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The evidence of bound solitons delocalization in o-TaS_3 under dc bias from sum rule

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ABSTRACT

The electric field dependence of the dielectric properties below 50 K along the chain direction of o-TaS_3 has been investigated. Under a dc bias field exceeding the lower threshold E'_T, the real part of dielectric constant $\epsilon_1(\omega)$ decreases possibly due to the decreasing concentration of bound solitons. We propose that the delocalization of bound solitons with the increasing applied electric field could be used to explain the reduction of $\epsilon_1(\omega)$ and the enhancement of the dc conductivity. This has also been confirmed by the quantitative comparison between the enhancement of the dc conductivity and the change of the dielectric spectral weight.

1. Introduction

The existence of charge density wave (CDW) phase solitons — the topological phase defects — has been confirmed in quasi-one-dimensional systems [1–4]. Up to date, more work has indicated that solitons contribution must be a key to the unconventional nature associated with CDW system [5–11]. Itkis et al. suggested that the lower threshold field E'_T in o-TaS_3 below 60 K was relevant to solitons [12]. In recent reports, Matsuura et al. observed a new ac-dc interference spectrum in the differential conductivities of o-TaS_3 and proposed the origin was the depinning of solitons from impurity potentials [7]. In very recent studies, Rojo-Bravo et al. presented 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 [13]. However, the contribution to the charge transport properties from CDW phase solitons is often revealed only at rather lower temperatures due to the low activation energy.

Temperature and field dependent complex dielectric or conductivity spectrum is a powerful probe for investigating the charge transports and polarization behavior of various systems [14–17]. At a fixed temperature, the complex dielectric spectrum $\hat{\epsilon}(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega)$ of a substance is ultimately defined by the polarizations of the total contributing charged microscopic particles. For CDW systems, under a weak applied dc electric field E, the bound soliton-antisoliton pair has no contributions to the dc transport due to its charge neutrality, but it does contribute polarizations nontrivially [18,19]. As the applied dc field increases, the probability of solitons tunnelling (delocalization tendency) enhances. The corresponding features in charge transport and dielectric properties will change accordingly. Further, the quantitative analysis could be carried out by comparing the enhancement of the dc conductivity with the change of the dielectric spectral weight. To the best of our knowledge, the field dependent dielectric response from phase solitons has not been clarified yet [15,20,21], even the characteristics of the charge carriers in CDW systems are still in disagreement [4,13,22–25]. Thus the investigations of dielectric properties for o-TaS_3 crystals under various dc bias would be of interest, especially at lower temperatures when the screening effect of normal electrons diminishes.

In this report we systematically study the electric field dependent charge dynamics along the chain direction of o-TaS_3 below 50 K. We focus on the dielectric properties under the applied E covering the lower threshold E'_T. Under $E > E'_T$, the real part of dielectric constant $\epsilon_1(\omega)$ starts to decrease possibly due to the decreasing concentration of bound solitons. We propose that the delocalization of bound solitons could be used to explain the reduction of $\epsilon_{r1}(\omega)$, $\epsilon_{r2}(\omega)$ and the enhancement of the conductivity as E increases. This has been also supported by the sum rule or the quantitative comparison between the enhancement of the dc conductivity and the change of the dielectric spectral weight.

2. Results and discussions

All the primary data, the temperature dependent conductivity, complex impedance spectra and the field dependent conductivity, have

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been obtained in our previous experiments as described in Ref. [15]. At $T = 185$ K, with the establishment of the CDW, one relaxation process due to the elastic pinned CDW screened by uncondensed carriers emerges at frequencies higher than 10^7 Hz. With decreasing temperature, it shifts towards low frequencies, indicating the gradually freezing of this process. Below $T = 70$ K, another process relevant to solitons (localized pinned CDW excitations) starts to dominate the polarization properties of o-TaS_3. To be consistent with Ref. [14], we call the first process α and the second one β. Two models of the β process are discussed by Staresinic et al.: one requires the screening of solitons by the free carriers; the other is based on the idea of the hopping of the solitons between the impurity sites, which relies on the theory of ion hopping in glasses [14]. As phase solitons have an appropriate activation energy of $(1{-}2)T_P$ and would create a band within CDW gap, it has been speculated that the CDW phase solitons would play an important role in the dc transport and the dielectric properties of o-TaS_3 crystals [4,12,14,18]. In the context, we will analyze the enhancement of dc conductivity above the lower threshold E'_T below $T = 50$ K in more detail based on the field dependent dielectric properties.

At a fixed temperature below 50 K, a part of the bound solitons are expected to be polarized locally whereas the others could be thermally excited over or tunnelled through the energy barrier with the help of the applied electric field [4,26]. The increase in the field strength suppresses the dielectric constant distinctly in the measured frequency region: at 1.0 kHz, the relative dielectric constant ϵ_{r1} decreases from about 5.0 $\times 10^6$ to 2.0 $\times 10^6$ in the field of 10 V/cm; whereas at higher frequency region, the suppression of ϵ_{r1} weakens gradually, as shown in Fig. 1(a). Qualitatively, as $\epsilon_{r1} = 1 + N\alpha/\epsilon_0$ (N is the number of the dipoles per unit volume, α is the polarizations coefficient), $N\alpha$ would decrease. If N remains unchanged under electric field, then α should decrease. However, as α presents the local polarization, its decreasing will not change the dc conductivity, which is in contradiction to the experimental data. Thus we propose that the concentration of the bound solitons decreases with increasing E. In an electric field with sufficient strength, after the total number of bound solitons are delocalized, the dielectric constant would no longer decrease, but saturate as observed in Ref. [15], see Fig. 1(a). Moreover, the possible suppression of the back flow effects from the solitons (free or unbound solitons could transport charges, so they can be considered as new carriers) may also play a role in reducing ϵ_{r1}, since under dc bias these new carriers should transfer at long distances.

The field dependent imaginary part $\epsilon_{r2}(\omega)$ of relative dielectric constant at $T = 30$ K is shown in Fig. 1(b). The loss peaks were suppressed with the increase of the field: the maximum of the $\epsilon_{r2}(\omega)$ decreases from about 1.4 $\times 10^6$ ($E = 0$) to 0.4 $\times 10^6$ ($E = 10$ V/cm), indicating an essential reduction of the polarization loss. For an idealized system with only one polarization mechanism, it could be described well by the Debye model; whereas in real experiments the relaxation behavior implies a generalized Debye model with a distribution of relaxation time. The thin curves are fitting results with the Cole-Cole function: $\hat{\epsilon}(\omega) = \epsilon_{HF} + (\epsilon_s - \epsilon_{HF})/[1 + (i\omega\tau_0)^{(1-h)}]$, which provides a phenomenological description of symmetrically broadened loss peaks, τ_0 is an averaged characteristic relaxation time, while ϵ_s and ϵ_{HF} are the dielectric constant in static and at higher frequency limit, respectively. The resultant h ranges 0.30–0.40, implying a distribution of relaxation time.

The dielectric behavior is attributed to bound solitons which could be locally polarized, rather than the free solitons that can transfer in long distances. For the equation of motion relevance to CDW system, the realistic pinning potential of the phase would have a periodic form as expected [4]. Thus the exact solutions of the equation should include the soliton-antisoliton 'doublet' [27]. Instead of separating into the discrete soliton or antisoliton which is infinitely far apart with each other in infinite time, the relative separation for the members of the doublets oscillate periodically [27]. This doublet solution is also a 'breathing' one and can be thought of as a bound soliton-antisoliton pair (BSP).

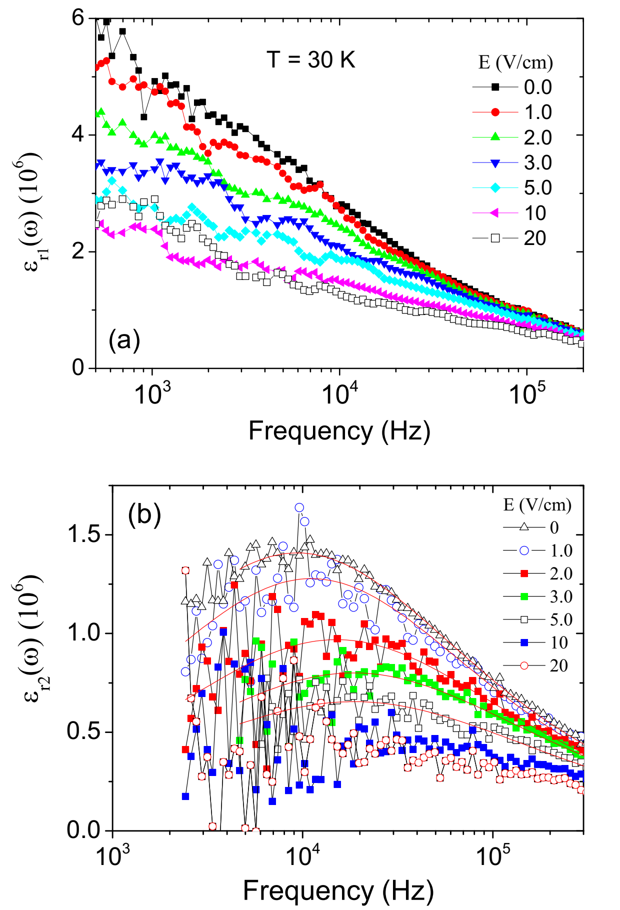

Fig. 1. (a) The dielectric spectra at various dc bias and $T = 30$ K. (b) The bias dependent imaginary part of the dielectric constant and the corresponding fittings at $T = 30$ K. Note that the dielectric response shows significant dependence at the lower electric fields but is independent above $E = 10.0$ V/cm. The fittings indicated by the thin lines are based on the generalized Debye model.

In a weak dc electric field E, the BSP has no contributions to the dc transport due to its charge neutrality, as shown in Fig. 2, but it does contribute to the polarizations nontrivially [18,19]. Though the intrinsic properties of the BSP are not known well by scientists up to now, it is plausibly assumed that the soliton and the anti-soliton are bound with each other, thus neither could move freely. As the applied dc field increases, the potential barrier for the solitons activation would decrease. For the solitons having positive charges, the preference direction of motion would be along the external electric field, whereas the inverse applies for the negatively charged solitons. Consequently, the probability of solitons tunnelling or the delocalization tendency enhances, driving an enhancement of the dc conductivity. This behavior may be necessary at lower temperatures for CDW to form a coherent condensate in a larger E exceeding the classical depinning threshold field E_{T-cl}, as we know that phase defects — the phase solitons — would inhibit the coherence of the CDW phase. In this respect, the delocalization process of the solitons could be seen as a preparation of the complete coherence of the whole CDW condensate.

To understand the electric field dependence of the dielectric spectra in our experiments, the quantitative analysis could be obtained from the Kramers-Kronig relations of the susceptibility [28,29]:

$$\sigma_1(\omega) = \frac{2}{\pi}\mathbb{P}\int_0^{\infty}\frac{s\sigma_2(s)}{s^2-\omega^2}ds \tag{1}$$

where $\mathbb{P}$ is for the principle integration. Considering that $\hat{\epsilon}(\omega) = \epsilon_0 + i\omega\hat{\sigma}(\omega)$, $\sigma_2(\omega)/\omega = \epsilon_0 - \epsilon_1(\omega)$, by setting $\omega = 0$, $\sigma_{dc} = \sigma_1(0)$