Possible origin of nonlinear conductivity and large dielectric constant in the commensurate charge-density-wave phase of $1T\text{-}TaS_{2}$

Yongchang Ma, Yanhui Hou, Cuimin Lu, Lijun Li, and Cedomir Petrovic

Phys. Rev. B **97**, 195117 — Published 10 May 2018

DOI: 10.1103/PhysRevB.97.195117

The possible origin of non-linear conductivity and large dielectric constant in the commensurate CDW phase of 1T-TaS_2

Yongchang Ma[1,2,3,*] Yanhui Hou[1], Cuimin Lu[1,4], Lijun Li[2,†] and Cedomir Petrovic[2]

[1] *School of Materials Science and Engineering, Tianjin University of Technology, Tianjin 300384, China;*

[2] *Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973, USA;*

[3] *Key Laboratory of Display Materials and Photoelectric Devices (Tianjin University of Technology), Ministry of Education, Tianjin 300384, China.*

[4] *Tianjin Key Lab for Photoelectric Materials and Devices, Tianjin 300384, China;*

(Dated: April 18, 2018)

Abstract

The electric field dependence of the dielectric properties and the non-linear conductance of 1T-TaS_2 below 50 K has been investigated. A large dielectric constant of about 10^4 is obtained up to 10^7 Hz, which can not be attributed to hopping of the localized carriers alone, the collective excitations of the commensurate charge-density-wave must be another contributor. The dielectric spectra disperse slightly in our measured temperature and frequency range. At a moderate dc bias field, the real part of dielectric constant $\epsilon_1(\omega)$ decreases. We propose that the separation of bound soliton-antisoliton pairs may be a contributor to the reduction of $\epsilon_1(\omega)$ and the accompanying non-linear conductivity with increasing dc bias.

*Email: ycma@tjut.edu.cn

†Permanent address: Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, Peoples Republic of China

I. INTRODUCTION

Non-linear conductivity and large polarization effects are the well-known phenomena in charge density wave (CDW) systems [1–3]. Though researchers made considerable progress in understanding the associated transport properties in quasi-one dimensional CDW compounds [4–12], less discussions for two-dimensional (2D) CDW crystals were carried out, especially the polarization properties [13–17]. In recent years, the investigations of 2D-CDW compounds have revitalized, for they would be helpful to understand the mechanisms of the high-T_c superconductors [18–20] and the origin of ultra-fast switching to a stable hidden quantum state [21, 22]. Hence, it is of interest to investigate the charge transport behavior in 2D-CDW systems.

The 2D-CDW often reveals itself in layered transition-metal dichalcogenides and rare-earth tritellurides [13, 14, 23–26]. Researchers began to study the charge transport properties of 2D-CDW systems (1T-TaS_2) about forty years ago: Refs. [14, 27, 28] suggested that the carriers localization induced by disorder may be responsible for the unique low temperature transport properties; whereas Uchida *et al.* proposed that the non-linear conduction in the presence of a moderate electric field at low temperatures might be explained by the collective excitations in pinned CDW — the creation of a soliton in pair with an antisoliton [16]. In the followed experiments, though the electronic nature was probed by various powerful spectroscopies [29–33], until recent years the mechanisms for the charge dynamics of the commensurate CDW (CCDW) phase in 1T-TaS_2 has not been disclosed yet. The discrepancy in understanding the transport properties at low temperatures has prompted us to study the fundamental question: whether the collective excitations of CCDW exist or not in 1T-TaS_2. As proposed by Rajaraman [34], for a system described by the equation of motion with soliton solutions, the soliton-antisoliton ′doublet′ also exists, in which the equally but oppositely charged solitons are in confinement and lead to a large static dielectric constant [35]. Considering that the localized electrons have a distinct feature on the dispersion of the dielectric spectrum, $\epsilon_{r1}(\omega) \propto \omega^s$ (s<0) [36, 37], investigations of the dielectric properties 1T-TaS_2 would be helpful to demonstrate whether the ′doublets′ have an essential contribution. On the other hand, by applying a dc electric field, the bound charges in the soliton-antisoliton ′doublets′ may obtain a larger probability to decouple into carriers with long mean-free-path, and accordingly driving a non-linear conductivity superposing on the Ohmic behavior.

Temperature and field dependent complex dielectric or conductivity spectrum is a powerful probe for investigating the charge transport and polarization behavior of various systems [9, 10, 38–41]. At a fixed temperature T_0, the complex dielectric or conductivity spectrum is ultimately defined by the polarization or conductance of the total contributing charged microscopic particles. In quasi-1D CDW systems, exceptionally large dielectric constants with strong dispersion and novel conducting behavior in radio and audio frequencies were observed [1, 10, 42]. Further, the dc biased charge dynamics would be of great interest, since non-classical characteristics often reveal, e.g. the well known current oscillations and mode-locking [1]. Clearly, these novel properties should not be attributed to normal carriers but to the new-born current carrying mechanisms associated with CDWs by applying electric field. For 1T-TaS_2, the non-Ohmic behavior manifests in current voltage characteristic (CVC), naturally, it is expected to be relevant to individual particles or collective excitations of CDW condensates [16, 17]. To the best of our knowledge, the discussions on dielectric or ac conductance behavior in 2D-CDW systems are limited, possibly due to strong screening effects from normal carriers thermally excited over the non-uniform CDW gap opening at the Fermi level [29, 31]. Usually, normal carriers respond extremely fast ($< 10^{-12}$ s), and if their density is large enough, the features of the conductivity in radio and audio frequencies relevant to the excitations of CDW condensates will become too weak to be identified well. However, below $T = 50$ K, the number of normal electrons excited by thermal energy remarkably decreases [14, 21], providing good opportunities to clarify the mechanisms of carriers motion in CCDW phase of 1T-TaS_2.

In this report we systematically study the polarization properties of 1T-TaS_2 flakes below 50 K. To clarify the origin of the non-linear conductance in CVC, the dielectric properties under various dc bias E_B are also probed. The Joule heating effects on ac conductivity under constant bias field $E_B \leq 20.0$ V/cm are analyzed by comparing with the pulsed approach. We propose that the large relative dielectric constant ($\epsilon_r \sim 10^4$) up to frequency 10^7 Hz can not originate mainly from hopping process of the localized states, but is possibly driven by the existence of 'bound soliton-antisoliton pairs' (BSPs). Besides the hopping of localized carriers, the separation of BSPs may also be the driving force for the reduction of $\epsilon_1(\omega)$ and the non-linear conductivity with increasing of dc bias.

II. EXPERIMENTS

The 1T-TaS_2 single crystals were successfully grown by the conventional chemical vapor transport method in a gradient furnace. The stoichiometric tantalum and sulfur pieces were put into an alumina crucible before sealing in a quartz tube and pre-reacted at 900 °C for 50 hours and then cooled to 760 °C within 12 hours followed by natural cooling. For single crystals growth, the temperature of hot (cold) end was 900 °C (800 °C). After one week growth time, the quartz tube was quickly pulled out and then quenched into cold water. Consistent with the earlier reports [24], the as-grown single crystals show thin flake-like shapes and gold-shining color, with typical sizes are in the range $(3 \sim 10) \times (2 \sim 3) \times (0.02 \sim 0.08)$ mm^3. Ref. [28] shows $\rho_\perp/\rho_\parallel \sim 500$ over the range 1.3<T<240 K, where $\rho_\parallel$ and $\rho_\perp$ are the resistivity perpendicular and parallel to the layers of 1T-TaS_2 crystal respectively, indicating an essential 2D system. In the experiments, we focused on the in-plane polarization and transport properties of the sample with surface area 3.0×0.4 mm^2 and thickness (along c-axis) about 25 μm, and the distance between two potential contacts is about 1.0 mm.

Temperature dependent resistivity was measured with current I=10 μA before probing the dielectric spectra. The CVCs showed linear behavior in weak electric field, confirming good ohmic contacts by silver paste. We also compared the contact resistance of the 4-contact with 2-contact probe and found the difference was negligible compared to the sample resistance from 300 K to 12 K. The Hall coefficient were probed under a magnetic field H=10.0 kGs parallel to the c-axis of the sample. In addition, we confirmed that the impedance spectra were unaffected by the difference between substrates by comparing data obtained with sapphire and the ultra thin mica sheets. The complex impedance properties under various dc bias up to 40 V/cm $\hat{z}(\omega) = |\hat{z}(\omega)| cos\theta(\omega) + i|\hat{z}(\omega)| sin\theta(\omega)$ were probed on Agilent 4294A Precision Impedance Analyzer through four-coaxial cables in two pairs (corresponding to two electrodes on the sample), where $\theta(\omega)$ was the phase difference between the ac driving potential and the response current at frequency ω. The sample was mounted on a cryostat of a closed cycle refrigerator (JANIS) with the experimental temperature range from 12 to 300 K monitored by a Cryo·con 32 temperature controller. All the impedance spectra were measured with driving voltage 30 mV(rms), where the non-linear conductance does not appear in I-V measurements. In analysis, the sample was seen as a leakage capacitor, both dielectric constant $\hat{\epsilon}(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega)$ and the complex conductivity spectrum $\hat{\sigma}(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ could be deduced from the raw data $\hat{z}(\omega)$ based on the universal relations: $\hat{\epsilon}(\omega) = \frac{\hat{\sigma}(\omega)-\sigma_{dc}}{i\omega}$ and $\hat{\sigma}(\omega) = 1/\hat{z}(\omega)$, thus $\epsilon_1(\omega) = \frac{-sin\theta(\omega)}{\omega|\hat{z}(\omega)|} \cdot \frac{d}{S}$, where $\epsilon_i(\omega) = \epsilon_0\epsilon_{ri}(\omega)$ (i=1,2), d and S were the length and the cross-sectional area of the sample, respectively.

III. RESULTS AND DISCUSSIONS

The temperature dependent resistivity $\rho_{dc}(T)$, the CCDW transition temperature T_{CCDW}=180 K, and the hysteresis feature upon warming are all consistent with previous reports [17, 21, 23], indicating good quality of the 1T-TaS_2 single crystal samples. The existence of non-linear conductivity in the CCDW phase was confirmed, as shown in Fig. 1. In our experiments, the electric field is limited within a moderate range, no more than 20 V/cm. The previous data have already shown that the carriers in the CCDW phase of 1T-TaS_2 are positively charged and the density is larger than 10^{19}/cm^3, hence the interface or the contact effects induced by Schottky-barrier could be neglected. In the following of context, the experimental data were obtained on decreasing temperature in the CCDW phase. We will focus on the polarization behavior of 1T-TaS_2 in the presence of the non-linear conductance below T=50 K.

To obtain the intrinsic CVCs, a narrow-width pulsed-current method is often required [4, 16, 17]. However, to investigate the polarization properties of the 1T-TaS_2 single crystals in the presence of the non-linear conductivity, a continuous dc bias would inevitably heat the sample (the Joule heating effects). The only question is how much does it change the temperature of the sample. Considering that the non-linear conductance is present in a moderate field, we compared the difference between the pulsed approach with pulse duration 0.1 ms and the continous method below T=50 K, see Fig. 1. At different temperatures, the data are similar except the initial slopes of the CVCs. Below E=5.0 V/cm at a fixed temperature, the CVCs differs only slightly, indicating a trivial Joule heating; whereas above E=8.0 V/cm, a deviation starts to appear. Generally, it is shown that self-heating has strong effects on dc transport properties in all systems with negative temperature coefficient of resistance [43, 44].

A. The self heating effects to conductivity spectrum under dc bias

The self-heating in external dc field and its influence on transport properties is important. To clarify this effect, the CVC at T=30 K in a wide electric field range was obtained, see Fig. 2 (a). For the current in continuous mode, an extra conductivity is superimposed on the pulsed results, indicating the effect of local Joule heating (the self-heating effect), not the intrinsic properties of 1T-TaS_2 system.

For the conductivity and dielectric spectra measured at constant dc bias using small ac voltage with changing ω: at low frequency, $\sigma_1(\omega \ll 1/\tau_{SH})$ is proportional to the differential conductivity (dI/dV), where τ_{SH} is the relaxation time from self-heating; at high frequency, $\sigma_1(\omega \gg 1/\tau_{SH})$ is proportional to the chordal conductivity (I/V). τ_{SH} is related to the specific heat by the expression [$\tau_{SH} = \frac{N_M c}{\kappa} \cdot \frac{d}{S}$], where N_M is the number of moles of material with specific heat c and thermal conductance κ [45]. In our experiments, $N_M \simeq 2.8\times10^{-7}$ mol, c=5.0 J/(mol·K) at 30 K [46], τ_{SH} is estimated to be ∼ms for heat flow parallel to ab-plane, larger than the perpendicular direction [47–49].

Nevertheless, above 10 kHz the thermal relaxation effect under E_B nearly disappear (Fig.2 b), being consistent with the measurement in pulsed approach with duration 0.1 ms. However, increase of frequency does not remove the local Joule heating. Specifically, the total conductivity: $\sigma_T = \sigma_{NM} + \sigma_{NL} + \sigma_{SH}$, where the index NM, NL and SH represent the contribution of normal carriers, the non-linear conductance and the self-heating effect. These three components behave differently with frequency increasing: σ_{NM} should reveal the well-known Drude free-electron properties and has no dispersion below GHz; σ_{NL} remains unresolved at present; whereas for σ_{SH}, as the averaged temperature of the sample elevates under dc bias, thus an increase of the frequency-independent conductivity is expected, since 1T-TaS_2 is a system with negative temperature coefficient of resistance.

In order to study the charged dynamics in the presence of the non-linear conductance in 1T-TaS_2, the evaluation of the temperature elevation under dc bias is necessary. We compared $\sigma_1(\omega)$ in Fig. 2(b) at 10 kHz with the CVC results measured in pulsed approach, and found that the enhancement of the averaged temperature at bias 10 V/cm is no more than 0.5 K in our experimental configuration. Above several MHz, $\sigma_1(\omega, E_B = 0)$ increases with increasing frequency. Clearly, this behavior should not originate from normal carriers or thermal effect, but be associated with a large dielectric constant.

B. The large dielectric constant in MHz region

In 1D-CDW systems, the exceptionally large dielectric constants and strong dispersion in audio frequency range have been extensively investigated [9, 10, 42, 50]. However, the investigations of dielectric behavior are rarely reported for 2D-CDW compounds, possibly due to the large screening effects from the normal carriers excited over the non-uniform CDW gap opening at the Fermi surface. Nevertheless, for the polarization properties of the 2D-CDW system 1T-TaS_2, a large static dielectric constant is expected for its highly polarizable structure, as suggested by Vaskivskyi *et al.* [17]. In our experiments, relatively small minus $\theta(\omega)$ could be identified below $T=$ 100 K, revealing capacitive responses, as shown in Fig. 3 (a). For dielectric spectra $\epsilon_{r1}(\omega)$, there are large noise below 2.0 MHz, whereas distinct decrease could be identified above 10^7 Hz, see Fig. 3 (b). At present, we can not obtain complete relaxation behavior of the corresponding polarization process for the limited measuring frequency range. With decreasing temperature below 50 K, the dielectric spectra are suppressed whereas the line-shapes remain unchanged.

The large dielectric constant ϵ_{r1} of the order of 10^4 in MHz region is of interest. In the incommensurate CDW system, extremely large polarization effects are expected due to weak pinning from impurities or defects [51]. For the CCDW state in 1T-TaS_2, unlike the incommensurate 2D-system $TbTe_3$ where Sinchenko *et al.* observed CDW sliding [25], for it would be difficult to slide in a moderate electric field due to the strong pinning by the underlying lattice and hence the contribution to the static dielectric constant would be much smaller [51]. Here one may argue that the mechanism in establishing the colossal dielectric constant in $(NbSe_4)_3I$ (due to the slight shift of the single chains against each other) can be a potential substitute for 1T-TaS_2, however, such similar features of the lattice structure has not been identified yet [10].

As discussed by Lunkenheimer *et al.* [36], hopping process of localized states can also give rise to large values of the dielectric constant. In Ref. [28] Hambourger and Di Salvo suggested that the disorder induced localization may explain the transport properties in 1T-$TaS_{2-x}Se_x$ system, as supported by Dardel *et al.* [29] and Kim *et al.* [30]. Indeed, our data of the temperature dependent Hall coefficient (Fig. 4) is consistent with Ref. [28], indicating the occurrence of carriers localization in the single band approximation. However, we note that the dielectric spectrum can not be fitted well by the hopping process alone [36, 37, 52]:

$\epsilon_{r1}(\omega) \propto \omega^s$ ($s < 0$). In Fig. 3(b), the dotted-curve fittings with s=-0.08 show a slightly upward curvature, which is inconsistent with the experimental data exhibiting a downward curvature above 10 MHz. Clearly, the cases at other temperatures behave similarly. Hence it seems that a relaxation process must exist besides the hopping of localized electrons based on the dielectric properties. On the other hand, the interfacial effects such as, e.g., a grain boundary or a surface depletion layer also contribute to colossal dielectric constant. However, the positively charged carriers with density larger than $10^{19}/\mathrm{cm}^3$ in the CCDW phase exclude the Schottky-barrier effects. As for the polaron contribution to the dielectric constant, we note that in the infrared conductivity spectrum, no such characteristic could be identified without photo-excitations [53].

We propose that the polarization properties of 1T-TaS_2 could be described by the well-known soliton model used in 1D-CDW. To the best of our knowledge, the complete theoretical model for the phase solitons in 2D-CDW materials is not available at the moment. The existence of CDW phase solitons — topological phase defects — is a particular form of collective excitation in the CDW condensate and has been confirmed in quasi-1D systems [54–57]. Recently, there are suggestions that soliton contribution must be rather important for the unconventional transport properties associated with CDW system [58–60]. Rojo-Bravo *et al.* indicated that the transport mechanism of travelling soliton lattice may open new perspectives in controlling correlated charges over long distances, explaining the main features of sliding CDW systems [61].

In Ref. [35], Krive and Rozhavsky proposed that the phase solitons significantly contribute the dielectric susceptibility. As shown in Fig. 5 (a), the localized compressions or rarefaction in the local condensed electron density (or deformations of the CDW phase φ) correspond to the soliton-antisoliton pair. A sketch of the charge density and the lattice arrangement is shown in Fig. 5 (b) [36, 62]. It is plausible that the central Ta atoms attract more electronic cloud which favors the commensurate CDW. In the equation of motion relevant to CDW phase, $d^2\varphi/dt^2 - c_0^2\nabla_x^2\varphi + \omega_F^2 dV(\varphi)/d\varphi = 0$, where c_0 is a characteristic velocity of the undeformed condensate and ω_F is the pinning frequency, the realistic pinning potential of the phase $V(\varphi)$ has a periodic form as expected [56, 57]. The exact solutions of the equation should consist of several forms characteristic of solitons, including the soliton-antisoliton 'doublet' [34] whose excitation energy is relatively lower due to the existence of coupling, compared with the case of the discreet soliton and antisoltion, see Fig. 5 (c). Instead of separating into the discrete soliton or antisoliton which are infinitely far apart with each other in infinite time, the relative separation for the members of the doublets oscillate periodically [34]. This doublet solution is also a ′breathing′ one and can be thought of as a bound soliton-antisoliton pair (BSP).

As the equally but inversely charged phase excitations are confined in BSPs, they would contribute to the dielectric constant of the system. It can be estimated using simple expression for dipole oscilliator [35]: $\epsilon_\varphi \sim \epsilon_\Delta(E_s/\omega_0)\ln(E_s/\omega_0)$, where $E_s/\omega_0 = \Delta/M\omega_Q$, Δ is the energy gap in the electronic spectrum, M is the commensurability index and ω_Q is bare frequency of phonon with momentum Q=2k$_F$ (Fermi wave vector). As suggested by Krive and Rozhavsky [35], $\epsilon_\varphi \sim 10^{1\sim2}\epsilon_\Delta$, where ϵ_Δ is about 10^3, hence a dielectric constant $\sim 10^4$ would be reasonable. In Ref. [16], Uchida *et al.* considered the collective excitation of a soliton in pair with an ′antisoliton′ under a moderate field, to explain the field independent Hall coefficient at T=4.2 K. However, solitons in CDW systems are always excited in pairs, whenever a soliton (φ compression) is created, an antisoliton (φ rarefaction) appears. Though the explanation of Uchida *et al.* demonstrates that mobile solitons and antisolitons are excited by applying electric field, they can not identify whether the excitation is Ground state→II or BSP→II, where II presents the state of free soliton and antisoltion [Fig. 5 (c)]. Ref.[16] and the followed research work associated with the dc transport did not provide the dielectric properties of the CCDW state of 1T-TaS$_2$. As bound charged microscopic states contribute to large electric polarizations, the BSPs are expected to exist.

Though the polarization properties of the BSPs are far from being understood, a Debye relaxation or Cole-Cole empirical equation may be plausible. In Fig. 3(b), good fittings to the dielectric spectra were provided by the sum of the hopping process and the Debye relaxation model, $\epsilon_0\epsilon_{r1} = \epsilon_{HF} + (\epsilon_s - \epsilon_{HF})/[1 + (i\omega\tau_0)] + A_{hp}\omega^s$, where τ_0 is a characteristic relaxation time, while ϵ_s and ϵ_{HF} are the dielectric constant in static and at higher frequency

TABLE I: The resultant parameters for fitting the dielectric spectra of 1T-TaS$_2$ at T=15 K and 25 K. ϵ_{HF} is not a dominant parameter and has been determined empirically.

T(K)	$\epsilon_s(10^3\epsilon_0)$	$\epsilon_{HF}(10^3\epsilon_0)$	$\tau_0(10^{-8}sec.)$	$A_{hp}(10^3\epsilon_0)$
15	10.6	1.0	2.7	40
25	12.2	1.0	2.2	56

limit of the Debye relaxation model, respectively; A_{hp} is the coefficient of the hopping term and s is assumed to be -0.2 [37, 52]. The resultant parameters are shown in Table I. It is remarkable that the feature above 10 MHz is associated with the Debye relaxation, whereas the hopping process would be responsible for the slight dispersion in low frequency region.

Other promising explanation for the observation of large dielectric constant is connected with the spontaneously arising interfaces. These are induced by the electronic phase separation or charge order as commonly observed in many transition-metal oxides. Though the true reason for the colossal polarization has not been completely clarified yet, it is possible that at least in some cases internal interfaces between the different phase-separated regions may play a role [36]. It is suggested that the dielectric behavior of these spontaneously forming heterogeneous regions represents a type of the long-known Maxwell-Wagner polarization effects, with the heterogeneity arising on a much finer scale close to crystal unit cell. Here we suggest that this interpretation is associated to the following two aspects: (1) the density of charges arranges periodically in real space [see Fig. 5(b)], however, whether such CCDW state of 1T-TaS_2 can be considered as a particular form of heterogeneity and drive large polarizations is not clear at present; (2) the domain wall configurations are on a scale of unit cell and accordingly it would remarkably contribute to the dielectric properties of the system [17, 33], therefore further investigations are necessary to clarify this issue.

C. The suppression of dielectric constant by dc bias

The dielectric behavior we discussed here is attributed to BSPs which could be locally polarized, rather than the free solitons that can transfer in long distances. In a weak applied dc electric field E, the bound soliton-antisoliton pair does not contribute to the dc transport due to its charge neutrality, but it does contribute to polarization [35, 59].

The increase in the field strength suppresses the dielectric constant distinctly in MHz frequency region, the relative dielectric constant ϵ_{r1} decreases from about 1.25 $\times 10^4$ to 1.05 $\times 10^4$ in the field of 10 V/cm at 2.0 MHz, as shown in Fig. 6(a). One may argue that under the dc bias, the true temperature of the 'heated' sample must be higher than the apparent temperature. Here we would clarify that E_B is moderate, and in contrast, stronger polarization is observed at higher temperatures [Fig. 6(b)]. Thus it is confirmed that the dielectric constant ϵ_{r1} decreases with the increasing of dc bias. Qualitatively, $\epsilon_{r1} \propto N\alpha$, where N is the number of the dipoles per unit volume, α is the polarizations coefficient. If N remains unchanged in electric field, then α should decrease. Since α describes the local polarization, its decrease will not change the dc conductivity, in contrast with the experimental data.

Hence, N would decrease in an external dc bias field, consistent with the soliton tunneling model [Fig. 7]. As the applied dc field increases, the potential barrier for the BSP activation would decrease. For the solitons having positive charges, the preferred direction of tunnelling would be along the external electric field, whereas the inverse applies for the negatively charged solitons, as shown in Fig. 7. Consequently, the probability of solitons tunnelling enhances, inducing the suppression of polarization by dc bias.

The ′grain and grain boundary′ model should also be discussed in connection with the suppression of the dielectric constant in dc bias [63]. The model describes a reduction of the dielectric constant with increasing dc bias in ceramic systems. In such materials, double back-to-back Schottky potential barriers in micrometer scales are created at interfaces between grains due to charge trapping. Under a dc bias, the total width of the depletion region increases and then the dielectric constant should be suppressed [63]. However, in our experiment below T=50 K, such polarization (often found at higher temperatures) should be frozen out and thus can be excluded.

D. The mechanisms of non-linear conductance in the CCDW phase

Intrinsic nonlinearity unrelated to CCDW ground state of 1T-TaS_2 should be discussed. The two most common single-particle mechanisms are field ionization and impact ionization of charged impurities. To ionize impurity electrons with a binding energy of about 20 K by applying an electric field of about 10 V/cm would require that the impurity wave function extend over lengths of an order of 1.0 μm. It is possible, however, to generate extra carriers in weak electric fields if free carriers can be accelerated to sufficient kinetic energies to ionize bound electrons upon collision. This would require the mobility of carriers to be about 10^5 $cm^2/(V{\cdot}s)$. However, σ_{dc} at T=20 K is about 2.5 $(\Omega{\cdot}cm)^{-1}$ and the density of carriers is order of $10^{19}/cm^3$ [16], thus the mobility is estimated to be only several $cm^2/(V{\cdot}s)$. Further, the experimental results from Ref. [28] also support a mobility less than 20 $cm^2/(V{\cdot}s)$. Hence it is plausible to believe that the non-linear conductivity is associated with the electrons localization or CCDW condensate.

The previous studies of 1T-TaS_2 reported that in the presence of the non-linear conductance the changes of Hall coefficient could be neglected at T=4.2 K [16]. Uchida *et al.* proposed that the non-linear conduction in 1T-TaS_2 might be explained in terms of the collective excitations in pinned CDW, if the creation of soliton in pair with antisoliton occurs in 2D-CDW systems. Such excitations are expected to occur in the current temperature range as Rice *et al.* [56] proposed that the average number of φ-particle pairs $N(T) \propto exp(-E_0/k_B T)$, where E_0 is the activation energy of solitons inside the Peierls gap. Therefore, the existence of BSPs bearing with relatively small excitation energies would be more plausible as Rajaraman discussed [34] and the associated contribution to non-linear conductivity may be understood: they cannot move freely without external excitations, as the applied dc field increases, the potential barrier for both of the positively charged and negatively charged solitons would decrease [Fig. 5(b)], driving a non-linear dc conductivity accompanied by the suppressed dielectric constant.

Vaskivskyi *et al.* also observed the non-linear CVCs and suggested that the unusual exponential curves can be understood within the model in terms of trapped carrier tunnelling through barriers resulting from the transient domain wall structure [17]. Nevertheless, as the localized or trapped electrons become easier to form delocalized charges in the presence of a dc field, the conductivity of the system enhances. However, we note that the CVCs in CCDW state of 1T-TaS_2 could not be fitted well by exponential relation in weak field range [17]. Thus the transport properties of 1T-TaS_2 at low temperatures may be a complex issue. Combining with our experimental results on dielectric properties, a process which can not be attributed to the hopping charge transport has been identified, thus we suggest that the collective excitations in the CCDW state of 1T-TaS_2 can not be neglected. In very recent reports, Cho *et al.* did observe two well-confined and non-metallic in-gap states, located on the domain wall center and edges of neighboring domains [33]. Further experiments are necessary to clarify whether the two in-gap states correspond to the two contributors: the localization states and the collective excitations (BSPs).

In Ref. [64], Rozhavsky *et al.* proposed a dynamic mechanism of topological charge creation in a commensurate 1D-CDW near the contact with a normal metal, and also predicts the presence of non-linear conductivity. However, the associated feature of the CVC has a downward curvature in their model, which is inconsistent with our experimental data. Any way, to disclose the mechanisms of the non-linear conductivity in 2D-CDW systems is a complex issue, it is always a problem to distinguish between solitons motion from other forms of non-uniform CDW excitations (e.g., creep) and thus more efforts should be needed in future research work.

IV. SUMMARY

In summary, we have studied the in-plane electric field dependent charge dynamics in the in-plane of 1T-TaS_2 single crystals in the CCDW phase. A large dielectric constant $\sim 10^4$ exhibits up to 10^7 Hz, which is possibly due to the occurrence of bound soliton pairs or charge ordering of the system. The dielectric spectra disperse only slightly in our measured temperature and frequency range. We propose that besides the hopping carriers, the delocalization of BSPs may be another contributor to suppress $\epsilon_1(\omega)$ and the non-linear conductivity with increasing of dc bias.

ACKNOWLEDGMENTS: The authors are very grateful for instructive discussions with DongWu. The research work was supported by the National Science Foundation of China (Grant No. 10704054). Work at Brookhaven National Laboratory is supported by the US DOE, Contract No. DE-SC0012704.

[1] G. Gruner, Rev. Mod. Phys. **60**, 1129 (1988).

[2] P. Monceau, Adv. Phys. **61**, 325 (2012).

[3] Chih-Wei Chen, Jesse Choe and E Morosan, Rep. Prog. Phys. **79** 084505, (2016).

[4] M. E. Itkis, F. Ya. Nad, and P. Monceau, J. Phys.: Condens. Matter **2**, 8327 (1990).

[5] Jean-Paul Pouget, Comptes Rendus Mecanique **17**, 332 (2016).

[6] F. Ya. Nad and P. Monceau, Phys. Rev. B **46**, 7413 (1992).

[7] G. Gruner, A. Zawadowski, and P. M. Chaikin, Phys. Rev. Lett. **46**, 511 (1981).

[8] I. G. Gorlova, S. G. Zybtsev, and V. Ya Pokrovskii, JETP Lett. **100**, 256 (2014).

[9] D. Staresinic, K. Biljakovic, W. Brutting, K. Hosseini, P. Monceau, H. Berger, and F. Levy, Phys. Rev. B **65**, 165109 (2002).

[10] D. Staresinic, P. Lunkenheimer, J. Hemberger, K. Biljakovic, and A. Loidl, Phys. Rev. Lett.

96, 046402 (2006).

[11] M. E. Itkis, F. Ya. Nad, S. V. Zaitsev-Zotov, F. Levy, Solid State Commun. **71**, 895 (1989).

[12] V. Nasretdinova, S. Zaitsev-Zotov, Physica B **407**, 1874 (2012).

[13] F. J. Di Salvo, J. A. Wilson, B. G. Bagley, and J. V. Waszczak, Phys. Rev. B **12**, 2220 (1975).

[14] J. A. Wilson, F. J. Di Salvo, and S. Mahajan, Adv. Phys. **24**, 117 (1975).

[15] A. A. Sinchenko, P. Lejay and P. Monceau, Phys. Rev. B **85**, 241104(R) (2012).

[16] S. Uchida, K. Tanabe and S. Tanaka, Solid State Commun. **27**, 637 (1978).

[17] I. Vaskivskyi, I. A. Mihailovic, S Brazovskii, J. Gospodaric, T. Mertelj, D. Svetin, P. Sutar, and D. Mihailovic, Nat. Commun. 7, 11442 (2016), and the Supplementary Information.

[18] J. Chang, E. Blackburn, A. T. Holmes, N. B. Christensen, J. Larsen, J. Mesot, R. Liang, D. A. Bonn, W. N. Hardy, A. Watenphul, M. V. Zimmermann, E. M. Forgan, and S. M. Hayden, Nat. Phys. **8**, 871 (2012).

[19] Manuel G. Velarde, Werner Ebeling, Alexander P. Chetverikov, Comptes Rendus Mecanique **340**, 910 (2012).

[20] R. Comin, R. Sutarto, F. He, E. H. da Silva Neto, L. Chauviere, A. Frano, R. Liang, W. N. Hardy, D. A. Bonn, Y. Yoshida, H. Eisaki, A. J. Achkar, D. G. Hawthorn, B. Keimer, G. A. Sawatzky and A. Damascelli, Nature materials **14**, 796 (2015).

[21] M. Yoshida, R. Suzuki, Y. J. Zhang, M. Nakano, and Y. Iwasa, Sci. Adv. 1, e1500606 (2015). Masaro Yoshida, Takashi Gokuden, Ryuji Suzuki, Masaki Nakano, and Yoshihiro Iwasa, Phys. Rev. B **95**, 121405(R) (2017).

[22] M. J. Hollander, Y. Liu, W. J. Lu, L. J. Li, Y. P. Sun, J. A. Robinson, and S. Datta, Nano Lett. **15**, 1861 (2015).

[23] H. Mutka, L. Zuppiroli, P. Molinie, and J. C. Bourgoin, Phys. Rev. B **23**, 5030 (1981).

[24] Alberto Ubaldini and Enrico Giannini, Journal of Crystal Growth **401**, 878, (2014).

[25] A. A. Sinchenko, P. Lejay, O. Leynaud, and P. Monceau, Phys. Rev. B **93**, 235141 (2016).

[26] Y. Huang, B. F. Hu, T. Dong, A. F. Fang, P. Zheng, and N. L. Wang, Phys. Rev. B **86**, 205123 (2012).

[27] DiSalvo and J. E. Graebner, Solid State Commun. **23**, 825 (1977).

[28] P. D. Hambourger and F. J. Di Salvo, Solid State Commun. **35**, 405 (1980).

[29] B. Dardel, M. Grioni, D. Malterre, P. Weibel, Y. Baer, and F. Levy, Phys. Rev. B **45**, 1462 (1992).

[30] Ju-Jin Kim, W. Yamaguchi, T. Hasegawa, and K. Kitazawa, Phys. Rev. Lett. **73**, 2103 (1994).

[31] L. Perfetti, P. A. Loukakos, M. Lisowski, U. Bovensiepen, H. Berger, S. Biermann, P. S. Cornaglia, A. Georges, and M. Wolf, Phys. Rev. Lett. **97**, 067402 (2006).

[32] B. Burk, R. E. Thomson, A. Zettl and John Clarke, Phys. Rev. Lett. **66**, 3040 (1991).

[33] Doohee Cho, Gyeongcheol Gye, Jinwon Lee, Sung-Hoon Lee, Lihai Wang, Sang-Wook Cheong and Han Woong Yeom, Nature Comm., 8, 392 (2017).

[34] R. Rajaraman, Phys. Repts. **21C**, 227 (1975).

[35] I. V. Krive and A. S. Rozhavsky, Solid State Commun. **55** 691 (1985).

[36] P. Lunkenheimer, S. Krohns, S. Riegg, S. G. Ebbinghaus, A. Reller, and A. Loidl, Eur. Phys. J. Special Topics **180**, 61 (2010).

[37] S. R. Elliott, Adv. Phys. **36**, 135 (1987).

[38] Y. C. Ma, Cuimin Lu, Xuewei Wang, Xueli Du, Lijun Li, and Cedomir Petrovic, J. Phys.: Condens. Matter **28**, 395901 (2016).

[39] T. Ivek, T. Vuletic, S. Tomic, A. Akrap, H. Berger and L. Forro, Phys. Rev. B. **78** 035110 (2008).

[40] P. Lunkenheimer and A. Loidl, Phys. Rev. Lett. **91**, 207601 (2003).

[41] A. K. Jonscher, Nature **267**, 673 (1977).

[42] R. J. Cava, R. M. Fleming, P. B. Littlewood, E. A. Rietman, L. F. Schneemeyer, and R. G. Dunn Phys. Rev. B **30**, 3228 (1984).

[43] A. Fischer, P. Pahner, B. Lussem, K. Leo, R. Scholz, T. Koprucki, K. Gartner, and A. Glitzky, Phys. Rev. Lett. **110**, 126601 (2013).

[44] N. Croitoru and C. Popescu, phys. stat. sol. (a) **3**, 1047 (1970).

[45] Marshall J. Cohen and A. J. Heeger, Phys. Rev. B **16**, 688 (1977).

[46] J. B. Balaguru Rayappan, S. A. Cecil Raj, N. Lawrence, Physica B **405**, 3172, (2010).

[47] A. V. Inyushkin, A. N. Taldenkov, and V. V. Florentiev, Synthetic Metals, 19, 843 (1987).

[48] M. D. Nunnez-Regueiro, J. M. Lopez-Castillo, and C. Ayache, Phys. Rev. Lett. 55, 1931 (1985).

[49] As thermal conductivity κ of 1T-TaS_2 below T=100 K is absent, the value κ of TaS_3 is taken as a reference. Moreover, the anisotropy of the thermal conductivity of 1T-TaS_2 should be considered: it is acceptable that d/S and κ in ab-plane (cleaved surface) would be remarkably different to the case of $\parallel$ c-axis. For our measurements configuration, the heat flow along the

c-axis is remarkable for the large ratio $'$contact area/thickness$'$, hence at low frequencies the heating of the substrate as a whole gives a large contribution to remove the Joule heating. That is, as most of the heating power flows into the sapphire substrate, the relaxation time τ_{SH} (of the sample and the substrate as a whole) enhances significantly. Thus the conductivity under dc bias mainly gives the chordal contribution. Nevertheless, the thermal relaxation effects could be neglected above 10 kHz, though the systematic research work on the thermal properties of 1T-TaS_2 is still necessary.

[50] P. B. Littlewood, Phys. Rev. B **36**, 3108 (1987).

[51] W.-y. Wu, L. Mihaly, and G. Gruner, Solid State Commun. **55**, 663 (1985).

[52] I. G. Austin and N. F. Mott, Advances in Physics **18**, 41 (1969).

[53] N. Dean, J. C. Petersen, D. Fausti, R. I. Tobey, S. Kaiser, L.V. Gasparov, H. Berger, and A. Cavalleri, Phys. Rev. Lett. **106**, 016401 (2011).

[54] S. V. Zaitsev-Zotov and V. E. Minakova, Phys. Rev. Lett. **97**, 266404 (2006).

[55] S. Brazovskii, C. Brun, Z.-Z. Wang and P. Monceau, Phys. Rev. Lett. **108**, 096801 (2012).

[56] M. J. Rice, A. R. Bishop, J. A. Krumhansl, and S. E. Trullinger, Phys. Rev. Lett. **36**, 432 (1976).

[57] B. Horovitz and J. A. Krumhansl, Phys. Rev. B **29**, 2109 (1984).

[58] T. Matsuura, J. Hara, K. Inagaki, M. Tsubota, T. Hosokawa, and S. Tanda, EPL **109**, 27005 (2015).

[59] J. H. Miller, C. Ordonez, and E. Prodan, Phys. Rev. Lett. **84**, 1555 (2000).

[60] K. Inagaki, M. Tsubota, and S. Tanda, Phys. Rev. B **81**, 113101 (2010).

[61] A. Rojo-Bravo, V. L. R. Jacques, and D. Le Bolloch, Phys. Rev. B **94**, 201120 (2016).

[62] S. Krohns, P. Lunkenheimer, Ch. Kant, A. V. Pronin, H. B. Brom, A. A. Nugroho, M. Diantoro, A. Loidl, Appl. Phys. Lett. **94**, 122903 (2009).

[63] Timothy B. Adams, Derek C. Sinclair, and Anthony R. West, Phys. Rev. B **73**, 094124 (2006).

[64] A. S. Rozhavsky, Yu. V. Pershin, and A. S. Kovalev, Low Temperature Physics **23**, 996 (1997).