Thermal hysteresis in low frequency dielectric response of charge density wave systems TaS_3 and $K_{0.3}MoO_3$

A. Akrap, D. Staresinic, K. Biljakovic, P. Lunkenheimer' and A. Loidl'

Institute of Physics, P.O. Box 304, 10001 Zagreb, Croafia
' University of Augsburg, Experimentalphysik V, Universitaetsstr. 2, 86135 Augsburg, Germany

Abstract. Low frequency dielectric response of charge density wave systems $K_{0.3}MoO_3$ and o-TaS_3 shows hysteresis on temperature cycling. The closing **of** the hysteresis at low temperature coincides in both systems with the closing of the hysteresis in DC conductivity and corresponds to the temperature of the glass transition observed in dielectric response of these two systems. AC conducitivity is higher on heating, while DC conductivity is lower, i.e. two loops have opposite directions. Higher AC conducitivity (or dielectric response) is a consequence of more corrugated CDW phase on heating.

For systems with charge density wave (CDW) ground state such as o-TaS_3 or $K_{0.3}MoO_3$ thermal hysteresis is typically observed in the temperature evolution of physical properties, such as DC conductivity [1-3], thermopower [4], thermal expansion [5] or IR transmission [6]. It indicates indirectly the differences of the configuration of the pinned CDW on cooling and heating, revealed due to the coupling of CDW with free carriers (DC conductivity, thermopower, IR transmission) or lattice (thermal expansion). On the other hand, the low frequency dielectric response of CDW systems has been shown to be a direct consequence of CDW dynamics, so we have considered it would be advantageous to study the hysteretic properties of CDW by this method.

Dielectric response of two typical CDW systems, $K_{0.3}MoO_3$ and o-TaS_3, has been measured in several temperature regions from room temperature down to 4.2 K. We have used impedance analysers HP4284 and HP4285, covering the frequency range from 25 Hz up to 30 MHz. The rate of temperature change has been varied from 0.1 Wmin up to 1 Wmin. Typical signal amplitudes have been 10-30 mV, verified at several temperatures to give linear response.

We have observed a large hysteresis in the low frequency dielectric response of both systems, as presented in Figure 1a) for o-TaS_3 and 1b) for $K_{0.3}MoO_3$. The hysteresis is entirely reproducible, and insensitive to the temperature sweep rate. The closing of the lowest frequency hysteretic loop at 50 K in o-TaS_3 and at 30 K in $K_{0.3}MoO_3$ coincides with the closing of hysteresis in the DC conductivities in these systems [1,3]. It also features all particularities observed in the DC conductivity hysteresis such as cooling and heating saturation curves, transient curves on reversal of temperature drift and slow ageing when the cooling or heating is interrupted [1]. However, the dielectric response (or AC conductivity) is higher on heating, and DC conducitivity lower, i.e. the direction of the hysteresis is inverted.

In Figures 1c) and 1d) the frequency dependence of dielectric response at selected temperatures is compared on heating and cooling. It shows that the hysteresis is a property of relaxational mode of CDW, which we call α process ([7,8] and references therein, also Starešinić *et al.* in these proceedings). Closing of the hysteretic loop occurs at temperatures of glass transitions T_g observed in the dielectric response of o-TaS_3 [7] and $K_{0.3}MoO_3$ [8], and it is related to the freezing of α process.

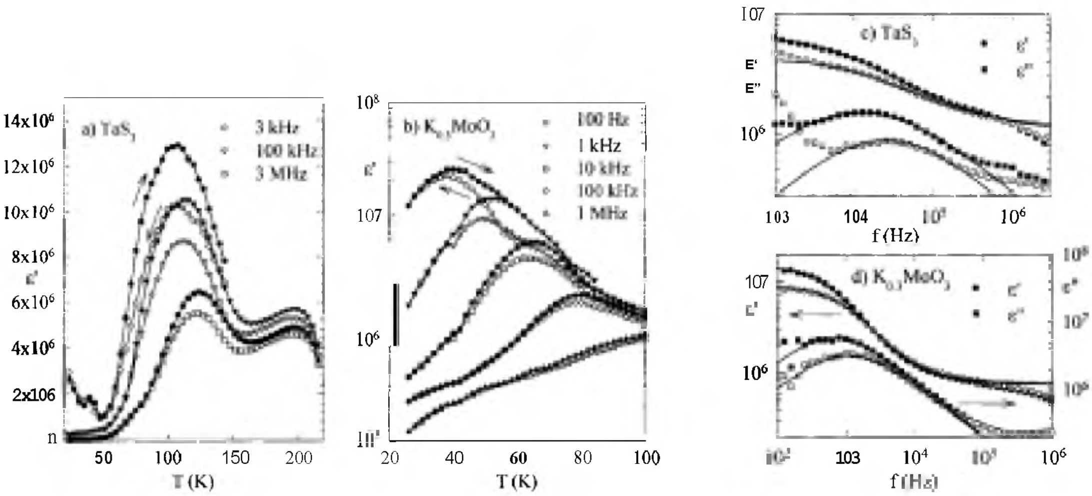

Figure 1. a) Hysteresis in the real part of low frequency dielectric response ε' of TaS_3 and b) $K_{0.3}MoO_3$ shown at several frequencies. c) Frequency dependences of real ε' and imaginary ε" part of the dielectric **response** on cooling and heating at **68** K for TaS_3 b) at 51 K for $K_{0.3}MoO_3$. Empty symbols **are** for cooling, and **filled** for heating.

Figures 1c) and 1d) show that the amplitude of α process is higher, and the relaxation time (inverse of the frequency of maximum in ε") lower on heating than on cooling. This remains true in the entire temperature region (68 K to 90 K for TaS_3, and 45 K to 80 K for $K_{0.3}MoO_3$) in which both parameters could have been evaluated. Higher amplitude and longer relaxation time correspond to the more corrugated CDW phase [9]. This is consistent with the hysteresis found in numerical simulation of CDW adjustment to the changes of equilibrium CDW wave wector Q_{CDW} [10] through phase slip. The microsopic model for the case of temperature dependent Q_{CDW} is found in [11].

Finally, the fact that the hysteresis in DC resistivity follows the hysteresis in the dielectric response can be understood if we suppose that at least a part of free carriers scattering of is on deformed CDW. In this case more corrugated CDW phase would lead to higher scattering rates and higher restivity.

In conclusion, by measuring low frequency dielectric response of two CDW systems we have directly shown that the thermal hysteresis is a property of the CDW condensate itself, or more precisely its low frequency relaxational, or α process. It is most probably related to the hysteresis in CDW phase configuration coming from adjustments to the temperature dependent CDW wave-vector. The hysteresis in DC resistivity seems to be simply a consequence of free carriers scattering on deformed CDW.

References

[1] Higgs W. and Gill J.C., *Solid State Commun.* **47** (1983) 737

[2] Wang Z.Z. and Ong N.P., *Phys. Rev. B* **34** (1986) 5967

[3] Xue-mei W. and Dian-lin Z., *Phys. Rev. B* **54** (1996) 1443

[4] Higgs **A.W.** in Charge Density Waves in Solids, edited by Hutiray Gy., Solyom J. (Springer-Verlag, Berlin, West Germany, 1985) pp.422-425

[5] Golovnya A.V., Pokrovskii V.Ya., and Shadrin P.M., *Phys. Rev. Lett.* **88** (2002) 246401

[6] Itkis M.E., Emerling B.M. and Brill J.W., *Phys. Rev. B* **56** (1997) 6506

[7] Starešinić D. *et al., Phys. Rev. B* **65** (2002) 165109

[8] Hosseini K. *et al., J. Phys.IV, France* **9** (1999) Pr10-41

[9] Littlewood P.B., *Phys. Rev. B* **33** (1986) 6694; *Phys. Rev. B* **36** (1987) 3108

[10] Littlewood P.B. and Rice T.M., *Phys. Rev. Lett.* **48** (1982) 44

[11] Pokrovskii V.Ya. and Zaitsev-Zotov S.V., *Synth. Metals* **32** (1989) 321