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Field-induced dielectric response saturation in *o*-TaS₃Yongchang Ma^{1,2,3,*}, Mengmeng Ma^{1,4}, Cuimin Lu^{1,4}, Xuewei Wang¹, and Cedomir Petrovic²¹ *School of Materials Science and Engineering, Tianjin University of Technology, Tianjin 300384, China;*² *Condensed Matter Physics and Materials Science Department,
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The temperature and electric field dependent conductivity spectra of *o*-TaS₃ sample with $10\ \mu\text{m}^2$ in cross section were measured. Besides the classical electric threshold E_{T-Cl} , we observed another novel threshold E_{T-N} at a larger electric field, where an S-shaped I - V relation revealed. The appearance of E_{T-N} may be due to the establishment of coherence among small charge-density-wave domains. Under a stable field $E > E_{T-N}$, a sharp dispersion emerged below kHz. At a fixed temperature, the scattering rate of the charged condensate was extremely small and decreased with increasing field. With decreasing temperature, the scattering rate decreased and in zero temperature limit, the Fröhlich conductivity would be expected to occur. We proposed that the Fröhlich-mode conductivity would be consistent with the meta-stable state.

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I. INTRODUCTION

The nonlinear conductivity in quasi-one-dimensional system, one of collective charge behaviors below Peierls transition temperature (T_p) under an applied electric field E exceeding the classical threshold E_{T-Cl} , has long been attributed to the sliding of a charge-density-wave (CDW)^{1,2}. Scientists often compare such phenomenon to the Fröhlich 'sliding-mode' conductivity, an early attempt to explain superconductivity in one dimensional system³⁻⁵. However, in a real CDW compound, most of the experimental results showed that the dc conductivity from CDW transport was not corresponding to the Fröhlich mode, the reason may come from the notion of pinning by impurities, the damping of uncondensed carriers screening and moreover, phason-phason or phason-phonon scattering^{5,6}. Littlewood proposed that at sufficiently low temperatures, the I - V characteristic develops an S-shaped bistability, in which the high-velocity branch describes a 'Fröhlich superconductor' in zero temperature limit. Up to date, only a few work has reported the evidences for the onset of Fröhlich-mode conductivity at liquid Helium temperature when $E > 1000\ E_{T-Cl}$ ³. Naively, one would wonder whether similar behavior could be observed in other CDW systems.

For the S-shaped I - V relation proposed by Littlewood, it has been speculated that the upper branch represents a rigid motion of the CDW in the manner of the original Fröhlich model, whereas the lower branch indicates a lower speed motion of the CDW. In experiments, measuring the S-shaped I - V curve is difficult since the transition from the lower branch to the higher one means from current driving to potential driving⁵. Further the phenomenon of meta-stable state at very large time scale makes the measurements and analysis much complex^{2,7}.

However, we noted that such difficulty may be due to the dc case, if the ac dynamics could be obtained, then we can extrapolate to zero-frequency limit and meanwhile, the behavior of rigid CDW condensates at finite frequencies could also be uncovered.

Temperature dependent complex dielectric or conductivity spectrum is a powerful method for studying charge dynamics of substances⁸⁻¹⁴. At a fixed temperature and meanwhile especially under various E established by dc bias, it would be of great interest since the depinned CDW could transport charges collectively or even possibly provide Fröhlich-mode conductivity. To the best of our knowledge, the temperature and bias dependent dielectric or conductivity spectra have been reported for K_{0.3}MoO₃^{1,15,16} and NbSe₃^{2,17,18}. For TaS₃, similar research work is relatively insufficient^{1,19,20}, the very recent report is the ac-dc interference spectra investigated by Matsuura *et al.*¹². Hence, investigating the charge dynamics of TaS₃ system under various electric field would be helpful to elucidate the behavior of the charged condensates.

In this report we studied the temperature and electric field dependent charge dynamics of *o*-TaS₃ crystals along the chain direction. Experimental results reproduced well in other specimens. We found that the sample with $10\ \mu\text{m}^2$ in cross section reveals two electric thresholds, the E_{T-Cl} and a novel threshold E_{T-N} at a larger value. With decreasing temperature, E_{T-N} increased monotonically. We observed S-shaped I - V relation under $E \gtrsim E_{T-N}$ at which the differential conductivity increases till a divergence. Further, in the conductivity spectra of the *o*-TaS₃ crystals under a static electric field $E > E_{T-N}$, a sharp dispersion revealed below kHz. At a fixed temperature, the scattering rate was extremely small and decreased with increasing field.

FIG. 1: (Color online) (a) The connecting structural diagram of the *o*-TaS₃ specimen for measuring the transport properties. Note that s1-screw, s2-steel plate, s3-spring, s4-sample. The two ellipses present Indium contacting. (b) The picture is for the sample *o*-TaS₃ probed in the experiments. The inset shows a gold silk with diameter 25 μm .

With decreasing temperature, the scattering decreases and the Fröhlich conductivity would expect to occur in zero temperature limit. We proposed that the Fröhlich-mode would be consistent with the meta-stable state, due to the sharp dispersion below kHz.

II. EXPERIMENTS

o-TaS₃ crystals were successfully grown by the conventional chemical vapor transport method²¹. We carefully selected separated fibrous crystals with typical length 4.0 mm. The cross section area of the sample was about 10 μm^2 , obtained by comparison with the resistivity at 300 K. The photograph of the specimen and the connecting diagram have been shown in Fig. 1.

Temperature dependent resistivity was measured with current $I=10 \mu\text{A}$ before probing the conductivity spectrum, similar to the method in previous reports²¹. We also compared the contact resistance of the 2-probe method with 4-probe configuration and found it was negligible within the experimental error 0.5%. The obtained transport properties were nearly the same for different substrates of LaAlO₃ and sapphire. The complex impedance properties under sweeping dc bias

$z(\omega, E) = |z(\omega, E)|\cos\theta(\omega, E) + i|z(\omega, E)|\sin\theta(\omega, E)$ from 0 to 20 V/cm were probed on Agilent 4294A Precision Impedance Analyzer through four-coaxial cables (into two pairs, corresponding to two electrodes on the sample), where $\theta(\omega, E)$ was the phase difference between the ac potential and current at frequency ω . The sample was mounted on a cryostat controlled by Cryo-con 32 with the experimental temperature range 50-300 K. All the impedance spectra were measured with driving voltage $V_{p-p}=20 \text{ mV(rms)}$, far below the electric threshold field obtained in *I-V* measurements. The complex conductivity spectrum $\sigma(\omega, E) = \sigma_1(\omega, E) + i\sigma_2(\omega, E)$ could be deduced from the raw data $z(\omega, E)$ based on the universal relations: $\sigma(\omega, E) = 1/z(\omega, E) \cdot (d/S)$, where d and S are the length and cross-section area of the specimen.

III. RESULTS AND DISCUSSIONS

The temperature dependent resistivity along the chain direction of *o*-TaS₃ specimens is consistent with previous results²¹⁻²⁴. Below Peierls transition temperature $T_p = 220 \text{ K}$, two electric thresholds reveal on the *I-V* curves, see Fig. 2(a): the classical electric threshold E_{T-CI} and a novel threshold E_{T-N} at a larger field strength. In contrast, the previous experimental data, only one threshold E_{T-CI} appeared above 70 K^{19,21,24-26}.

In our experiments, the $E_{T-N} \sim 2E_{T-CI}$ at 180 K, where $E_{T-CI}=2.0 \text{ V/cm}$ is larger than the data previously reported due to the enhancing of the restoring forces contributed by a large ratio of surface-to-bulk^{2,27}. Here we emphasize that the emerging of the classical non-linearity in *I-V* relation could be attributed to the small separated CDW segments or domains, which start to transport charges collectively for $E > E_{T-CI}$ ^{26,28}. Though these small domains are depinned, they are expected to be non-coherent with each other, see Fig. 2(b)-III, where the case I and II are for $0 \leq E < E_{T-CI}$, the corresponding phase of the CDW deforms slightly to response the applied electric field. For the appearance of E_{T-N} , the finite size effects should be considered: generally the smaller diameter means the larger of the perpendicular dynamical coherence of the CDW phases^{2,29}. If the coherence among the small CDW domains eventually establishes, the whole CDW would act as an one rigid throughout coherent 'domain', as shown in Fig. 2 (b)-IV, but for thicker samples case would be changed inversely and thus no E_{T-N} features. As expected, typical volume of the throughout coherent CDW domain (TCCD) would be up to a macroscopic volume^{13,23}. With decreasing temperature, the absolute value of E_{T-N} increases monotonically.

In Fig. 2(c), the details near E_{T-N} has been shown. Clearly, the S-shaped *I-V* relation tends to more remarkable with decreasing temperature. It can be expected that as E increases up to E_{T-N} , a divergent differential conductivity dI/dV emerges. Littlewood suggested that the upper branch represents a rigid motion of the CDW in

FIG. 2: (Color online) (a) The I - V relation of o -TaS₃ crystal at 180 K and 140 K. The vertical bar shows E_{T-CI} , whereas the arrow shows E_{T-N} . The dashed lines are drawn to the naked eyes. (b) The diagram for the charge transport of CDW domains. Note that I: $E=0$; II: $0 < E < E_{T-CI}$; III: $E_{T-CI} < E < E_{T-N}$; IV: $E > E_{T-N}$. The triangles present the impurities by which the CDW is pinned down when $E < E_{T-CI}$. (c) The enlarged I - V curve near the E_{T-N} region of o -TaS₃ crystal at 180 K, 160 K and 140 K. The S-shaped feature could be identified well.

the manner of the original Fröhlich model⁵. For the measurement of the S-shaped I - V curve, as the sample with small cross section area could not endure large current, it is difficult to finish the transition from the lower branch to the higher one in a wide range. However, we noted that such difficulty may be due to the case of dc current, for the charge dynamics in kHz region, see Fig. 3, the

FIG. 3: (Color online) The bias dependent conductivity spectrum of o -TaS₃ crystal at various frequencies and temperatures. Note the jumps indicated by the ellipses at different frequencies occur at nearly the same field.

probing of transport properties could be extended to a much higher applied electric field. Hence it is possible for us to uncover the behavior of rigid CDW condensates at finite frequencies.

As shown in Fig. 3, above $E_{T-N}=7.0$ V/cm at 140 K, the specimen still remains well, not broken down. With increasing frequency, the responses reveal a slight but identifiable dispersion in low frequency range. As the features of 1.0, 2.0, 5.0 and 10.0 kHz occur nearly at the same dc bias in each case as shown by the ellipses, the contribution of narrow band noise (NBN)^{26,30,31} could be excluded. Thus it is plausible to attribute the low-frequency dispersion in conductivity to other mechanisms. The dispersion in conductivity spectra under external field $E > E_{T-N}$ have been clearly shown in Fig. 4. Astonishingly, the conductivity below 1.0 kHz could not be constantly extrapolated to dc limit. Comparing with the results from Hundley *et al.*¹⁶ on K_{0.3}MoO₃, our experimental data again confirms no NBN features. As E increases, the dispersion or inductive-like response becomes more remarkable, and meanwhile shifts towards lower frequency.

In previous report²⁰, Bhattacharya *et al.* reported another inductance feature in the real conductivity and a concomitant occurrence of negative imaginary part of conductivity near the washboard frequency at 120 K. However, our experimental data could not be explained by classical deformable CDW model as suggested by Coppersmith and Littlewood³². The conductivity spectra in Ref.²⁰ and Ref.¹⁹ show downturn curvatures near MHz, whereas our data reveal an upturn below kHz when $E > E_{T-N}$. Here one may also speculate that the dispersion below kHz may be contributed by the broad band noise (BBN)²⁶. We propose that this could also be excluded for the following reasons: (1) It expects that BBN could not have such a large amplitude as shown in Fig. 4 for $E > E_{T-N}$; (2) In Ref.²⁶, under a electric field that BBN could appear (in the range 1.1-1.8 E_{T-CI}), there is

FIG. 4: (Color online) The bias dependent conductivity spectrum of *o*-TaS₃ crystal at 150 K (a) and 120 K (b). Note the fittings [thin curves obtained from Eq. (2)] present good results when $E > E_{T-N}$ at corresponding temperatures.

no jump-like feature in I - V curve, and thus they are independent with each other; (3) As shown in 4(b), at $E=2.5$ V/cm (about $2.2 E_{T-Cl}$) where BBN should be present, however, there are no remarkable features in lower frequency region of $\sigma_1(\omega)$.

The present experimental results are not due to heating effect proposed by Mihaly *et al.*⁷. The reason is that, when we applied a stable dc bias field, not electric pulses, the thermal equilibrium could be obtained and thus the system should be in a stable conducting state. In their experiments⁷, the sample was heated by a large current pulse, however, the ac dynamics were not probed. Our experimental data does show a rather extremely dispersion below kHz, then the present experimental data must be a intrinsic response of the system, neither it is noise nor heating effect.

The novel dispersion below kHz in $\sigma_1(\omega)$ maybe the origin of the spontaneous decay in *o*-TaS₃ crystal as reported in Ref.⁷. As we have known well for a ideal resistor, we can always observe a stable response since the system has a constant $\sigma_1(\omega)$. For our present experiment, when added a step voltage on the specimen, at the very beginning of the switch (corresponding to a much

FIG. 5: (Color online) The picture is for the response of the specimen to the applied periodic electric pulses. In experiments, the specimen connects with a resistor 1.0 k Ω in series. We took this photo from an oscilloscope.

FIG. 6: (Color online) (a) The bias dependent fitting parameters at various temperatures of *o*-TaS₃ crystal with 10 μm^2 in cross section. (b) The temperature dependent electric thresholds of *o*-TaS₃ crystal. Note that with decreasing temperature, E_{T-N} increases monotonically.

shorter time scale), we see the response at very high frequencies — the lower conductivity region; whereas as time elapses, the responses at lower frequencies would gradually dominate the properties, that is the higher conductivity. As $\sigma_1(\omega)$ disperses very sharply towards zero frequency limit, the response of the system would

be different at various time scales, driving spontaneous decay or meta-stable states^{7,33,34}, as also observed in our experiments, see Fig. 5. Considering that in Mihaly's experiments⁴, the spontaneous decay could even be observed till 10^5 s, indicating that the dispersion could be maintained up to 10^{-5} Hz at least.

For the dispersion in lower frequency region, the origination must be related to the rigid coherent CDW motion. Qualitatively, we find that the conductivity decreases with increasing frequency. It is expected to fit the conductivity spectra by two components: $\sigma(\omega) = \sigma_{UC} + \sigma_{CDW}(\omega)$, where the constant σ_{UC} is for the conductivity of uncondensed carriers while $\sigma_{CDW}(\omega)$ presents the contribution from TCCD. To the best of knowledge, the theory for field dependent ac conductivity of TCCD is relatively insufficient. In Ref.²⁶, Maeda *et al.* suggested that in the high-field region, the CDW condensate would be drifting and could be described by the 'classical particle' oscillator model. We postulate that the movement of TCCD has similar behavior^{2,35},

$$\sigma_{CDW}(\omega) = \frac{n_c e^2}{i\omega m^*} \frac{\omega^2}{\omega_0^2 - \omega^2 - i\omega/\tau} \quad (1)$$

Here m^* is the effective mass; τ^{-1} is the scattering rate of the TCCD, which could be attributed to the scattering from impurities, phasons and phonons at non-zero temperatures. For $E > E_{T-N}$, it is plausible that the pinning frequency (corresponding to the restoring force) $\omega_0 \rightarrow 0$, and then from Eq. (1) the real part of $\sigma(\omega)$ is

$$\sigma_1(\omega) = \sigma_{UC} + \frac{n_c e^2}{m^* \tau} \frac{1}{\omega^2 + 1/\tau^2} = \sigma_{UC} + \frac{\sigma_{dc}}{1 + \omega^2 \tau^2} \quad (2)$$

where $\sigma_{dc} = n_c e^2 \tau / m^*$ is the dc conductivity of the TCCD. As proposed by Sridhar *et al.*³⁶, Eq. (2) could also be obtained from an equation of motion $d^2x/dt^2 + \tau^{-1}(dx/dt) = (-e/m^*)E(t)$ which describes the damped inertial response of the CDW condensate (by ignoring pinning).

The fitting results have been shown in Fig. 6 (a). At a fixed temperature, σ_{dc} increases whereas the scattering rate decreases with increasing field. Comparing

with the experimental data from Ref.³⁶, we have noted that the present scattering rate is extremely small, which maybe due to the establishment of the TCCD in macroscopic scale when $E > E_{T-N}$, as shown in Fig. 3 (b)-IV. In other words, the motion of the TCCD for current carrying would be hard to change comparing with the case of non-coherence. In zero temperature limit, we can speculate that the damping of the system would diminish and thus the Fröhlich conductivity establishes. Nevertheless, the existence or establishment of the TCCD in *o*-TaS₃ should be further supported by other methods.

In Fig. 6(b), we show the phase diagram of temperature dependent electric thresholds for *o*-TaS₃ crystals in our experiments. The Fröhlich conductivity would be consistent with the meta-stable states: in zero temperature limit, the ideal zero-resistivity for dc current maybe satisfied, and meanwhile due to the sharp dispersion below kHz, the meta-stable state or the spontaneous decay could also be satisfied.

IV. SUMMARY

In summary, we measured the temperature and electric field dependent conductivity spectra of *o*-TaS₃ sample with $10 \mu\text{m}^2$ in cross section. We observed the classical electric threshold E_{T-Cl} and a novel threshold E_{T-N} at a larger electric field due to the establishment of coherence among small CDW domains. Near E_{T-N} , an S-shaped *I-V* relation revealed. Under a stable field $E > E_{T-N}$, a sharp dispersion revealed below kHz. At a fixed temperature, the scattering rate of the specimen was extremely small and decreased with increasing field. In zero temperature limit, the scattering would diminish and thus the Fröhlich conductivity would occur, considering the sharp dispersion in low frequency region, the Fröhlich conductivity would be consistent with the meta-stable states.

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