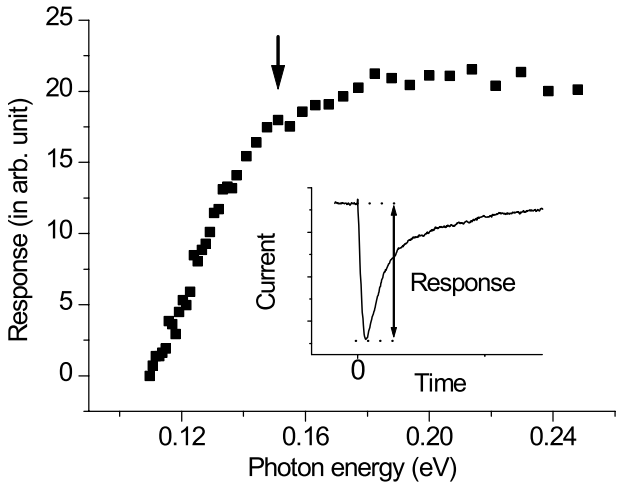

FIG. 5. Action spectrum of the transient photoresponse (as indicated in the inset) by laser irradiation measured at 7.5 K. The incident intensity is lower than 600 nJ/pulse. The transient current decrease disappears below 0.15 eV, which matches the reported optical gap indicated by an arrow.

acting only on the surface becomes irrelevant whereas its effect is fully felt in the creeping motion against a pinning site wherever it is located.

A few more remarks are in order before closing. (i) The photoinduced effect is qualitatively unaffected by the density of disorders. Samples doped with varying amount of Rb (1%, 5%, and 80%) show similar effects, although the intrinsic V_s is much higher than that of the undoped one. (ii) *In situ* x-ray diffraction indicates the photoinduced relaxation of strain which is very shallow as is expected from the small penetration depth of light [21]. (iii) We are aware of one photoconductive experiment done around the optical gap (CO_2 laser = 0.13 eV) [22]. The intensity of the light ($\sim 1-10$ W/cm^2)cm^2) used was orders of magnitude larger than the current experiment, and we believe that a direct comparison with our observation is not feasible.

In conclusion, we observed photoinduced dynamic phase transition from slide to creep states. We attributed this effect to the photoinduced phase slip and redistribution of the CDW phase. The drastic change of the conductance indicates the long-range coherence of the sliding CDW and its sensitivity to the phase deformation localized only near the sample surface.

We thank Professor N. Nagaosa, Professor Y. Murakami, Professor T. Tamegai, and Professor H. Matsukawa for valuable comments and discussions. The work was supported in part by Grant-in-Aid for COE Research of the Ministry of Education, Culture, Sports, Science and Technology.

[1] For a review, see, for example, G. Grüner, Rev. Mod. Phys. **60**, 1129 (1988); G. Grüner, *Density Waves in Solids* (Addison-Wesley, Longmans, MA, 1994).
[2] G. Blatter *et al.,* Rev. Mod. Phys. **66**, 1125 (1994).
[3] H. Fukuyama and P. A. Lee, Phys. Rev. B **17**, 535 (1977).
[4] An earlier description of the transition from the pinned to the sliding state in CDW in terms of a dynamic phase transition is given by D. S. Fisher, Phys. Rev. Lett. **50**, 1486 (1983). Some aspects of the theoretical prediction have been numerically confirmed [e.g., H. Matsukawa, Synth. Met. **29**, F343 (1989)].
[5] A. B. Kolton, D. Dominguez, and N. Gronbech-Jensen, Phys. Rev. Lett. **86**, 4112 (2001).
[6] L. Balents and M. P. A. Fisher, Phys. Rev. Lett. **75**, 4270 (1995).
[7] A. E. Koshelev and V. M. Vinokur, Phys. Rev. Lett. **73**, 3580 (1994); F. Pardo *et al.,* Nature (London) **396**, 348 (1998).
[8] For the case of Abrikosov lattice, see, e.g., Y. Paltiel *et al.,* Nature (London) **403**, 398 (2000).
[9] For the case of CDW, see, e.g., A. Zettl and G. Grüner, Phys. Rev. B **29**, 755 (1984).
[10] A. Wold *et al.,* Inorg. Chem. **3**, 545 (1964).
[11] L. Mihaly, K.-B. Lee, and P. W. Stephens, Phys. Rev. B **36**, 1793 (1987).
[12] A. Maeda, M. Notomi, and K. Uchinokura, Phys. Rev. B **42**, 3290 (1991).
[13] L. Degiorgi *et al.,* Phys. Rev. B **44**, 7808 (1991).
[14] S. G. Lemay, R. E. Thorne, Y. Li, and J. D. Brock, Phys. Rev. Lett. **83**, 2793 (1999).
[15] The sliding voltage V_s may be defined as the voltage at which $d\log I/d\log V$ is maximum. The appearance of V_s vs light intensity remains the same as in Fig. 4 except for an overall shift of the ordinate.
[16] R. S. Kwok and S. E. Brown, Phys. Rev. Lett. **63**, 895 (1989).
[17] J. Demsar, K. Biljakovic, and D. Mihailovic, Phys. Rev. Lett. **83**, 800 (1999).
[18] J. Levy and M. S. Sherwin, Phys. Rev. B **43**, 8391 (1991).
[19] G. Travaglini *et al.,* Solid State Commun. **37**, 599 (1981).
[20] B. M. Emerling, M. E. Itkis, and J. W. Brill, J. Phys. IV (France) **9**, Pr10-125 (1999), and references therein.
[21] N. Ogawa *et al.* (to be published).
[22] R. Gaal *et al.,* Phys. Rev. Lett. **69**, 1244 (1992).