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Modalities of self-organized charge response in low dimensional systems

S. Tomic[1], T. Vuletic[1], M. Pinteric[1,2] and B. Korin-Hamzic[1]

[1] *Institut za Fiziku, P.O. Box 304, 10001 Zagreb, Croatia*
[2] *Faculty of Civil Engineering, University of Maribor, Maribor, Slovenia*

Abstract. We present modalities of self-organized charge response in low dimensional systems, like diverse organic and quantum spin systems, studied by the low-frequency (10 mHz – 1 MHz) dielectric spectroscopy. Density wave structures with the order of commensurability N ≈ 4 can be recognized as phasons in a random impurity potential, whereas those with N ≈ 3 can be viewed as topological defects like charge domain wall pairs in the background domain structure.

1. INTRODUCTION

Density wave (DW) is a complex, deformable object, which exhibits a self-organized response to externally applied fields. Density waves, mostly studied so far are incommensurate (IC) with respect to the underlying lattice with the order of commensurability N close to 4. The spatial variation of the DW phase in an IC structure is referred to as a phason. **A** dynamical mode associated with this DW variation, also referred to as the phason, is theoretically gapless long wavelength excitation. However, in the real crystals there is a small gap in the phason spectrum due to defects. The phason mode, as long as free carrier excitations are present, couples to an applied dc and ac electric field and gives novel contributions to the electrical conductivity. The coupling to the former gives rise to the non-linear conduction of electrical current above a finite electric field accompanied by narrow and broad band noise, whereas the coupling to the latter leads to a frequency dependent conductivity [1]. Theoretically, transverse phason mode couples to electromagnetic radiation and yields a narrow absorption close to the pinning frequency, whereas longitudinal screened mode results in a broad low frequency relaxation at frequencies typically lower than 1MHz. The latter is observed due to inherent randomness of density waves. This feature results also in that the observed absorptions are neither purely longitudinal nor transverse [2].

In this paper we summarize modalities of DW dielectric response observed in diverse systems. Section 2 describes the well established phason response for N ≈ 4 DW whose randomness is due to inhomogeneously distributed positions of impurities in the real crystals. Section 3 presents results that show the charge domain wall relaxation for commensurate DW whose randomness is associated with domain structure of the ground state. **A** DW relaxation observed recently in a low-dimensional quantum spin system with chains and ladders is presented in Section 4.

2. PHASON RELAXATION IN RANDOM IMPURITY POTENTIAL

The response of N ≈ 4 DW is characterized by dielectric constants of the order 10^6-10^9, and is broader than the Debye one, which is expected for the system with a single degree of freedom [3]. This feature reflects a distribution of relaxation times associated with a single process due to a distribution of metastable states around the equilibrium position. These metastable states correspond to local changes of the phase of the pinned DW in a random impurity potential. Dielectric relaxation is strongly influenced by the free-carrier screening so that the response gradually slows down with temperature, with the activation energy equal to the single-particle activation energy. **As** an example, we show in Fig.1 and 2 dielectric relaxation observed in N ≈ 4 SDW, established below metal-semiconductor phase transition at T_C = 12 K in $(TMSTF)_2PF_6$ [4]. The observed behaviour indicates that SDW responds to the outer ac perturbation field by its long-wavelength phason excitations, and that the interaction with free carriers yields the dominant dissipation. In order to get an insight what happens when there is not enough

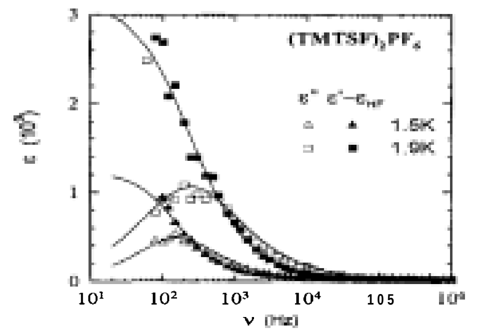


Figure 1. Frequency dependence of the real and imaginary parts of the dielectric function at two selected temperatures.

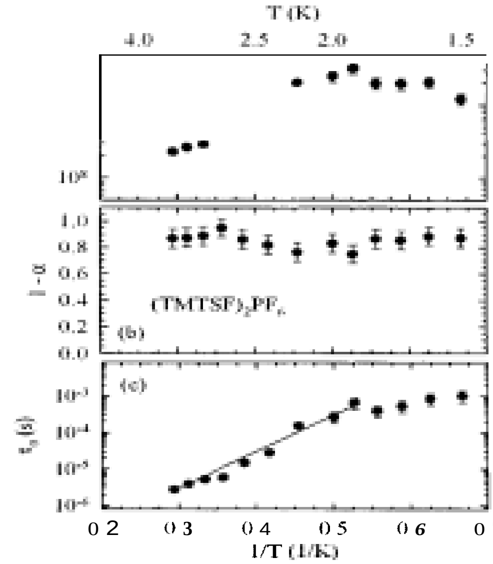
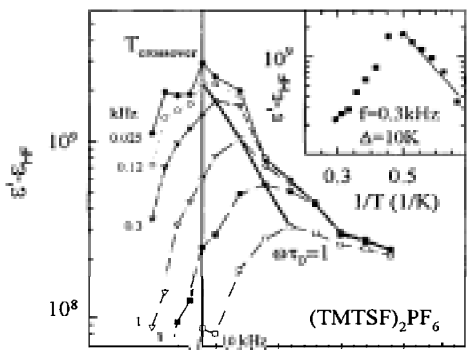


Figure 2. (a) Relaxation strength ($\Delta\varepsilon$), (b) shape parameter (1-a) and (c) mean relaxation time (τ) versus inverse temperature.



free carriers to screen, we present Fig.3. At temperatures $T < T_{CO} \approx 2$ K the free carrier screening becomes gradually ineffective since the free carrier density becomes smaller than one electron per the phason characteristic length. The latter scales inversely with dc threshold field and is estimated to be about $L_{ph} \approx 25$ µm, whereas an effective one-electron length can be deduced taking into account the resistance rise of about 4 orders of magnitude below ~12 K. At these low temperatures the dynamics of DW system starts to be governed by low energy barriers $A \approx 10$ K $< \Delta_{fc}$. It is important to note that the position of T_{CO} depends on sample's purity and shifts to lower temperatures for higher impurity levels. We also point out that this consideration is only possible to make below $\omega\tau_0 = 1$ line, where DW ceases to be able to follow the ac perturbation and consequently dielectric function decreases. Another dielectric study claims a glassy transition at 2 K and explains barriers of about 10 K to be due to solitons [5]. We note that the existence of a subtle structure inside the SDW phase and its possible effects on the dielectric response in (TMTSF)$_2$PF$_6$ [6] are beyond the scope of this paper.

3. CHARGE DOMAIN WALL RELAXATION IN DOMAIN STRUCTURE

Theoretically, a commensurate structure limits the spatial variation of DW to rather narrow localized regions, which are topologically equivalent to solitons in 1D or domain walls in 2D and 3D. For $N \geq 3$ DW has a complex order parameter and, in contrast to IC DW, there are well defined, energetically most favourable, positions in the underlying lattice at which it is pinned. Consequently, the pinning energy depends on the phase of the order parameter that determines the relative position of the DW and the lattice. The importance of this term, i.e. the dependence of the pinning energy on phase, strongly diminishes with increasing N [7]. Experimentally, the impurity pinning, i.e. the pinning of DW to randomly distributed impurities, appears to be dominant pinning mechanism for $N \equiv 4$ DW, which usually prevails over the commensurability one (see Section 2). In this section, we show that commensurate DW whose randomness is associated with domain structure of the ground state are

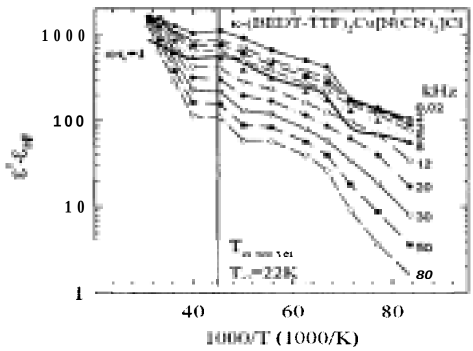


Figure 4. Real part of the dielectric function versus temperature at a few selected frequencies

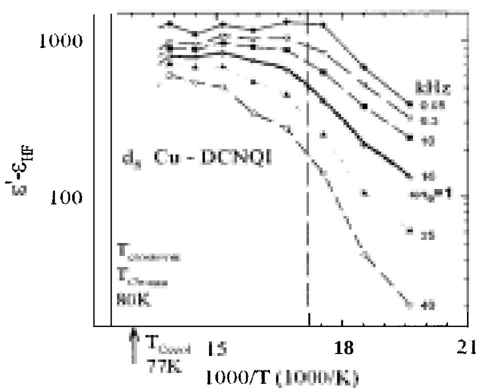


Figure 6. .eal part of the dielectric function versus temperatui at a few selected frequencies.

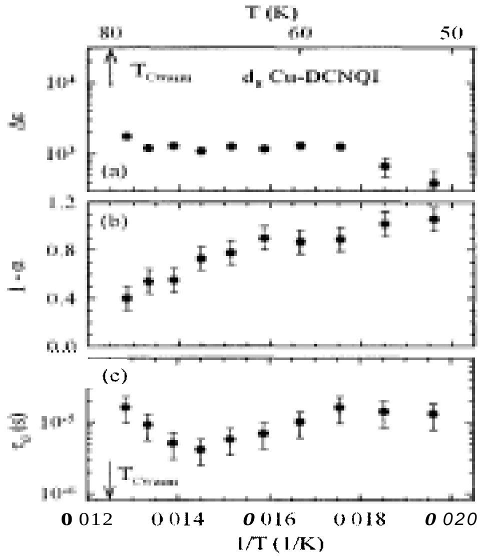


Figure 5. *(a)* Relaxation strength (Δε), (b) shape parameter (1-a) and (c) mean relaxation time (τ) versus inverse temperature.

not able to use phason excitations to respond to outer ac fields. These excitations are frozen and replaced by charge domain wall pairs, which are short wavelength excitations. First example is dielectric response of the domain structure in weak ferromagnetic state of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, which is basically commensurate SDW with canted spins (N is not known). The magnetic phase transition is at $T_C = 22$ K, where charges are already rather well localized. Therefore, antiferromagnetic (AF) order is of localized spins in contrast to AF order of itinerant spins in (TMTSF)$_2$PF$_6$ (Section *2)*. The charge response is characterized by dielectric constant Δε of about 1000, broad relaxation time distribution, whose width is characterized by parameter (1-a), that significantly narrows at low temperatures and temperature independent mean relaxation time τ_0 [8]. T_{CO} is estimated to be close to T_C, so that in the whole T range below T_C free carrier screening is substantially reduced. At $T > T_C \approx T_{CO}$ and $T < T_C \approx T_{CO}$ DW dynamics is governed by $\Delta_{fc} \approx 150$ K and by low energy barriers $A \approx 50$ K $< \Delta_{fc}$, respectively (Fig 4). Qualitatively same kind of response is used by domain structure of N = 3 CDW in the deuterated Cu-DCNQI d_8 system shown in Fig.5 [9]. The first order metal-insulator phase transition in the warming cycle is at $T_C = 80$ K. Random domain structure is due to a coexistence of CDW insulating and metallic domains in a broad hysteretic region. This random pattern gradually evolves into a regular one yielding a narrow, Debye like, response at low temperatures. In addition to the relaxation time distribution, the characteristic energy also depends on the domain pattern. The response of the random domain structure is broad and governed by low energy barriers $A \approx 50$ K, whereas the response of the regular domain structure is narrow and governed by high energy close to the free carrier one $\Delta_{fc} \approx 500$ K (Fig.6). In the former pattern, estimated $L_{dw} \approx 0.1 - 1$ μm is much smaller than the one-electron length of about 1 mm, explaining clearly inefficiency of the free carrier screening. In the regular low temperature pattern, where N=3 DW is established in the whole bulk, much smaller L_{dw} should be expected.

4. COLLECTIVE CHARGE RELAXATION IN Cu-0 BASED CHAIN-LADDER SYSTEM

The low dimensional quantum spin system $Sr_{14}Cu_{24}O_{41}$ contains interpenetrating subsystems of CuO_2 chains and Cu_2O_3 two-leg ladders. The nominal valence of Cu is + 2.25. Optical studies indicate that holes are strongly localized at chains, whereas the self-doping of 0.07 holes/ladder Cu site indicates that the system is not a genuine insulator. Our DC resistivity data indicate a phase transition at $T_C = 210$ K and activated behaviour with $\Delta_{fc} \approx 1300$ K below [10]. In spite an extensive research, the nature of the insulating state, as well as the very existence of phase transition, has remained elusive.

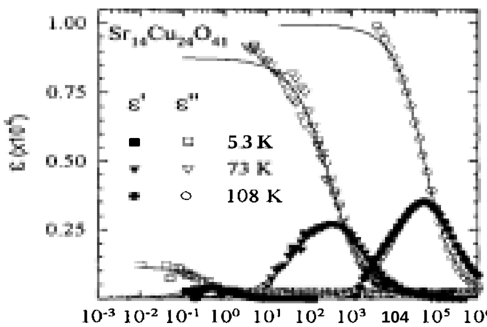


Figure **7.** Frequency dependence of the real and imaginary parts of the dielectric function at three selected temperatures.

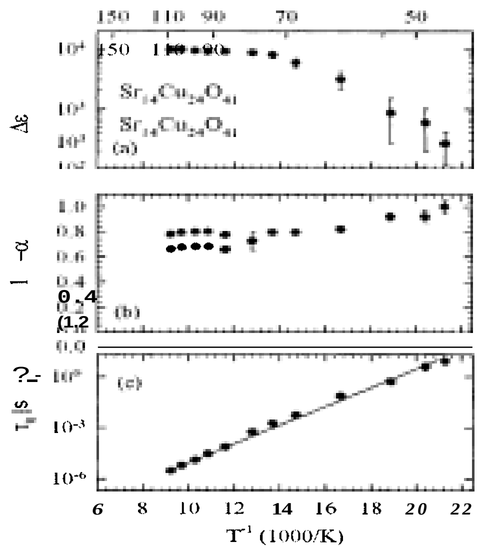


Figure **8.** (a) Relaxation strength (Δε), (b) shape parameter $(1-\alpha)$ and (c) mean relaxation time (τ) versus inverse temperatures.

Dielectric response is displayed in Fig.7 and 8. The relaxation is characterized by dielectric constant of the order 10^4, gradually slows down with temperature with the activation energy equal to Δ_{fc} and relaxation time distribution is broad [10, 11]. These features indicate that the observed response might be attributed to the phason relaxation in a random impurity potential with the crucial influence of free carrier screening on dissipation. Consequently, we are lead to conclude that the insulating state below $T_C = 210$ K is charge density wave state established in the ladder subsystem. However, a substantial reduction of Δε and an important narrowing of relaxation time distribution at low temperatures, together with no finite threshold field for dc non-linearity indicate that a standard picture as described in Section 2 cannot be fully applied in this case. **A** full account of this work that incorporates the whole series $Sr_{14-x}Ca_xCu_{24}O_{41}$ will be published elsewhere [11].

References

[1] G.Grüner, *Rev.Modern Physics* **60** (1988) 1129-1182 and **66** (1994) 1-24.

[2] P.B.Littlewood, *Phys.Rev.**B*** **36** (1987) 3108-3116.

[3] R.J.Cava, R.M.Fleming, R.G.Dunn and E.A.Rietman, *Phys.Rev.**B*** **30** (1984) 3228-3239.

[4] S. Tomić, N.Biškup and A. Omerzu, *Synth. Metals* **85** (1997) 1597-1598.

[5] F.Nad', P.Monceau and K.Bechgaard, ***Solid State** Commun.* ***95*** (1995) 655-660.

[6] T.Vuletić, D.Herman, N.Biškup, M.Pinterić, A.Omerzu, S.Tomić and M.Nagasawa, *J.Phys.IV France* **9,** Pr10 (1999) 275-277 and references there-in.

[7] P.A.Lee, T.M.Rice and P.W.Anderson, *Solid State **Commun.*****14** (1974) 703-709.

[8] M.Pinterić, M.Miljak, N.Biškup, O.Milat, I.Aviani, S.Tomić, D.Schweitzer, W.Strunz and I.Heinen, *Eur.Phys.J.**B*** **11** (1999) 217-225.

[9] M.Pinterić, T.Vuletić, M.Lončarić, S.Tomić and J.U.von Schütz, *Eur.Phys.J.**B*** **16** (2000) 487-493.

[10] B.Gorshunov, P.Haas, T.Room, M.Dressel, T.Vuletić, B.Korin-Hamzić, S.Tomić, J.Akimitsu and T.Nagata, submitted to *Phys.Rev.**B**.* (2002) and cond-mat/0201413 and references there-in.

[11] T.Vuletić et al, in preparation.