Direct measurement of spatial distortions of charge density waves in $K_{0.3}MoO_3$

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Using x-ray scattering and multiple diffraction on a charge density wave (CDW) material, $K_{0.3}MoO_3$, under applied voltages, 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 exceeds 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 the disordered pinning state to ordered moving solid state and then to disordered moving liquid. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2213198]

The homogeneous phases in low dimensional materials, such as $K_{0.3}MoO_3$, $NbSe_3$, and Bechgaard salt molecules, undergo 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 interactions, or sometimes electron-electron ones.[1] Among many other interesting aspects of CDW, transport property in the presence of finite driving electric field has attracted lots of attention from both experimental and theoretical sides. It is generally believed that the current is suppressed at small biased voltage, where the CDW is pinned by impurity potential. Above the threshold voltage, the sliding motion along the applied electric field starts and the current increases significantly. It has been known that this dynamic behavior involves the phase slippage of the density waves. In fact, similar phenomena occur in many other systems, such as moving vortex lattice,[2] Wigner crystal,[3] charge/spin stripes in manganites, high-T_C superconductors,[2,4] $La_{2-x}Sr_xNiO_4$,[5,6] and so on.

Theoretically, investigations[7,8] predicted that the pinning forces become irrelevant when the system enters the sliding phase. This indeed provides a natural explanation for the dynamical narrowing of the half-width above the threshold voltage. However, to understand how the CDW adjusts to the pinning forces at different driving voltages, a direct measurement for the spatial distortions of CDW is desirable. In this letter, in addition to the usual transport and full width at half maximum (FWHM) measurements of the CDW satellite reflections, we establish the connection between the lattice distortions and the triplet phase in x-ray scattering and thus demonstrate how the spatial distortions of CDW can be measured directly. While the sliding transition is already well studied, the technique we developed here can be applied to general periodic media driven by external sources and provides a new perspective into many interesting strongly correlated systems.

A single crystal $K_{0.3}MoO_3$ of good quality was prepared for the transport measurement and x-ray scattering. The crystal structure belongs to the monoclinic with the space group $C2/m$. The lattice parameters of $K_{0.3}MoO_3$ are $a = 18.162$ Å, $b = 7.554$ Å, $c = 9.816$ Å, and $\beta = 117.393°$.[9] The sample was characterized with a mosaic width of $\sim 0.005°$ and a transition temperature of ~ 180 K, and prealigned using an x-ray rotating anode source so that the scattering plane coincided with $a^* \times c^*$ plane. The *in situ* measurements were carried out on the Taiwan beam line BL12B2 of SPring-8 synchrotron facility. The incident x-ray wavelength was selected to be 1 Å. Two gold stripes spaced about 3 mm were evaporated onto the sample surface as shown in the inset of Fig. 1. The sample was then glued on the cold head of a cryostat mounted on a six-circle diffractometer. The voltage was applied along the b^* axis ([010] direction). A Keithley 2400 source meter was used to generate the driving voltage, and the *I-V* curve was measured by the two-probe setup.

Figure 1 shows the nonlinear conductivity of the sample at $T = 70$ K, indicating the dynamical transition from the pinned CDWs to sliding motions. While the nonlinearity is not as robust as at low temperatures, the current below the critical voltage $V_C \sim 0.165$ V can be fitted remarkably well with the prediction from thermal creep.[10–14] To make the critical transition more transparent, one can plot dR/dV (as shown in the inset of Fig. 1), which shows clear singularity near the critical voltage V_C. The nice fit with the thermal creep behavior indicates that our two-probe measurement does not suffer poor contacts or serious current inhomogeneity in the sample. One may notice that there is no switching phenomenon in our measured *I-V* curve due to thermal fluctuations at $T = 70$ K.[15,16]

In addition to the transport measurement, the evolution of CDW satellite reflections as a function of applied fields was also probed using x-ray scattering. The widths of the

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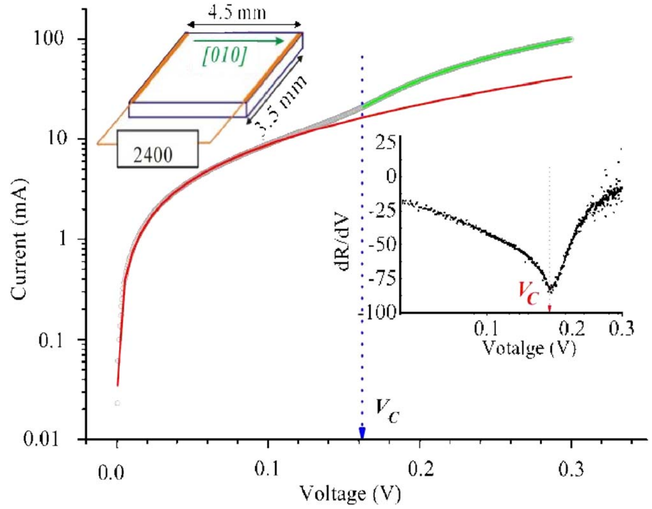

FIG. 1. (Color online) *I-V* characteristic of $K_{0.3}MoO_3$ in a two-probe transport setup at $T=70$ K. The red line is the fit to the predicted thermal creeps when the CDW is pinned, and the green line shows the *I-V* for $V > 0.165$ V. The left inset shows the dimension of the sample and the experimental setup. The right inset for (dR/dV) shows a transition point at $V_C = 0.165$ V.

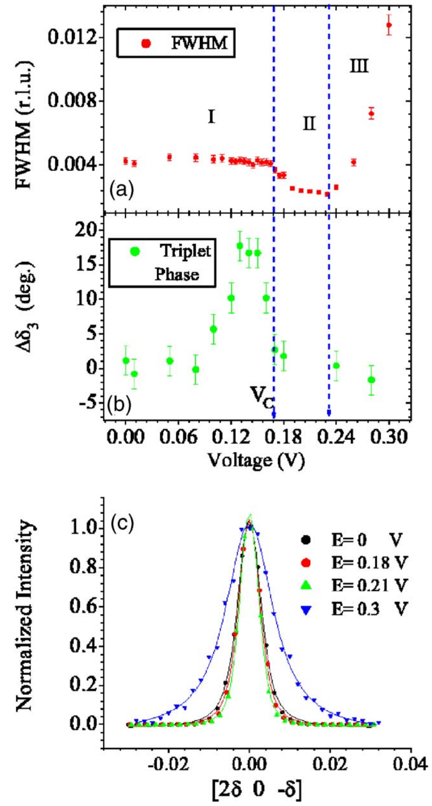

FIG. 2. (Color online) (a) Evolution of the half-width of the Bragg peak vs the applied voltage. According to the changes of the half-width, one can classify CDW into three phases: (I) the creeping CDW state, (II) the moving solid, and (III) the moving liquid. (b) The triplet phase change $\Delta\delta_3$ at different voltages. Note that in the sliding phase, $\Delta\delta_3=0$ is a direct evidence that the pinning forces become irrelevant. (c) Peak profiles at different voltages. The fits were obtained by convolution to the resolution function as measured on a nearby Bragg peak (12 0 −6). The bar shows the resolution, 0.0033 [reciprocal lattice unit (r.l.u.)], along the longitudinal direction [2 0 −1].

CDW peak and triplet phase at different driving voltages are summarized in Fig. 2. In this letter, we focus on the particular satellite reflection located at $G_1=(13q-6.5)$ with $q \sim 0.748$. Scans were performed along the longitudinal direction of [2 0 −1] and the data were convoluted with resolution function obtained from the nearby Bragg peak (12 0 −6). The FWHM of the satellite reflection, $(13q-6.5)$, in Fig. 2(a) remains more or less unchanged below the critical voltage $V_C \sim 0.165$ 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.

This can be understood as that a driving force steers a pinned lattice and results in inhomogeneous flow.[17,18] As the voltage exceeds 0.18 V, the lattice shows a long-range ordered state. A motional ordered behavior by a driving force has been reported in $NbSe_3$.[19] In contrast to the lattice existing nearby the threshold ($V_C=0.165$ V), where both pinned and flowing regions coexist, there only the flowing part remains in this moving state, namely, a moving solid phase.[14,19,20] This interesting dynamical narrowing of half-width is a strong indication that the pinning forces due to random potentials become irrelevant (or less efficient) when the CDW starts to slide.[14,19,20] When the voltage goes beyond 0.22 V, nonequilibrium effects, amplitude fluctuations, and others become important and the Bragg peaks disappear. This is clearly evidenced by the sharp increase of FWHM.

The other key quantity we studied in this letter is the triplet phase δ_3 of a three-wave multiple diffraction at different biased voltages. To set up a three-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 φ to bring in the secondary reflection $\boldsymbol{G_2}$ which 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 asymmetric distribution versus φ gives the information about δ_3, which is the triplet phase of $F_{G_2}F_{G_3}/F_{G_1}$ involved in the three-wave diffraction.[21,22] Previously,[23] 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.

The origin of azimuthal angle ($\varphi=0$) was determined to be the direction where [1 0 0] lies on the scattering plane. Through the azimuthal scan around the primary reflection $G_1=(13q-6.5)$ at $T=70$ K, we obtained the three-wave diffraction pattern containing lots of multiple diffraction peaks.[24] We concentrated here only on the particular three-wave diffraction, (0 0 0), $(13q-6.5)$, and (4 −8 4) at $\varphi = 108.53°$, 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 φ at $V=0$ is typical for $\delta_3=0$. The phase variation $\Delta\delta_3$ due to nonzero applied voltage was analyzed based on the dynamical theory for multiple diffractions.[21,22] In Fig. 3, 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 by a driving force. This results in a nonzero of $\Delta\delta_3$, suggesting the internal deformation of the original charge density distribution. The Darwin width of a

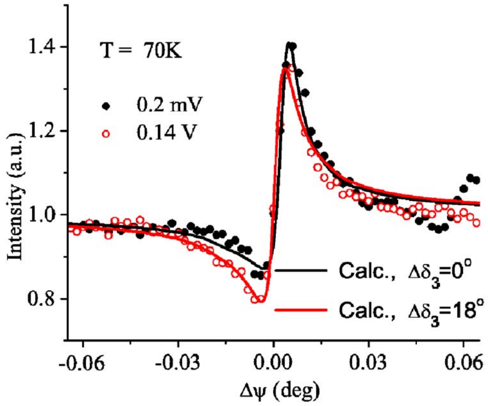

FIG. 3. (Color online) Triplet phase change $\Delta\delta_3$ extrapolated from curve fitting of the three-wave diffraction profiles. The analysis is based on the dynamic theory for multiple diffraction, giving $\Delta\delta_3=0°$ and 18° at $V=0.2$ mV and 0.14 V, respectively.

Bragg reflection was also monitored in order to make sure the crystal was not destroyed by applied voltages. As shown in Fig. 2(b), the $\Delta\delta_3$ reaches the top at $V=0.12-0.14$ V. This can be understood as that the internal distortion of CDWs is saturated just before the sliding motion. Classically, upon the application of bias voltage, the free energy is minimized by the elastic energy cost due to CDW distortion. After threshold, it is energetically favorable to slid (increasing kinetic energy) rather than hold up the large elastic energy. The estimated $\Delta\delta_3$ from curve fittings at 0.1, 0.12, 0.13, 0.14, 0.15, and 0.16 V are about 6°, 10°, 18°, 17°, 17°, and 10°, respectively, and then back to 0° for $V>0.18$ V, as shown in Figs. 2 and 3. This evidences 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.[25,26]

All experimental results given above evidence the correlation of the nonlinearity and the dynamic phase transition of a periodic medium. The phase measurement using three-beam diffraction is also demonstrated for the first time as a sensitive method to study the dynamic phenomena in nonlinear systems. While it is already exciting to observe these dynamic motions of CDW, it also opens up many interesting issues requiring further studies.

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