Single-Particle And Spin-Density Wave Charge Dynamics In $(TMTSF)_2PF_6$ And $(TMTSF)_2AsF_6$: A Comparative Overview

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Abstract. We present the results of DC and AC (100 mHz – 1MHz) electrical transport measurements in low and high electric fields performed in the spin-density wave (SDW) state of the Bechagaards salts $(TMTSF)_2PF_6$ and $(TMTSF)_2AsF_6$. We argue that a degree of complex structure of the SDW ground state which is unfolded in a particular experiment depends strongly on the chosen experimental probe and the crystal measured.

Twelve years ago, the NMR measurements of the spin-lattice relaxation rate ($1/T_1$) [1] have shown a changeover at 3.5 K from a temperature independent behaviour at higher temperatures to an activated behaviour at lower temperatures, with an activation energy lower than the free carrier resistivity one. In addition, another anomaly in $1/T_1$ was detected at about 2 K. Takahashi et al. have suggested that the $1/T_1$ anomalies indicate the existence of successive transitions inside the SDW ground state, which divide the SDW phase into three subphases. Further, a calorimetric transition at 3.5 K with large hysteretic phenomena in the temperature range 2.5 K – 4 K caused by the sample history has been observed [2]. In addition, the same authors have suggested that the dielectric relaxation follows the critical slowing down behaviour indicating a glass transition around 2 K. Clark et al. [3] have reported that the temperature of the anomaly of the spin-lattice relaxation rate also depends on measuring frequency. However, they find that the temperature shift follows Arrhenius behaviour with free carrier activation energy. The latter result contradicts the critical slowing down suggested on the basis of the dielectric relaxation. The authors have pointed out that the phason fluctuations are responsible for the $1/T_1$ relaxation and that the observed NMR behaviour might result from a gradual slowing down of the phason fluctuations. Hence, no need for the existence of a glass transition is invoked. Since then an impressive number of publications have been dedicated to this subject and we apologise to all the authors whom we do not mention here due to the lack of space. Nevertheless, the above differences have not been clarified yet, leaving open the intriguing question of a complex nature and the possible existence of a glass transition inside the SDW phase of $(TMTSF)_2PF_6$.

The purpose of this communication is to briefly summarize and discuss results of the experiments we have performed during the last four years in order to understand the complex nature of the SDW ground state [4]. In particular, we address the following issues:

(a) A change in slope in the log R vs. $1/T$ plots at about 2.5 K indicating changes in the activation energy of the single-particle conduction channel. It should be noted that this feature is also visible in the same kind of plots obtained 9 years ago [5], but the authors were not aware of it at that time. Recent magnetoresistance results for the current direction along the intermediate axis by B. Korin-Hamzić et al. [6] were the first to reveal the change in the activation energy at about 4 K, which depends on magnetic field. In Fig. 1 we show the results obtained on 6 single crystals from different sources. Symbols KB S and KB P which stand for the PF_6 crystals describe the samples from the same batches which were studied and reported in [2]. Symbol KB stands for the PF_6 crystal prepared by K. Bechgaard on which the nonlinear transport and dielectric function measurements were also performed (see Figs. 3(a) and 4). Symbols MN1 (reported in [1(c)]) and MN2 describe the AsF_6 samples from the same batch prepared by M. Nagasawa on which nonlinear transport and dielectric response were also studied (see Figs. 2, 3(b), 5). The measured crystals had very different resistivity ratios and different numbers of cracks which caused a different amount of the total increase of resistance on cooling (ranging from zero to several hundred percent). The latter has influence on the absolute value of the activation energy, nevertheless a break in the slope is always present.

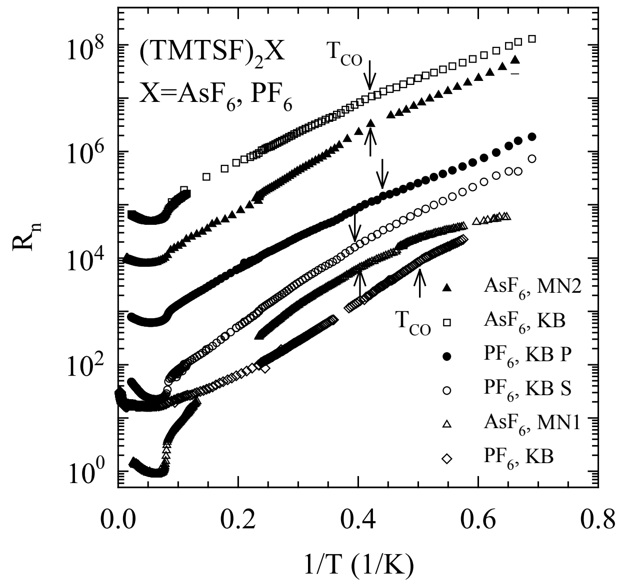

Figure 1: Normalised resistance vs. inverse temperature. Curves are shifted for clarity. Note a break in the slope at about 2.5 K.

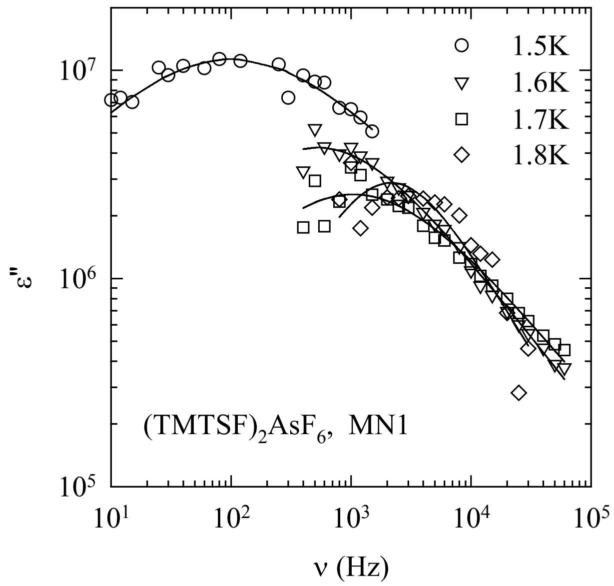

Figure 2: Imaginary part of the dielectric function vs. frequency.

(b) Dependence of the threshold field (E_T) and of the collective conductivity on temperature below 4 K, the former showing a maximum in the temperature range 3 K - 1.6 K (Figs. 3(a), 3(b)). The shape of E_T vs. T curve, that is the position and the width of the maximum is sample (batch) dependent. It should be noted that while this maximum is clearly visible for all the studied AsF_6 samples, no firm evidence is obtained down to 1.5 K for the PF_6 samples with dominant impurity pinning (KB S and KB). Only PF_6 crystals, in which the commensurability pinning prevails over the impurity one (KB P), show a similar temperature behaviour of E_T as the AsF_6 ones.

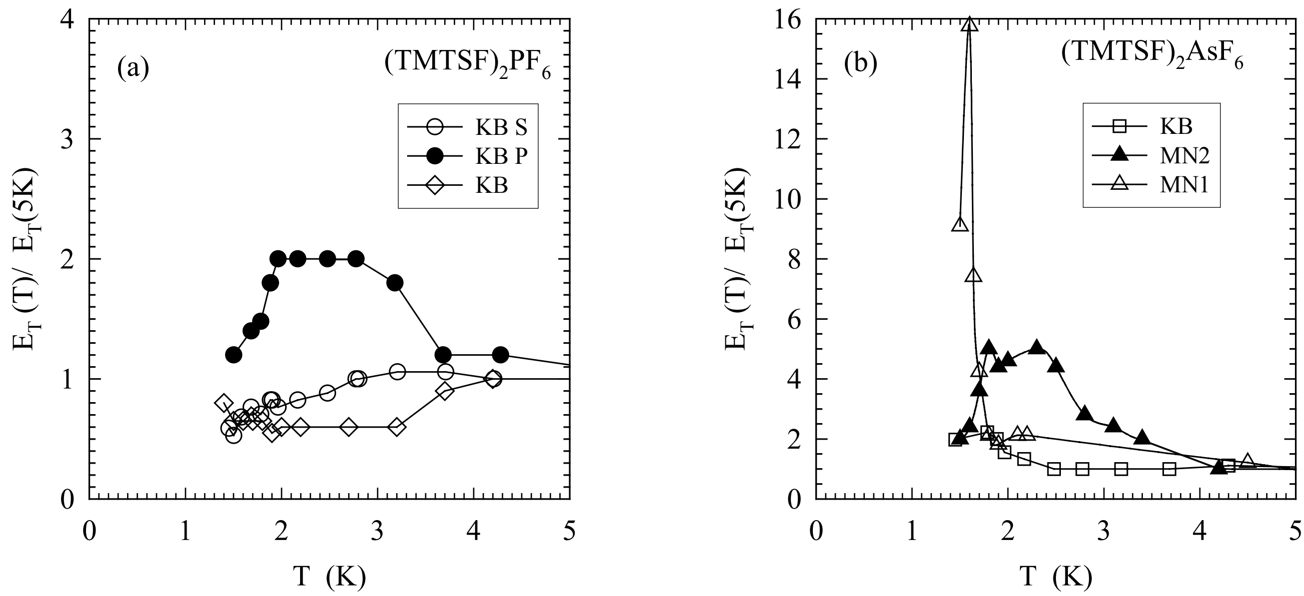

Figure 3: Normalised threshold electric field vs. temperature for (a) three PF_6 crystals (P: commensurability pinning; S: weak impurity pinning) and (b) three AsF_6 crystals (MN1: critical slowing down; MN2: gradual slowing down).

(c) The low-frequency dielectric relaxation below 5 K is broader than the Debye ($\alpha = 0.8$) and for the 5 PF_6 samples with dominant impurity pinning is thermally activated in a manner similar to the single-particle conductivity (Fig.4). T_{CO} found in DC resistivity curves is also visible in (1 - α) vs. $1/T$ behaviour for these samples, while it is clearly identified for only one AsF_6 sample (MN2) in log τ vs. $1/T$ curves (Fig. 5). The crossover to an almost saturated behaviour below 2 K is observed for the two PF_6 samples. Only for the one AsF_6 single crystal (denoted as MN1) the critical slowing down, accompanied by broadening of the response, was observed below 1.7 K (Figs. 2, 5). In addition, it should be noted that the magnitude and the temperature dependence of the relaxation strength is also sample dependent (Figs. 4 and 5).

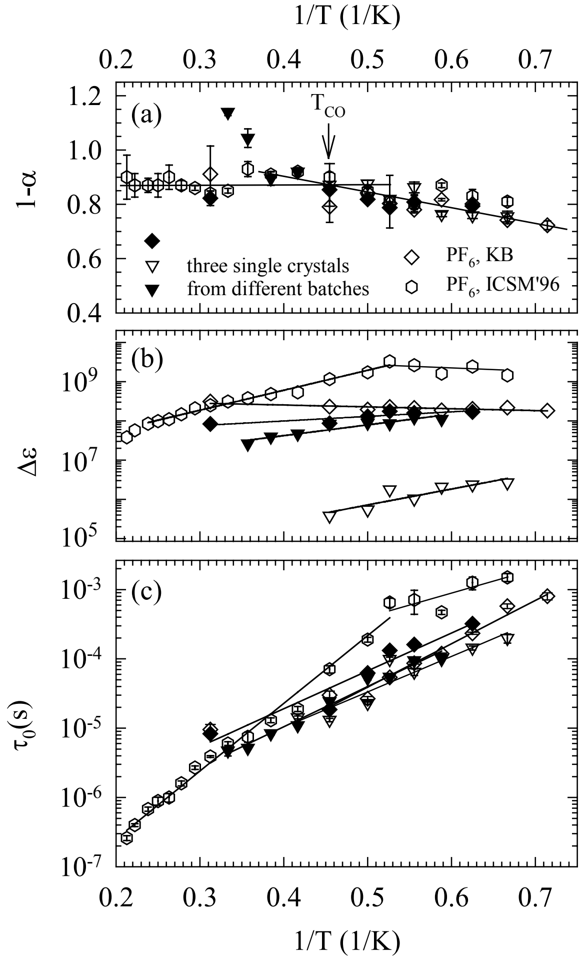

Figure 4: (a) Shape parameter, (b) relaxation strength and (c) relaxation time vs. inverse temperature for five PF_6 crystals.

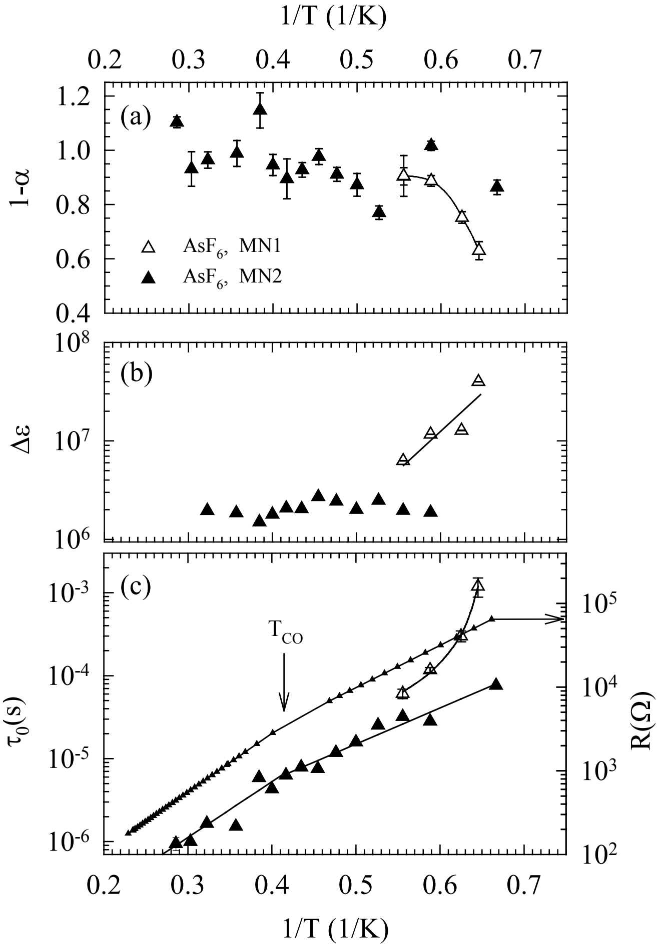

Figure 5: (a) Shape parameter, (b) relaxation strength and (c) relaxation time vs. inverse temperature for two AsF_6 crystals.

In conclusion, we have shown that the features of both the collective conduction channel in the SDW ground state and the SDW low-frequency dielectric response are sample (batch) dependent. In the PF_6 samples in which the SDW is pinned by impurities the electric threshold field which characterizes the sliding mechanism gradually weakens with temperature, concomitantly as the SDW dielectric relaxation gradually slows down. In contrast, in the PF_6 and AsF_6 samples in which the commensurability pinning prevails [5, 7], E_T displays the maximum below 3 K, and a critical slowing down of the SDW dielectric relaxation might be observed. It should be noted that the latter behaviour was only observed in the sample in which an extremely narrow E_T peak was found. Keeping in mind that the SDW wave vector is found to be very close to commensurability [8], we propose that subtle variations of the disorder level in nominally pure samples might be responsible for the different behaviours observed.

References

1. T. Takahashi, Y. Maniwa, H. Kawamura, K. Murata and G. Saito, Synth. Metals **19**, 225 (1987).
2. J. C. Lasjaunias, K. Biljaković, F. Nad', P. Monceau and K. Bechgaard, Phys. Rev. Lett.**72**, 1283 (1994).
3. W. G. Clark, M. E. Hanson, W. H. Wong and B. Alavi, J. de Physique IV, C2, **3**, 235 (1993).
4. (a) N. Biškup, Ph. D. Thesis, University of Zagreb, unpublished (1996); (b) S. Tomić, N.Biškup and A. Omerzu, Synth. Metals **85**, 1597 (1997) and (c) N. Biškup, T. Vuletić, D. Herman, S. Tomić, M. Nagasawa and K. Bechgaard, Proceedings of ICSM'98.
5. S. Tomić, J. R. Cooper, W. Kang, D. Jerome and K. Maki, J. Phys. I France **1**. 1603 (1991).
6. B. Korin-Hamzić, M. Basletić and K. Maki, Europhys. Lett. **43**, 450 (1998) and B. Korin-Hamzić et al., These Proceedings.
7. M. Nagasawa, T. Sambongi, K. Nomura and H. Anzai, Solid State Commun. **93**, 33 (1995).
8. T. Takahashi, Y. Maniwa, H. Kawamura and G. Saito, J. Phys. Soc. Jpn. **55**, 1364 (1986).