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STM OBSERVATION OF SLIDING MOTION OF CDW IN $K_{0.3}MoO_3$

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The sliding motion of CDW was investigated with the STM observation in a $K_{0.3}MoO_3$ single crystal. For bias current exceeding the threshold value for CDW depinning, sharp peak was found in the spectra of tunneling current with fixing the tunneling tip position. The peak frequency increased with the current carried by the CDW condensate. This peak was explained by the modulation of tunneling current due to the sliding motion of CDW; the sliding motion of CDW at the surface was observed for the first time. The sliding velocity was obtained directly. It was confirmed that the CDW begins to slide at the surface just above the threshold current with much higher velocity than the interior of sample.

SINCE THE sliding of CDW condensate was proposed for the non-linear conduction with a threshold electric field, many experimental researches and theoretical ones have been done in several quasi-one-dimensional materials such as $NbSe_3$, TaS_3 and $K_{0.3}MoO_3$, up to now. Through various characteristic phenomena such as the non-linear electric conduction, the frequency dependent conductivity, the narrow band noise and the a.c.–d.c. interference phenomena, the sliding motion of CDW has been explained as that of a rigid CDW [1] qualitatively and at the same time it has become recognized that the degree of freedom of CDW deformation [2] is important. The sliding motion itself was confirmed by the NMR experiment clearly [3]. But the controversy is continued [4] about the origin of narrow band noise which is considered to reflect the sliding velocity directly.

The STM (Scanning Tunneling Microscope) has demonstrated its usefulness by direct observation on the image of reconstruction of Si surface recently [5]. This technique serves various possibilities for investigation of surface of materials. Measurements by STM in CDW materials with two dimensional structure have been reported; the superlattice structure due to CDW was successfully observed together with the atomic image [6, 7]. These results suggest that the STM can reveal both the structure of CDW and the sliding motion of CDW in quasi-one-dimensional conductors as well. The STM has its non-contacting tunneling tip for a probe. Therefore, we can observe the sliding state of CDW with much less disturbance than the usual electric measurements.

In the present letter, we report the first STM observation of sliding motion of CDW in $K_{0.3}MoO_3$, with the incommensurate wave vector below 180 K, and discuss the sliding velocity of CDW from the tunneling current spectra.

A single crystal of $K_{0.03}MoO_3$ was prepared by the electric reduction method from the melt of K_2MoO_4 and MoO_3 [8]. The sample crystal was cut and cleaved to a rectangular shape. After the electric leads were attached to the sample with indium metal by using an ultra-sonic bonder, the sample was cleaved again parallel to the $(\bar{2}, 0, 1)$ plane to obtain a clean surface. The sample dimension was $2.38 \times 0.43 \times 0.18\,mm^3$. We adopted the two probe method in the electric measurement.

The principal part of our STM unit is made of a scanning tip of tungsten and a tube type actuator. The sample was pasted with epoxy adhesive on the holder whose position is able to be adjusted coarsely by a screw. The scanning tip was arranged to be normal to the cleaved surface of sample and the distance of the tip from the sample surface was controlled finely by the tube actuator. The scanning tip was also scanned along the sample surface by the tube actuator. The tunneling unit was laid in a cell filled with thermal exchange helium gas. The temperature was controlled in a metal dewar. To avoid vibration the tunneling tip and the metal dewar were mounted on a vibration isolator, which was equipped with air suspensions. The metal dewar was immersed in liquid nitrogen filled in another outer dewar laid on the floor. The temperature was measured by using a platinum resistance thermometer near the sample in the cell. The obtained temperature, 80 K, was a little higher than the liquid nitrogen temperature.

We measured the current–voltage curve in the

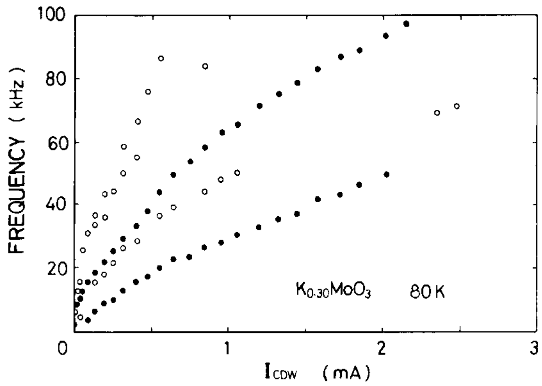

Fig. 1. Frequencies of narrow band noise vs current carried by CDW condensate. Solid and open circles represent two different fundamentals and their corresponding second harmonics.

sample being set in the tunneling unit. Under the current control condition, the voltage noise spectra (narrow band noise) were also observed with use of a FFT analyzer (Iwatsu SM2700). It has a function of Fast Fourier Transform of the digitized voltage signal with a band width of 100 kHz.

In the tunneling measurement, we adopted the current detection mode, where the tip height was controlled to be constant during the transverse scanning and the variation of tunneling current was detected. In the measurement under dc bias current, we fixed the tip position and monitored the tunneling current signal. The tunneling current was analyzed with the same FFT analyzer as for the narrow band noise measurement. The observed spectra of tunneling current were accumulated with a computer for improving the signal to noise ratio.

We obtained the non-linear current–voltage curve with the threshold field $E_T = 88\,\mathrm{mV\,cm^{-1}}$ in the sample of $K_{0.3}MoO_3$ at 80 K and could observe the narrow band noise under a current exceeding the threshold value. Two fundamental peaks and their harmonics were found in the noise spetra. Their frequencies increased with the current carried by the CDW as shown in Fig. 1. According to the classical sliding model, the following well-known relation holds between the fundamental frequency F_1 and the current density carried by CDW J_{CDW},

$$F_1/J_{CDW} = 1/(en\lambda), \tag{1}$$

where n and λ represent the electronic density condensed to the CDW and the wavelength of CDW, respectively. In $K_{0.3}MoO_3$, this ratio F_1/J_{CDW} have been obtained in the homogeneous sample to be $11\,\mathrm{kHz\,A\,cm^{-2}}$ at 77 K [9]. The values of fundamental frequencies in the present experiment were much larger than the above value in the homogeneous crystal. Moreover, the linear relation does not hold precisely in our present sample. These results are attributed to the inhomogeneous current flow of CDW in the present sample; the CDW is sliding not through the whole cross sectional area of the sample but through a limiting area. When there are some portions of the cross section, where the CDW is pinned and is not sliding, the averaged current density always causes an under-estimation of real current density of CDW, because we know only the average current density for a sample. Therefore, the higher value of F_1/J_{CDW} was brought about in our present sample. The observed two fundamental peaks suggest that there are at least two portions where the CDW is sliding with different velocity from each other. While, the nearly linear dependence show that the areas, through which the CDW is sliding, are not much changed with increasing the current. But the bend of F_1–J_{CDW} curve in Fig. 1 suggests the existence of another portion, where the CDW is sliding with a different velocity.

We tried to observe the image of CDW by the STM, but without success to obtain the atomic image nor the CDW superlattice structure. It is presumably because of the surface contamination. We analyzed the tunneling current spectra with scanning the tip. But the obtained signal was too poor to discuss the CDW structure. The control of gas absorption is the future problem to obtain a clear STM image in this material.

We investigated carefully the tunneling current spectra with fixing the tip position and found a relatively sharp peak under dc bias current. As shown in Fig. 2, a new peak (indicated by arrows in the figure) emerged in the spectra of tunneling current for current exceeding the threshold value ($I_c = 0.95\,\mathrm{mA}$). The peak position shifted to the high frequency with increasing the current. In the spectra, we find several extremely sharp peaks with large amplitudes. These peaks were observed even below the threshold current and neither their shape nor amplitude changed for varying bias current. Therefore, we can distinguish the relatively sharp peak from these peaks, which are attributed to the system noise mainly generated by the STM control circuit and the computer.

In Fig. 3, we show the dependence of peak frequency on the CDW current. The latter was calculated from the current–voltage curve. The current dependence of peak frequency is somewhat different from that of the narrow band noise. The frequency of the peak was rapidly increased just above the threshold current and showed the tendency of saturation with increasing the current. The frequency of the peak is 10 times just above the threshold and about twice at

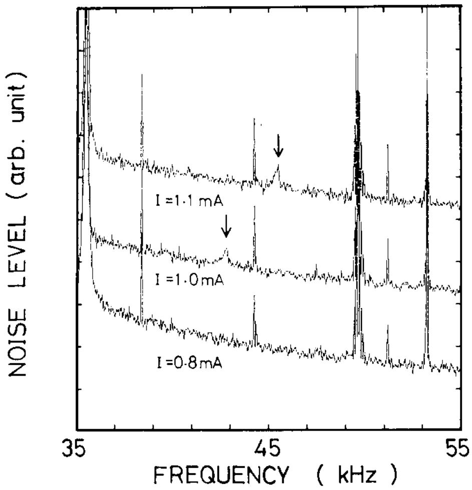

Fig. 2. Spectra of tunneling current under bias currents. The position of tunneling tip was fixed and the tunneling current was analyzed. For currents exceeding the threshold value of 0.95 mA, a new peak (indicated by arrows) appears in the spectra.

$I_{CDW} = 1.5$ mA as large as that of the narrow band noise.

According to the simple tunneling theory, the tunneling current is due to the overlap of the electronic wave function between the tip metal and the sample surface, which are separated by vacuum, and is expressed as,

$$I = I_0 \exp(-d/d_0), \tag{2}$$

where d is the distance between the tip and the nearest atom of sample. The characteristic current I_0 contains the product of electronic state densities in tip metal and the sample and the length d_0 characterizes the extension of electronic wave function. If the sample is normal metal with a constant density of electronic states, the tunneling current is proportional to the bias voltage for small bias. But our sample is a Peierls type semiconductor. In the CDW phase of Peierls semiconductor, e.g. $K_{0.3}MoO_3$, the tunneling current-voltage has to show a gap structure* similar to that of a superconductor [10]. For the bias voltage lower than the Peierls gap, no current can flow at $T = 0$ K. While at finite temperature, the tunneling current is associated with the thermally activated normal carriers. Our experiment was carried out with the tunneling voltage less than the Peierls gap. It is understood that the tunneling current flows between the metallic band of tip and the states of thermally excited electrons across the Peierls gap in the sample material. Therefore, the CDW condensate is not converted to normal electrons even at the nearest position to the tip. Accordingly, we can observe the CDW with a relatively small disturbance. Then, the tunneling current is mainly determined by the distance d.

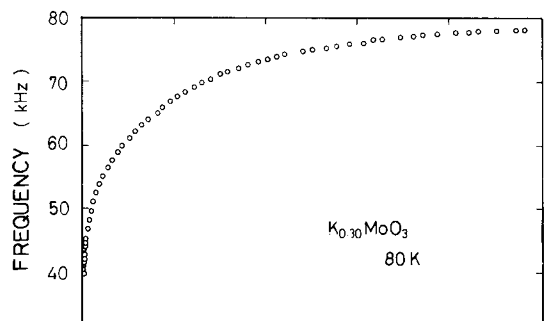

Fig. 3. Peak frequency in tunneling current spectra vs CDW current.

The CDW state is described by the electronic charge density as,

$$\varrho(x) = \varrho_0 + \varrho_1 \cos(Qx + \phi), \tag{3}$$

where Q represents the wave number of CDW and is equal to $2k_F$. The phase ϕ denotes the position of CDW in a rigid CDW model. Correspondingly, the lattice displacement $\mathbf{u}$ is described as,

$$\mathbf{u}(x) = \mathbf{u}_1 \cos(Qx + \phi). \tag{4}$$

Therefore, the sliding motion of CDW modulates the distance d periodically. As the hill and valley of CDW pass near the tunneling tip alternatively, the overlap of electronic wave functions is modulated with a time period $\tau = \lambda/v$, where v is the sliding velocity of CDW. Thus the tunneling current is modulated with a frequency $f = 1/\tau$. Therefore, the modulation frequency of tunneling current satisfies the same relation as the narrow band noise (equation (1)).

The observed peak in the tunneling current clearly reveals the above mentioned mechanism and supplies the most direct evidence for the sliding motion of CDW. We estimate the sliding velocity of CDW at the surface from the peak frequency of tunneling current. Using the relation $v = \lambda f$, we obtained $v_s = 5.7 \times 10^{-3}\,\mathrm{cm\,s^{-1}}$ at $I_{CDW} = 0.1$ mA and $v_s = 7.8 \times 10^{-3}\,\mathrm{cm\,s^{-1}}$ at $I_{CDW} = 2$ mA. The sliding velocity of CDW in the interior of sample v_b is calculated as $v_b = 6.3 \times 10^{-4}\,\mathrm{cm\,s^{-1}}$ at $I_{CDW} = 0.1$ mA and $v_b = 5.1 \times 10^{-3}\,\mathrm{cm\,s^{-1}}$ at $I_{CDW} = 2$ mA, from the low frequency component of fundamental frequencies of narrow band noise (Fig. 1). Accordingly, the velocity of CDW is much higher at the surface than the interior of sample. These results show that

* Such a tunneling spectroscopy in $K_{0.3}MoO_3$ will be discussed elsewhere.

just above the threshold the CDW begins to slide at the surface of sample, while the CDW is pinned or sliding but with very small velocity in the interior region of sample. The non-linear current mainly flows through the sample surface just above the threshold. It might be probable that the CDW is pinned and cannot slide at the surface by some reasons. The surface energy may pin the CDW and, more directly, the electric leads attached to the sample surface pin the CDW. However, our results clearly show that the CDW is easy to slide at the surface. This sliding at the surface may be proper to the two-lead configuration.

The width of peak in the tunneling current spectra is much narrower than that of the narrow band noise, but broader than that of the system noises. The modulation of tunneling current is generated at the gap point between the tip and the sample. Therefore, as far as the sliding velocity of CDW is constant, the peak must have an infinitesimal width. We averaged the spectra of tunneling current for about 1 min to obtain the clearer spectra. The observed width is equivalent to the sliding velocity fluctuation of 4×10^{-3} for 1 min. The stability of the current generator used in our experiment, better than 1×10^{-4}, cannot explain the observed width. Although it has not been made clear whether the observed width comes from a rapid fluctuation of sliding velocity or from a gradual change in metastable CDW configurations, it is clear that such a time variation of sliding velocity is present even for a constant bias current.

In summary, we investigated carefully the tunneling current with fixing the tip position in the STM measurement and found a relatively sharp peak in the spectra of tunneling current for bias current exceeding the threshold value. This peak is generated by the sliding CDW; the sliding motion of CDW at the surface was observed for the first time. It is confirmed that the CDW at the surface is easy to move and the sliding velocity is much larger at the surface than that in the interior of the crystal.

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