Depining Process and Dynamic Phase Transition of Charge-density Wave in $K_{0.3}MoO_3$

The homogeneous phase in low-dimensional materials, such as $K_{0.3}MoO_3$, $NbSe_3$, 2H-$NbSe_2$ and TMTSF molecules, undergoes a phase transition to charge-density waves (CDWs) at low temperatures. The instability toward spontaneous formation of charge-density modulations is driven by electron-phonon or electron-electron interactions [1]. Among many other interesting aspects of CDWs, the transport property has attracted much attention from both experimental and theoretical sides. It is generally believed that current is suppressed at a small bias voltage, where the CDW is pinned by impurity potential. Above some threshold voltage, the sliding motion along the applied electric field starts and the current increases significantly. It is known that this dynamic behavior involves the phase slippage of the density waves [2]. In addition, the dynamics of the disordered state driven by an external force in a periodic medium are also of the characteristic common to many physical systems, such as density waves and vortex lattices [3].

An interesting question naturally arises: How does the internal structure of density modulations change in the presence of the applied electric field? In particular, in the non-equilibrium sliding motion the robust periodic structure, originally developed in static limit, might become unstable. In this report, we address this issue by applying the techniques of multiple (three-wave) X-ray diffraction and X-ray scattering to measure the changes in the CDW lattice and its internal structure with respect to the applied electric fields. From the extracted phase $\delta_3(V)$ of the three-wave diffraction and the peak profile of the CDW satellite reflection, we demonstrate that the occurrence of nonlinear conductivity caused by the periodic media is through the internal deformation of the CDW lattice, i.e., a phase jump of 2π, as the applied voltage exceeding the threshold. From the evolution of the measured peak width of satellite reflections as a function of the field strength, we also report that the CDW lattice can be driven to move and undergo a dynamic phase transition from a disordered pinning state to an ordered moving solid state and then to a disordered moving liquid.

A good-quality single-crystal $K_{0.3}MoO_3$ was first characterized and oriented at room temperature. The non-linear conductivity [4] was confirmed by measuring the *I-V* curve at 70 K, as shown in Fig. 1(a), indicating the dynamic transition from the pinned CDWs to sliding motions at the threshold observed at $V_c \approx 0.165$ V. It is noteworthy that the nonlinearity is not as robust as that at low temperature due mainly to the thermal fluctuations smearing out the switching behavior.

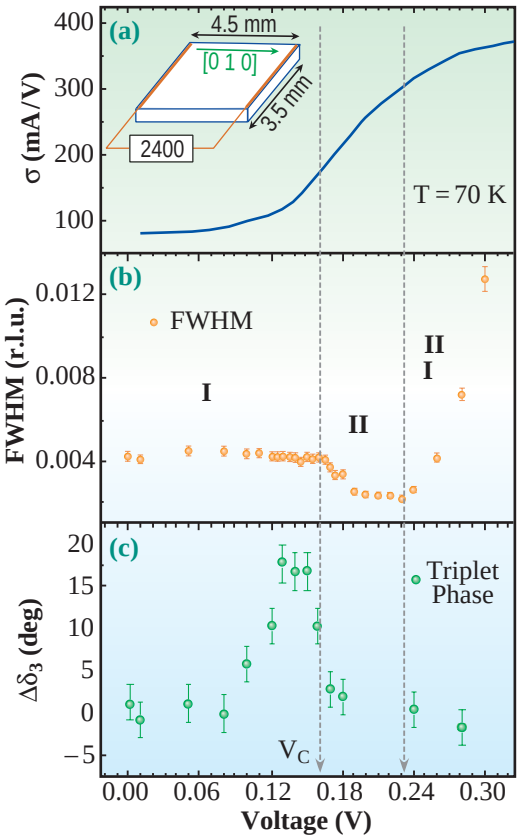

Fig. 1. **(a)** Resistance versus voltage at 70 K. The curve shows the nonlinear behavior with a threshold voltage $V_c \approx 0.165$ V. The inset shows the dimensions of the sample and the experimental setup. **(b)** Evolution of the FWHM of the CDW satellite reflection as a function of applied voltage. One can classify CDW into three phases: (I) the creeping CDW state, (II) the moving solid and (III) the moving liquid. **(c)** Triplet phase $\Delta\delta_3$ at different voltages. Note that in the sliding phase, $\Delta\delta_3 = 0$ is direct evidence that the pinning forces become irrelevant.

The *in situ* measurements were carried out in the Taiwan beamline **BL12B2**. The incident X-ray wavelength was selected to be 1 Å. Satellite reflections signal the formation of CDW, located at the Bragg position G = (13 q 6.5), where $q \approx 0.748$. Scans were performed through the longitudinal direction of [2 0 -1], and the data was convoluted with the resolution function obtained from the nearby Bragg peak (12 0 -6). Figure 1(b) shows the evolution of the peak width of CDW reflection as a function of applied voltage at T = 70 K.

As shown in Fig. 1(b), the FWHM of the satellite reflection, (13 q -6.5), remains more or less unchanged below the critical voltage $V_c \approx 0.165$ V determined from the transport measurement. Above the critical voltage, where the CDW enters the sliding phase, the FWHM decreases as predicted by previous theoretical investigations. As the voltage exceeds 0.18 V, the lattice shows a long-range ordered state; namely, the moving solid phase [5]. This interesting dynamical narrowing of FWHM is a strong indication that the pinning forces due to random potentials become irrelevant (or less efficient) when the CDW begins to slide. When the voltage exceeds 0.22 V, nonequilibrium effects, amplitude fluctuations and other factors become important and the Bragg peaks disappear. This is clearly evidenced by the sharp increase of FWHM.

The other key quantity we studied is the triplet phase δ_3 of a 3-wave multiple diffraction at different bias voltages. To set up a 3-wave ($\boldsymbol{O}$,$\boldsymbol{G_1}$,$\boldsymbol{G_2}$) multiple diffraction experiment, the crystal is first aligned for a primary reflection $\boldsymbol{G_1}$. It is then rotated around the reciprocal lattice vector $\boldsymbol{G_1}$ with an azimuthal angle ψ to bring in the secondary reflection $\boldsymbol{G_2}$, that also satisfies Bragg's law. Namely, both $\boldsymbol{G_1}$ and $\boldsymbol{G_2}$ reflections take place simultaneously. $\boldsymbol{O}$ stands for the incident reflection. The interaction of the multiply diffracted waves modifies the intensity of the primary reflection. Intensity variation showing an asymmetric distribution versus y gives the required information about δ_3, which is the triplet phase of $F_{G2}\,F_{G3}\,/\,F_{G1}$ involved in the 3-wave diffraction. Previously [6], we demonstrated that the triplet phase δ_3 due to the coupling between the CDW lattice and its host lattice can be probed using multiple diffraction. Here we further demonstrate that measuring the relative change in δ_3 , $\Delta\delta_3$, caused by a driving force makes the study of the internal deformation of the CDW lattice possible.

Through the azimuthal scan around the primary reflection $\boldsymbol{G_1}$ = (13 q -6.5) at T = 70 K, we obtained the 3-wave diffraction pattern containing numerous multiple diffraction peaks. We concentrated here only on a particular 3-wave diffraction, (0 0 0), (13 q -6.5) and (4 -8 4) at ψ = 108.53º, where the primary reflection is (13 q -6.5) and the coupling reflection is (9 8 +q 10.5). The profile asymmetry of the diffraction intensity of (13 q -6.5) versus ψ at V = 0 is typical for δ_3 = 0. The phase variation $\Delta\delta_3$ due to nonzero applied voltages was analyzed based on the dynamical theory for multiple diffractions [7]. In Fig. 2, the profile develops different asymmetries at different finite voltages. In the static CDW state, the crystal lattice possesses a centrosymmetric structure, and a change in the peak profile means that this centrosymmetry is broken due to the relative motion of ions by a driving force. This results in a nonzero $\Delta\delta_3$, suggesting the internal deformation of the original charge density distribution by the driving force. As shown in Fig. 1(c), the estimated $\Delta\delta_3$ values from curve fitting at 0.1, 0.12 0.13, 0.14, 0.15, and 0.16 V are about 6º, 10º, 18º, 17º, 17º, and 10º respectively, and then back to 0º for $V > 0.18$ V. This proves experimentally that the occurrence of nonlinear transport behavior is through a phase jump of 2π at the sliding threshold, i.e., from the pinned to sliding states [8].

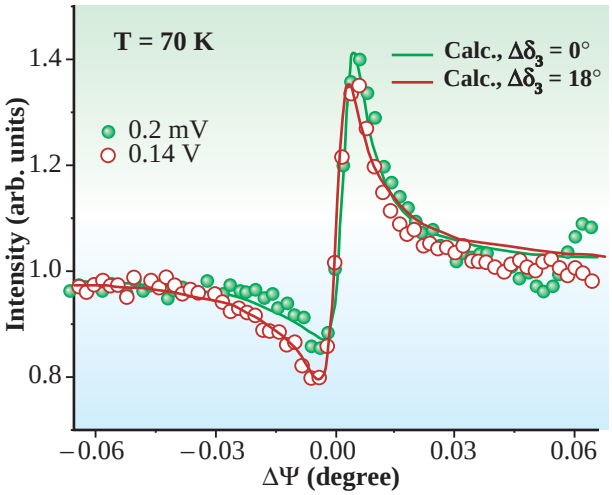

Fig. 2. Triplet phase change $\Delta\delta_3$ extrapolated from curve fitting of the 3-wave diffraction profiles of the primary reflection $\boldsymbol{G_1}$ = (13 q -6.5) at V = 0.2 mV and 0.14 V. The analysis is based on the dynamical theory for multiple diffractions [7].

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