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Glass transition in charge-density-wave systems o-TaS_3 and $K_{0.3}MoO_3$

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Abstract. We present experimental evidence for the glass transition in charge density wave (CDW) superstructure of two quasi one-dimensional systems, o-TaS_3 and $K_{0.3}MoO_3$. Low frequency dielectric response of both systems exhibits typical glass-like phenomenology, featuring the splitting of the relaxational spectrum into two processes on decreasing the temperature and the subsequent freezing of primary or α process at finite temperature of glass transition T_g. Below T_g secondary, or β process becomes dominant. Activation energies obtained from the temperature evolution of the characteristic relaxation times of α and β processes correspond to the activation energies of the temperature evolution of the DC conductivity above and below T_g respectively. The results are discussed in respect to the relevant theories of low frequency CDW dynamics, with the emphasis on the Coulomb hardening of CDW in absence of screening by free carriers. An attempt to understand observed differences in freezing of CDW in o-TaS_3 and $K_{0.3}MoO_3$ is made.

Ideal properties of charge density wave (CDW) systems are substantially changed in real systems due to the interaction with impurities and uncondensed free carriers. Impurity pinning prevents CDW condensate to slides below some finite threshold field E_T, shifts the CDW phase mode from zero to finite frequency (pinning resonance), and its inherent randomness leads to the distortion of the CDW phase and proliferation of metastable states that are responsible for the low frequency realxation mode. In addition, extra charge associated with phase distortions (both homogeneous and inhomogeneous) is responsible for the intra CDW electrostatic (Coulomb) interaction. At high temperatures the screening of phase distortions by uncondensed free carriers minimizes this effect. However, lowering of temperature decreases substantially the density of free carriers due to the semiconducting nature of majority of CDW systems and Coulomb stiffening becomes important in the descreened limit. Descreening leads to significant changes of the properties of CDW systems between the high and low temperature range, which occur in a narrow temperature region. Among others, extra contribution to the DC conductivity and second threshold field are observed. In this paper we present evidences based on the extensive study of the low frequency dielectric response of two CDW systems, o-TaS_3 and $K_{0.3}MoO_3$, that these changes are due to freezing of the inhomogeneous elastic excitations of CDW phase, which bears close resemblence to the ordinary glass transition.

We have used impedance analysers specially designed for high precision dielectric measurements at low frequencies and low driving signal amplitudes of highly resistive probes like CDW systems at low temperatures [1,2]. It enabled us to cover substantially wider frequency range than in previosuly reported measurements.

In Figure 1. we present low frequency dielectric response of TaS_3 (Figure 1a and 1b) in the temperature range below 220 K down to 5 K and $K_{0.3}MoO_3$ (Figure 1c) in the temperature range below 80

K down to 5 K. Dielectric response is overdamped (relaxational) and can be described in parts with the modified Debye response with variable width (Cole-Cole function) [1,2].

It is clearly seen that the well-known relaxational mode reported in many publications [3] can be observed only down to finite temperature, below which the relaxation frequency decreases below the accesible frequency range. On the other hand, the second relaxational process developes as a high frequency wing of the relaxational mode and dominates at low temperatures. In order ot distinguish between these two relaxational (overdamped) processes we have labeled the high temperature one *a* process, and the low temperature one β process. β process in TaS_3 has already been observed in [4], however it has not been distinguished from *a* process. On the other hand, observation of β process in $K_{0.3}MoO_3$ is entirely new.

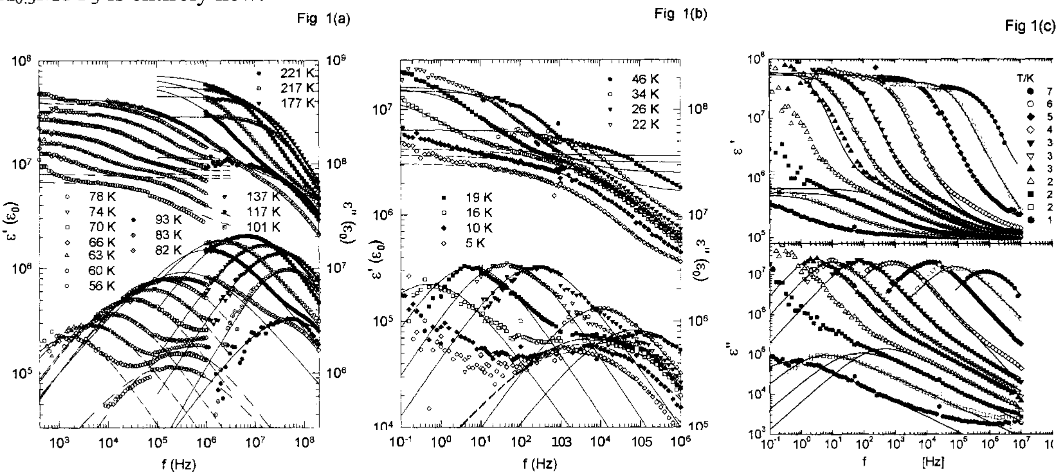

Figure 1. Frequency dependences of real ε' and imaginary ε" part of the dielectric response of TaS_3 at selected temperatures between 220 K and SO **K** (a) and 50 K and 5 K (b) The same for $K_{0.3}MoO_3$ between 80 K and 5 K (c). The lines are Cole-Cole tits of the experimental data.

In Figure 2 the temperature dependence of the characteristic relaxation times τ for both processes is presented in usual Arrhenius plot, together with the temperature dependence of DC resistivity ρ_{DC}. Temperature evolution of τ for both processes shows roughly activated behaviour presented by solid lines in Figure 2. Activation energies E, are given in Table 1. Interestingly, the temperature evolution of ρ_{DC} shows two regimes of activated behaviour, where the high temperature activation energy is close to the activation energy of *a* process, wheras the low temperature activation energy is close to the activation energy of β process.

We can see that τ of α process, τ_α, becomes very long on decreasing temperature. Continuous slowing down (i.e. increase of the relaxation time) of the relaxational response is a general feature of glasses, observed on approaching the glass transition temperature T, [5]. The convention is that the transition to glass occurs at the temperature T, where τ of corresponding relaxation process (typically named *a* process) becomes longer than 10^2-10^3 s. It signifies that the excitations contributing to *a* process become frozen on experimental time scales and the corresponding degrees of freedom are not accesible any longer. We believe that it is important to consider our systems in this framework, as it tells us that the CDW excitations reponsible for the relaxational mode are frozen at low temperatures.

Extrapolation of the activated decrease (solid line) of τ_α to 10^2 s enables the estimate of T„ of TaS_3 to be about 30 K and of $K_{0.3}MoO_3$ to be about 23 K. However, at least for TaS_3 τ decreases faster than activated, and the suitable VF fit (dashed line) shifts the estimate of T, to about 42 K.

Slowing down of the relaxational mode of CDW has been throroughly considered in literature [6,7]. Regardless of the initial model, the relavant theories consider the dynamics of the slowly varying CDW phase in the pinning potential corresponding to the phase domains. Descreening prevents the domains to relax independently, leading to the cooperative dynamics and increase of the relaxation time. However, this picture cannot hold at arbitrary low temperatures as the relaxation occurs at finite length scales of the order of the domain size. Once there is not enough free carriers per domain to screen it efficiently, the relaxation would be inhibited, or frozen. Based on the estimated value of T, and expected decrease of the density of free carriers we could get a rough value of the critical density for the freezing of *a* process. **As** seen in Table 1, at T, there is approximately one free carrier per $3 \cdot 10^{-15}$ cm^3 in TaS_3 and $3 \cdot 10^{-13}$ cm^3 in $K_{0.3}MoO_3$. This is consistent with the estimate of the phase coherence volume V_{coh} of about 10^{-14} cm^3 [8]. Therefore we can set a criterion for glass transition saying that relaxational mode *(a* process) freezes when there is less than one free carrier per domain of phase coherence.

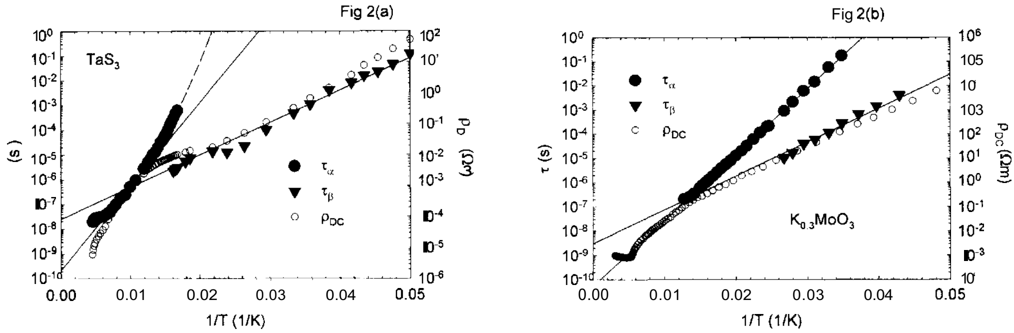

Figure 2. a) Temperature evolution of the characteristic relaxation times τ of *a* and β process, as well as DC resistivity ρ_{DC} in TaS_3 b) the same for $K_{0.3}MoO_3$. solid lines are activated tits, and dashed line Vogel-Fulcher fit.

Table 1. Peierls transition temperature (T_P), room temperature free carrier density (n, (RT)), activation energy obtained from the temperature evolution of DC resitivity (E, ρ_{DC}), and characteristic relaxation time of *a* (E, τ_α) and β (E, τ_β) process, glass transition temperature (Tg), estimated density of free carriers at T_g (n, (T,)) and the corresponding critical volume (V_{cr}) for freezing of *a* process in TaS_3 and $K_{0.3}MoO_3$.

	T_P (K)	n_e (RT) (cm^{-3})	E_a ρ_{DC} (K)	E_a τ_α (K)	T_g (K)	E_a ρ_{DC} (K)	E_a τ_β (K)	n_e (T_g) (cm^{-3})	V_{cr} (cm^3)
TaS_3	215	10^{22}	830	820	42	300	300	$3\,10^{14}$	$3\,10^{-15}$
$K_{0.3}MoO_3$	176	$3\,10^{21}$	530	625	23	300	320	$3\,10^{12}$	$3\,10^{-13}$

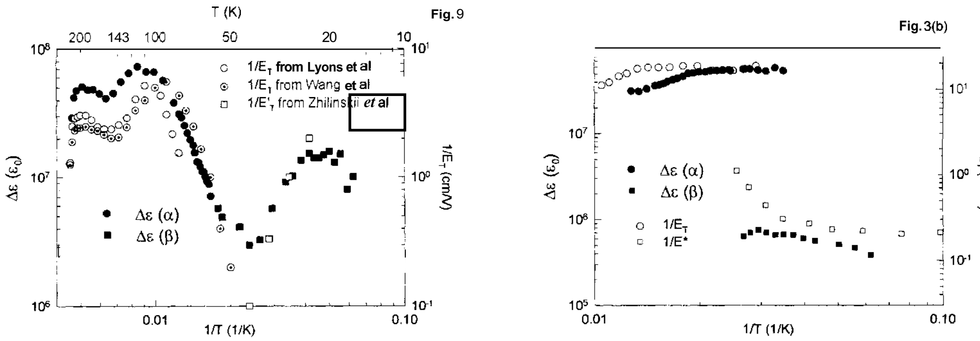

Figure 3. a) Temperature evolution of the amplitude $\Delta\varepsilon$ of a and β process, as **well** as threshold **field** E_T for sliding (from [13,14]) and slight nonlinearity offset E' (from [10]) in TaS_3 **b)** Temperature evolution of the amplitude $\Delta\varepsilon$ of a and β process, as **well** as threshold fields for sliding (E_T) and steep current E' in $K_{0.3}MoO_3$ (from [15]).

It has been shown [15] that the soliton lattice can contribute to the low frequency dielectric response of CDW, and that the increase of the soliton length leads to the slowing down of the relaxation time. As the soliton size still increases with the decrease of temperature even in the nominal absence of free carriers [12], it would thus lead to the temperature dependent low frequency response. The hopping of free carriers between the pockets situated at soliton centers can provide also non-negligable contribution to the linear conducitivity. Weather this could explain the fact that the temperature evolution of τ of β process follow closely that of σ_{DC} is not clear, however this is a basis for further consideration.

Higher amplitude of β process relative to α process in TaS_3 could be explained by higher abundance of solitons than in $K_{0.3}MoO_3$, as suggested by lower critical volume and cooresponding ly higher impurity content. This is also consistent with higher excess contribution to σ_{DC} at low temperatures. Stronger overlapping of solitions in TaS_3 could lead to the intermediate regime of correlated soliton hopping above E' [10], which is not observed in $K_{0.3}MoO_3$.

In conclusion, we have shown that the transition between the low and the high temperature CDW phase is the consequence of freezing of inhomogeneous elastic phase distortions due to the Coulomb hardening in the descreened limit. The freezing scenario resembles in many aspects the glass transition, which here occurs on the level of superstructure, and we have deduced the criterion for freezing. We have suggested the origin of the relaxational mode below T_g to be the dynamics of topological defects.

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