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Glass-like behavior in the CDW state of the quasi-one-dimensional conductor $K_{0.3}MoO_3$

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Abstract. By measuring the electric conductivity, dielectric susceptibility and thermally stimulated depolarization of the charge density wave system $K_{0.3}MoO_3$, we have found two relaxation processes in the temperature range between 100-10 K. The main relaxation process representing the CDW viscosity shows a critical slowing down behavior and disappears from the measured frequency window at about 28 K. The second one appears below 50 K and becomes the dominant relaxation below 30 K with a relaxation time decreasing with temperature slower than thermally activated, probably corresponding to tunneling between pinned CDW regions. The evidence for this interpretation comes from the temperature and frequency dependence of the conductivity. Together, this gives evidence for a glass-like behavior similar to chalcogenide glass semiconductors.

1-Introduction

Within the last few years, charge density waves (CDW) have been the subject of many experiments to establish similarities to glasses [1]. In real crystals, the random distribution of impurities pin the CDW and prevent its motion. At low temperatures the CDW becomes rigid due to many metastable states, its ground state properties change drastically. There are many manifestations of glassy behavior in electric, dielectric and thermal properties. In particular, thermodynamical investigations at low temperatures have revealed low energy excitations showing all the characteristic features of glasses [1]. The long relaxation times with a critical slowing down behavior of the primary process [2] and the existence of a new collective mechanism with low-energy excitation as solitons, which give rise to an extra contribution to the conductivity [3], are the motivation for studying the characteristics of these phenomena.

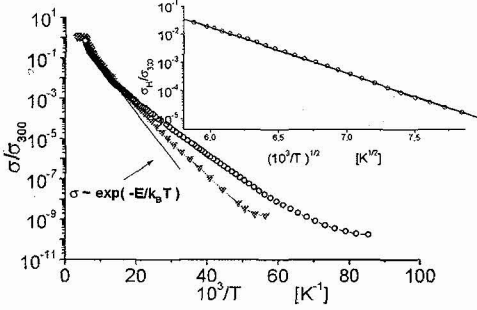
2-Experimental

We investigated three samples of $K_{0.3}MoO_3$ by measuring the electrical conductivity, dielectric susceptibility and thermally stimulated depolarization (TSD) in the temperature range 10 K to 350 K and in a wide frequency range ($10^{-1} - 10^7$ Hz). The measurements were carried out with four probe configurations for conductivity measurement, and two probe configurations for frequency-dependence and TSD measurements. The contacts were made by clamping 25 μ m gold wires with silver paste to gold pads evaporated on the crystals.

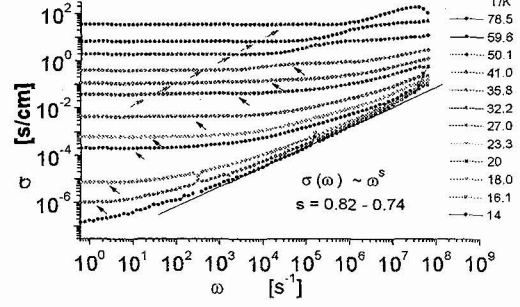
3-Results

The temperature dependence of electric conductivity σ is represented in figure 1, normalized to its value at 300 K for two samples. In the temperature range below $T_P=179$ K (T_P =Peierls Temperature) to $T \approx 75$ K the behavior of $\log \sigma$ is close to linear in a plot versus the reciprocal temperature $1/T$. Different activation energies are obtained for different samples. It varies from about $\Delta_{ac}=530$ K for sample 1 to 670K for sample 3. Below 75 K, the electric conductivity changes its behavior quantitatively from thermally activated and becomes flatter. In the temperature range 75K to 10K, one can distinguish the quality of crystals [3]. Sample 1, which has a larger value of the conductivity below 75 K, has a better quality than the others. The deviation from thermal activation caused by another process can be better represented, if the activated conductivity $\sigma_a(T)$ with activation energy $E/k_B=530$ K is subtracted from the total conductivity $\sigma(T)$ ($\sigma_H(T)=\sigma(T)-\sigma_a(T)$). The dependence of $\log \sigma_H(T)$ of sample 1 can be approximated by the law $\sigma_H(T)=\sigma_0 \exp[-(A_0/T^{1/2})]$ with $A_0 \approx 118$ K in the temperature range between 30 K and 15 K. This corresponds to hopping conduction with variable length of the hopping distance for one dimensional conduction [4]. Further evidence is provided by the frequency dependence of conductivity at low temperature, which should follow the law $\sigma_{ac} \sim \omega^S$ [4]. Figure 2 shows the strong frequency dependence of

the real part of the conductivity for various temperatures. For the sample 1, the values were obtained of S is 0.82 at 14K and decreasing to 0.74 at 24 K.



(1)



(2)

Figure 1: Temperature dependence of the relative conductivity of the $K_{0.3}MoO_3$ of the sample 1 \circ and sample 3 ∇ . Insert shows the dependence of σ_H/σ_{300} below 30 K of the sample 1.

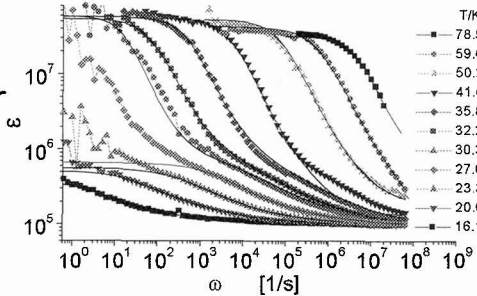
Figure 2: Frequency dependence of the real part of the conductivity below 80 K of the sample 1.

The real part and imaginary part of $\varepsilon(\omega)$ in a wide temperature range from 16 K to 80 K are shown in figure 3 and 4. The detailed analysis of relaxation processes were made by fitting the function:

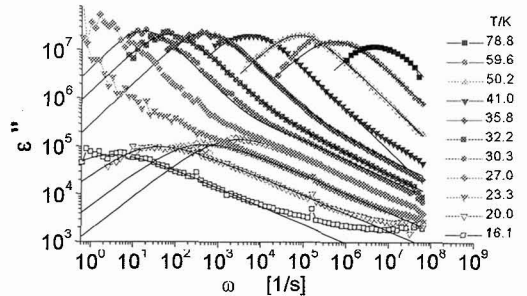
$$\varepsilon(\omega) = \varepsilon_{HF} + \frac{\varepsilon_0 - \varepsilon_{HF}}{[1 + (i\omega\tau_0)^{1-\alpha}]^\beta} \quad \text{with} \quad \varepsilon(\omega) = \frac{\sigma(\omega) - \sigma_{dc}}{i\varepsilon_0\omega}$$

where ε_{HF} , ε_0 are the high frequency and static dielectric constant and α and β describe the width and nonsymmetry of the distribution of relaxation times around τ_0 respectively.

The large value of ε reaches over 10^7 which reflects the extreme polarizability of the CDW. Below 80 K the peak of the pinned CDW is the most prominent feature of $K_{0.3}MoO_3$ and moves with decreasing temperature to lower frequency [5]. An additional process appears in the temperature range from 50 K to 16 K. While the first process moves out from the measured frequency range, the second one becomes dominant below 28 K.



(3)

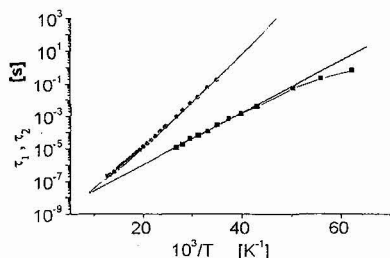


(4)

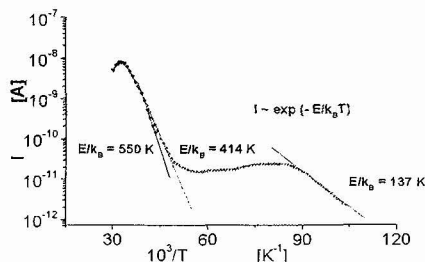
Figure 3 – 4: Frequency dependence of real and imaginary part of dielectric function below 80 K

We fitted the first and second process with $\beta=1$ (Cole-Cole-Function) and $\beta \leq 1$ (Havriliak- Negami-Function) respectively. Figure 5 presents the variation of $\log\tau_0$ against $10^3/T$ for the two processes, which were calculated from the fit. Both relaxation times display a deviation from Arrhenius behavior. The first process can be fitted by the Vogel-Flucher-law $\tau = \tau_0 \exp[E_0/k_B(T-T_0)]$ with the parameters $E_0 = 44.5$ meV (518 K), $T_0 = 4$ K and $\tau_0 = 1.84 \times 10^{-10}$ s and the critical slowing down equation $\tau = \tau_0 (1-T/T_c)^{-Z_v}$ with the parameters $T_c = 6.4$ K, $\tau_0 = 2.28 \times 10^{-10}$ s and $Z_v = 81$. The dependence of the second relaxation time has a different curvature with decreasing temperature and becomes flatter corresponding to the hopping mechanism. Both processes indicate in the high temperature region with their relaxation times a thermally

activated behavior by $\tau = \tau_0 \exp(E/k_B T)$ with $E/k_B \cong 600$ K close to the Peierls energy gap ($\Delta_{dc} = 530$ K) for the first process and $E/k_B \cong 421$ K for the second one.



(5)



(6)

Figure 5: Temperature dependence of the relaxation times of the two processes

Figure 6: Logarithm of TSD and partial TSD current in the presence of a field 8 V/cm and heating rate of about 10.5 K/min

4-Discussion

Below the Peierls transition temperature T_P thermally excited electrons contribute to the conductivity. The CDW is pinned by impurities, which are distributed randomly in the crystal. There are definite regions, between these regions the hopping occurs. The conductivity in $K_{0.3}MoO_3$ is dominated by the thermally excited electrons and has an activation energy $\Delta_{dc} = 530$ K. The classical CDW model predicts the same characteristic of the mean relaxation time and the normal conductivity in the same temperature dependence and the microscopic connection between the CDW relaxation and the CDW-induced band gap. The interesting result is the activation energy $E/k_B = 600$ K of the mean relaxation time of the CDW (the first process) at high temperature. However, this activation energy has a small difference from the dc band gap, which probably comes from the contribution of the hopping-effect. With decreasing temperature, the dominant thermally activated carries decrease rapidly and the pinned CDW becomes viscous with longer relaxation times. The conductivity is then dominated by the hopping process. It can be seen from the dielectric susceptibility experiments that the characteristics of the main relaxation time changes its thermal activation below 50 K as the static dielectric constant and goes to a critical slowing down behavior. Below ~ 28 K the main relaxation times are larger than our measurement conditions. The CDW is practically frozen out in the enormous metastable states with a long relaxation time. An evidence of this frozen polarization state is the maximum of the TSD current (figure 6) in this temperature range with an activation energy about 550 K. The second process leads to a TSD maximum at about 15 K.

5-Conclusion

We have investigated the dynamical features of $K_{0.3}MoO_3$ in a wide frequency and temperature range. A connection between the temperature dependence of dc and ac conductivity, with dielectric relaxation and TSD has been established. We have found that the hopping mechanism plays an important role at low temperature and there is a deviation from Arrhenius behavior of the relaxation of the CDW. From our result, the CDW shows the same features of the glassy state as the chalcogenide glass, which was reported by Jonscher [6]. The increase of the metastable states with decreasing temperature, the freezing of these polarization states of the CDW, the hopping conductivity and the very large relaxation times, which show a critical slowing down at low temperature, are characteristics of a transition to a glass-like state in systems with disorder.

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