Doping effects on the low-energy excitations of the charge density wave system o-TaS_3

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Abstract. We report on the effect of doping on the relaxational dynamics and the specific heat (C_p) of the charge density wave (CDW) system o-TaS_3. Isoelectronic substitution of Ta atoms with 0.2%-0.5% of Nb suppresses the primary relaxation process responsible for the glass-like dielectric response of pure o-TaS_3 but only slightly affects the secondary process as well as the low energy excitation (LEE) contribution to C_p. Our results show that the primary relaxation process is mainly due to long range deformations of the CDW, which are prevented by doping. The secondary process and the LEE contribution to C_p originate from local topological defects of the CDW still remaining in doped samples.

The low frequency relaxation dynamics of a CDW superstructure, together with the pinning resonance, bears striking resemblance with canonical glass forming systems. Wide temperature and frequency range dielectric spectroscopy of CDW systems o-TaS_3 [1a] and $K_{0.3}MoO_3$ [1b] demonstrates the splitting of the relaxation spectrum in two processes, slower α (or primary) and faster β (or secondary), on decreasing temperature. Here we show how these processes in o-$TaS_3$are affected by a slight doping.

Inset of Fig. 1b shows the most conventional presentation of the glass transition - the Arrhenius (1/T) plot of characteristic frequencies of the two relaxational processes α and β for the pure system and the slowing down of the remaining β process in the doped one. Cooperatively relaxing CDW domains of coherent phase coupled through electrostatic interaction give rise to the primary process. The absence of free carrier screening at lower temperatures leads to the freezing of the elastic degrees of freedom and α process slows down. Doping prevents the long range deformations of the CDW so that α process becomes suppressed. The remaining β process represents the dynamics of topological defects developed on the domain boundaries which is not affected by doping, as this process keeps basically the same amplitude and relaxation time in both, pure and doped samples. Glass transition is manifested also in the nonlinear properties as a well known transition from weak pinning to strong pinning. In the pure sample the amplitudes of α and β processes are correlated with the high temperature E_T and low temperature E_T*, threshold fields for nonlinear conductivity [1]. In other words, we might say that doping completely changes the energy landscape in the way that increases enormously the energy barriers between the adjacent valleys, hindering the corresponding relaxation. However, the corrugation within the valley remains almost unchanged.

We have shown previously that the stiffening of the CDW affects the phonon dynamics and consequently C_p, governed essentially by phonons. This occurs through a change in the localization, i.e. the correlation length of phonons [2]. Doping strongly influences the lattice as well by decreasing the Debye temperature (i.e. transverse sound velocity), as demonstrated in Fig. 2a. Coupling of the chains to domains changes as they become shorter in the doped system, so does the band bending. Change of the effective dimensionality by doping might be also relevant [3]. However, C_p does not change considerably below 1 K where the CDW low-energy excitations (topological defects) give the dominant contribution. The excess specific heat C_{LEE} (Fig. 2b) is only slightly higher for the doped sample.

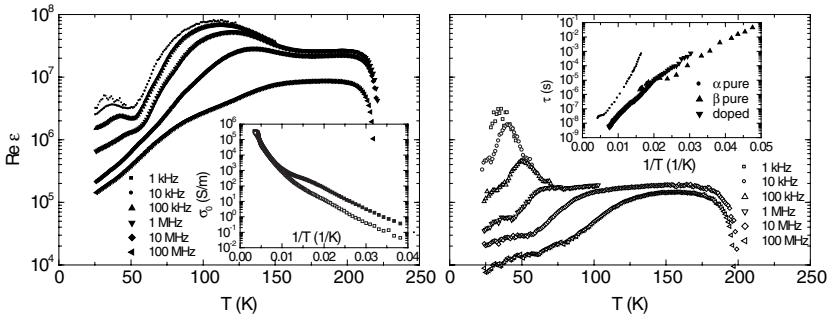

Figure 1. Temperature dependence of the dielectric constant Re ε at various frequencies of pure o-TaS_3 sample (a) and 0.5% Nb doped one with completely suppressed α -relaxation (b). Insets present Arrhenius (1/T) plots of the conductivity for pure and doped samples (a) and their dielectric relaxation times (b).

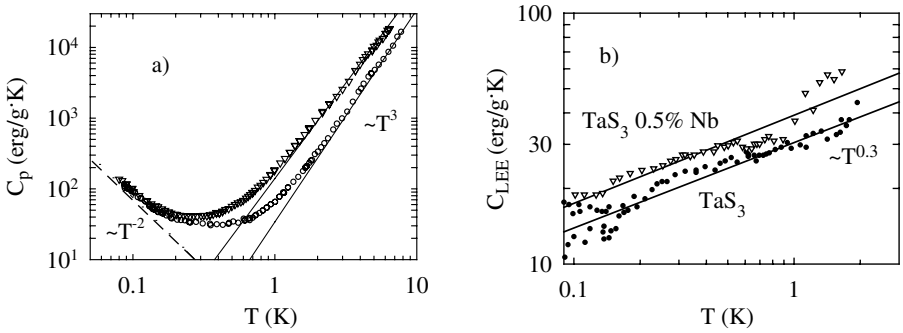

Figure 2. The specific heat of pure and doped o-TaS_3 (a) and the power-law contribution to specific heat $C_{LEE} = CT^{\nu}$ with only slightly larger amplitude for the doped sample (b) (adapted from ref. 2).

The secondary process (remaining below T_g) as an universal feature and an important aspect of the scenario of the glass transition in general is in the focus of the wide field of glasses [4]. Our findings on the doping effects on the CDW relaxational dynamics might have important impact in these topics.

Acknowledgement

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References

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