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Low frequency dielectric spectroscopy of the Peierls-Mott insulating state in the deuterated copper-DCNQI systems

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Abstract. We report a detailed characterization of an unique 3-fold commensurate insulating state in single crystals of the organic π - inorganic d hybrid $Cu(DMe\text{-}DCNQI)_2$ systems with deuterated and partially deuterated DCNQI ring, by means of low-frequency dielectric spectroscopy. A broad relaxation mode of strength $\Delta\varepsilon \approx 10^3 - 10^4$ centred at $\nu_0 \approx 1 - 10$ kHz is observed in the hysteresis temperature region in which the insulating phase coexists with metallic islands. At lower temperatures, outside the nucleation range, the relaxation narrows, approaching a Debye-like form for an overdamped response of a system with a single degree of freedom. Both, the relaxation strength and the mean relaxation time ($\tau_0 = 1/2\pi\nu_0$) are much larger than that expected for single-particle excitations. These features suggest the origin of the dielectric relaxation as an intrinsic property of the $N = 3$ charge density wave state.

PACS. 72.15.Nj Collective modes, low dimensional conductors – 71.45.Lr Charge density wave systems – 71.30.+h Metal-insulator transitions and other electronic transitions – 77.22.Gm Dielectric loss and relaxation

1 Introduction

Since 1987 an important amount of research has been devoted to a novel class of charge-transfer salts $(R_1R_2\text{-}DCNQI)_2Cu$ (where DCNQI stands for dicyanoquinonediimine and R_1, R_2 = CH_3, CH_3O, Cl, Br, I etc.), due to the possibility to control easily, by varying external (pressure, magnetic field) and internal (isotope substitution, doping) parameters, unique physical properties inside an extremely rich phase diagram [1–3]. In particular, a partial or total deuteration of DMe-DCNQI molecules has been shown to be equivalent to the application of a small pressure above which a metal-insulator transition occurs [3]. Compared with conventional organic metals, the novel electronic states have emerged associated with the hybridization between π- orbitals of DCNQI molecule and d-orbitals of Cu ions. According to XPS and infrared measurements [4,5], an averaged intermediate valence of Cu being somewhat smaller than 1.3 gives the very high room temperature (RT) conductivity of the order of 1000 Scm^{-1} and stabilizes a metallic state down to very low temperatures. The three-dimensional (3D) conduction network is achieved through mutually interconnected DCNQI chains via tetrahedrally coordinated Cu ions. The amount of distortion of the coordination tetrahedron is intimately related to an extra charge transfer from Cu to DCNQI sites and, consequently, to the nature of the low-temperature state. A simple estimate of the crystal-field splitting energy for Cu 3d-orbitals in the tetrahedral field formed by NC ligands has shown that deviations from the ideal tetrahedral symmetry are rather large already at RT in all Cu compounds, and much more important than additional distortion gained on lowering the temperature [6]. Then, an extremely small enhancement of the tetrahedron distortion is needed to lock the Cu valence to the 4/3 value and to switch the system into a highly insulating state.

Such a scenario has been recently confirmed through Cu 2p core-level photoemission measurements by Tanaka et al. [7]. The line-shape analysis have revealed 27% and 32% of Cu^{2+} ions at RT in the metallic DMe-Cu with no phase transition and in MeBr-Cu with M-I transition at 160 K, respectively. Further, at the M-I phase transition temperature, the lattice parameter c is shortened and the coordination angle N-Cu-N enlarged by a few percent as shown by crystal structure [8] and interferometric crystal length [9] determinations data. An insulating state is characterized by formation of the three-fold (N = 3) superstructure [10,11] which is associated with the lattice distortion and a periodic ordering of Cu ions. That is, the intensity of observed satellite superstructure spots is too strong to be due only to the copper ordering [12]. The former would imply the establishment of a charge-density wave (CDW) on DCNQI chains. However, it should be noted that no 1D precursor diffuse scattering has been

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observed as one should expect for a traditional Peierls instability. The latter is confirmed by the observation of a Curie-Weiss type of susceptibility [5] and recent ^{13}C NMR study by Kawamoto et al. [13]. The reported data have shown that the spin is localized exclusively at 1/3 of the Cu sites and have provided final evidence that the charge transfer $(1+\rho)$ is locked at the critical value $\rho = 1/3$ in the insulating phase.

The metal-insulator phase transition has been also studied theoretically by several authors [14–17]. Recent most complete theoretical model, proposed by Ogawa et al. [17], has shown that, in addition to electron-phonon (g) and electron-electron interactions, a 3D coupling is a necessary ingredient which has to be taken into account in order to fully describe the experimentally observed phase diagram. The metal-insulator phase transition is explained as a result of the Peierls transition with three-fold lattice distortion in the presence of strongly correlated Cu d-states. It is worth of noting that effective on-site Coulomb correlation energies between valence electrons, measured by Auger spectroscopy [7], have been found to be very high on both, carbon, as well as on copper sites, attaining values of 6.5 eV and 8 eV, respectively. The large gap at the wave vector $k = \pi n/3$ (n = 1, 2) is triggered by electron-phonon Peierls interaction only if the magnitude of g attains a value larger than a critical one. The existence of the critical value of g, which scales with pressure, is a consequence of the strong correlation which leads to the competition between the energy gain of the Peierls and the hybridization gap. The π - d hybridization is disconnected and the localized half-filled level is located at the chemical potential. The latter explains the discontinuous nature of the phase transition followed by large hysteresis effects.

On the basis of these results, it is generally accepted that in the insulating state a $N = 3$ charge density wave exists in DCNQI network together with the charge order of the same periodicity on Cu sites. However, the collective mode properties of the charge-density wave in this state are still unknown. Density waves (DW), both CDW and spin-density wave (SDW), mostly studied so far are incommensurate (IC) with respect to the underlying lattice. For IC DW it is now well established that below their respective phase transitions, as long as free-carrier excitations are present, DW condensate can slide above a finite threshold field (E_T) contributing so to the electrical conductivity [18]. Below E_T DW cannot move due to the pinning to randomly distributed defects in the underlying lattice. The dielectric response of the IC DW is broader than the Debye one which is expected for the system with a single degree of freedom. 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 the random defect potential. Both the collective DC conduction and dielectric relaxation are strongly influenced by the free-carrier screening so that the response of IC DW gradually slows down with temperature.

On the other hand, not much is known about the collective charge dynamics of commensurate (C) DW. Theoretically, one should expect the Debye form of the dielectric relaxation, and a large threshold field with a little contribution to the collective conduction which scales with the order of commensurability N. Indeed, our recent low-frequency dielectric measurements in the low-temperature state of DMeO-Li have identified, for the first time, a Debye-like relaxation with Arrhenius-like decay determined by the resistive dissipation of the $N = 4$ CDW mode pinned by commensurability [19]. On the other hand, threshold fields and non-linearity observed in current-voltage characteristics have not been found to differ appreciably from the ones usually encountered for IC DW close to $N = 4$ commensurability [20]. Finally, it is worth of mentioning two very recent publications which address the low-frequency dielectric relaxation in two distinct commensurate phases of organic π- conductors with strong charge localization. The first one concerns $N = 4$ short range CDW and $N = 4$ long range ordered SDW in $(TMTTF)_2Br$ [21]. The second one deals with a weak ferromagnetic phase in κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ [22]. An interesting aspect of the latter work is in that it addresses the dielectric relaxation of the collective excitation like charge-domain wall at the crossover between thermally activated decay at high temperatures and quantum tunneling at low temperatures where the resistive dissipation ceases to be effective.

In order to deepen the characterization of the commensurate $N = 3$ novel electronic state and to verify the proposed existence of $N = 3$ charge density wave in DCNQI-Cu materials, we have performed measurements of the low-frequency dielectric response. We have investigated two systems: (a) the partially deuterated system with a small amount of d_6 in h_8 (d_6 is the system where all of the hydrogen in the methyl groups are replaced by deuterium), that is the alloy $Cu[(2,5(CH_3)_2\text{-DCNQI})_{0.70}(2,5(CD_3)_2\text{-DCNQI})_{0.30}]_2$ (abbreviated as h_8/d_6 70%:30%) and (b) the fully deuterated system $Cu[(2,5(CD_3)_2\text{-DCNQI}]$ (abbreviated as d_8) where additionally all of the hydrogens in the DCNQI ring are replaced by deuterium. The observed dielectric relaxation data give clear evidence of a collective mode contribution to the dielectric properties which might be attributed to DW excitations like phase-vortex pairs and charge domain walls in a commensurate density wave structure.

2 Experimental and results

The measurements were performed on single crystals with varying lengths of about 2 mm and cross sections of typically 0.01 mm^2. All measured samples exhibited qualitatively the same behaviour. The RT conductivity (σ_{RT}) was in the range 800 - 1200 Scm^{-1}. The DC resistance measurements were performed using a standard DC technique or Keithley 617 electrometer in V-I mode. The observed resistance *versus* temperature curves have reproduced the previous ones in the literature [23,24]. The complex conductance ($G(\omega)$, $B(\omega)$) was measured by a

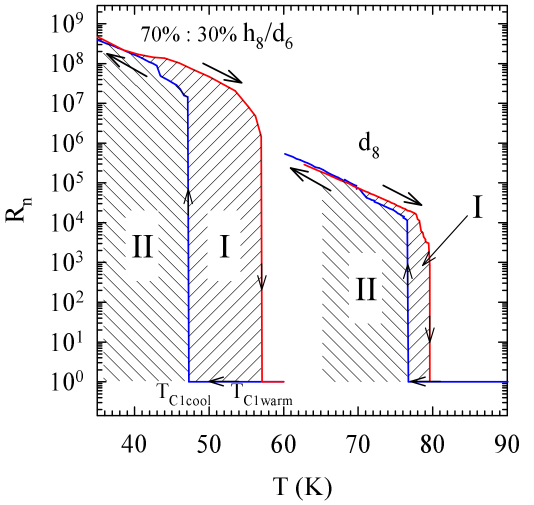

Fig. 1. Temperature dependence of the normalized resistance ($R_\mathrm{n} = R(T)/R(100\ \mathrm{K})$, where $1/R(100\ \mathrm{K})$ corresponds to a conductivity of about 1000 Scm^{-1}) in the cooling and warming cycle of the h_8/d_6 70%:30% system and the d_8 system.

Hewlett Packard HP4284A impedance analyzer (ν = 20 Hz - 1 MHz) in the two-probe configuration in the temperature range between 35 K and 80 K. Contacts were attached to the crystal either by the silver paste or the carbon paste diluted in the hexyl-acetate solvent. The higher limit of the contact resistances measured in the insulating phase were estimated to be smaller than 1% and 0.001% of the sample resistance, when the silver and carbon paste were used, respectively. Further, we have assured, by taking into account the results of open-circuit measurements, that the stray capacitances do not influence the real part of the conductivity in the frequency window 20 Hz- 1 MHz and in the temperature range of our study. Complex conductivity measurements were made at increasing temperatures until T_{C1warm} was reached, after the sample was cooled from 100 K through the M-I transition at T_{C1cool} down to 35 K. It should be noted that the latter temperature is higher than T_{C2cool} which designates the transition into the reentrant metallic state in the h_8/d_6 70%:30% system. $T_{\mathrm{C1cool}} \approx 47$ K and 77 K and $T_{\mathrm{C1warm}} \approx 57$ K and 80 K for the h_8/d_6 70%:30% and the d_8 system, respectively (see Figure 1). It should be noted that the respective hysteretic regions (denoted as the region I in Figure 1), in which cooling and warming *R versus T* traces differ, are situated between 40 K - T_{C1warm} and 70 K - T_{C1warm}, respectively [24]. In the cooling cycle the sample resistance was monitored in the two and the four probe configurations. It is also worth of noting that we have observed a variation of the value of T_C inside a few degrees range for the same single crystal after thermal cycling. This *sample memory effect* is not so surprising if we acknowledge the first order nature of the metal-insulator phase transition. However, the observed difference in T_C does not have an influence on the obtained results reported in this paper, since we have done the reported measurements in the well defined conditions as described above. In addition, we have looked for possible non-linear effects. Electric-field-dependent measurements have revealed clear, albeit weak in magnitude, non-linear current-voltage characteristics above large threshold fields of the order of 1 V/cm. In the region II (see Figure 1), the threshold field was found to increase substantially reaching values between 10 and 100 V/cm. The full account of this work will be published elsewhere [25].

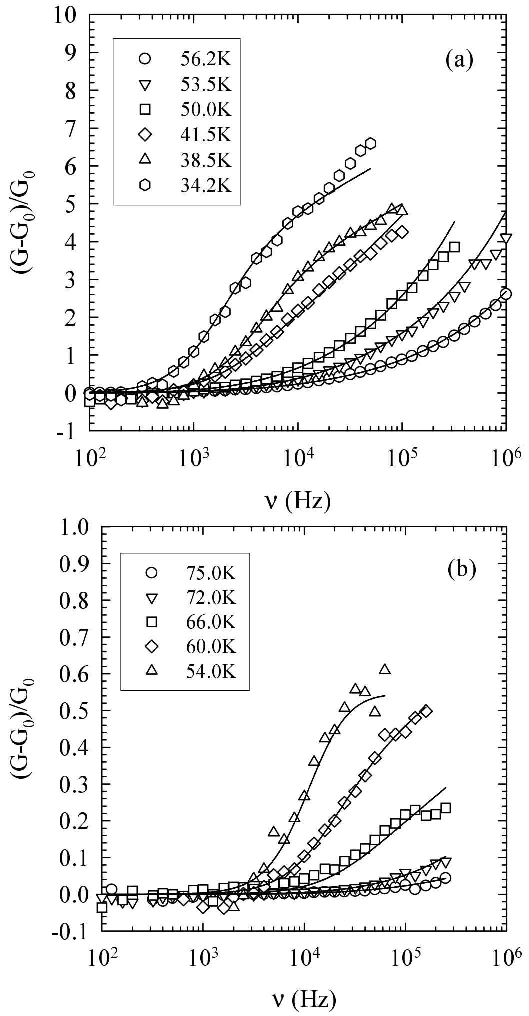

Fig. 2. The real part of the conductivity normalized to the DC value ($(G - G_0)/G_0$) *versus* frequency for (a) a temperature range between 56 K and 34 K in the h_8/d_6 70%:30% system and for (b) a temperature range between 75 K and 54 K in the d_8 system. Full lines are fits to the HN form.

The real part of the conductivity normalized to the DC value $(G(\omega) - G_0)/G_0$ as a function of frequency at a few selected temperatures is shown in Figure 2 (a) and (b) for the h_8/d_6 70%:30% and the d_8 system, respectively. It should be noted that an enhanced frequency-dependent conductivity grows in importance as the temperature lowers. Dielectric functions (Figure 3) were extracted from the conductivity using the relations $\varepsilon'(\omega) = B(\omega)/\omega$, and $\varepsilon''(\omega) = (G(\omega) - G_0)/\omega$, where G_0 is the DC conductivity obtained from the $G(\omega)$ measured at low frequencies

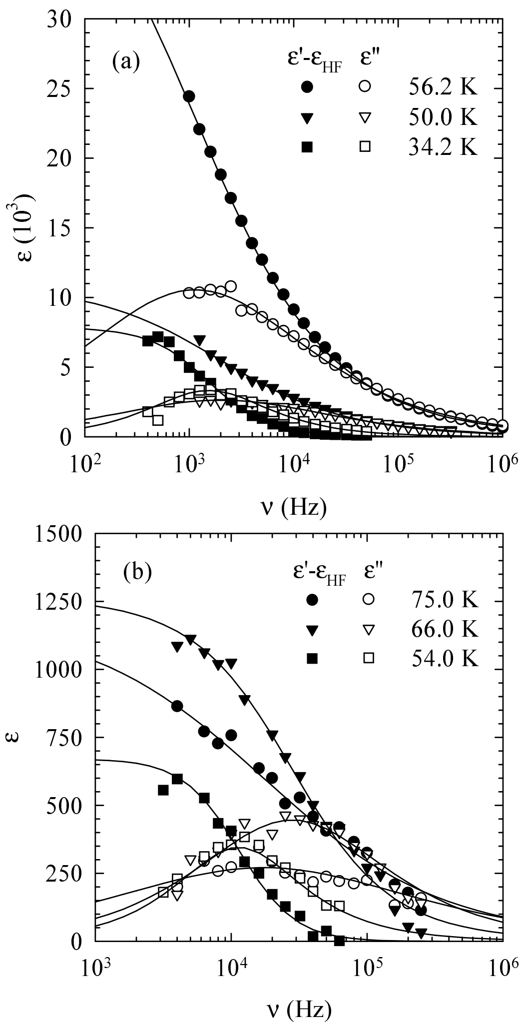

Fig. 3. Frequency dependance (log scale) of the real and imaginary parts of the dielectric function at three selected temperatures for (a) the h_8/d_6 70%:30% system and for (b) the d_8 system. Full lines are fits to the HN form.

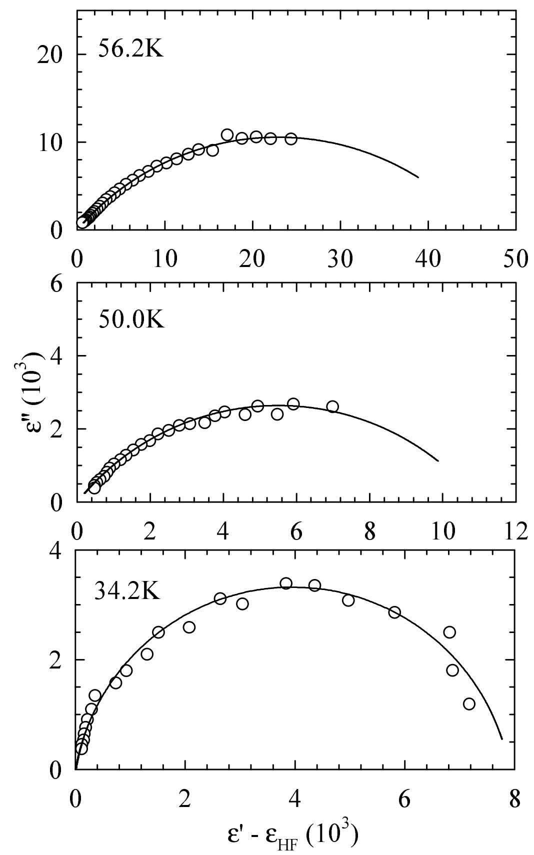

Fig. 4. Cole-Cole plots of the dielectric response at three selected temperatures in the h_8/d_6 70%:30% system. Full lines are from fits to the HN form.

where $G(\omega)$ was independent of ω ($\omega = 2\pi\nu$). The calculated Cole-Cole plots are presented in Figures 4 and 5. The intersection of the arcs with $\varepsilon' - \varepsilon_{\rm HF}$ axis at high $\varepsilon' - \varepsilon_{\rm HF}$ values, corresponding to low frequencies, indicates that the value of the relaxation strength $\Delta\varepsilon = \varepsilon_0 - \varepsilon_{\rm HF}$ is of the order of 10^4 and 10^3 for the h_8/d_6 70%:30% and the d_8 system, respectively. ε_0 and $\varepsilon_{\rm HF}$ are the static and the high frequency dielectric constant, respectively. The observed dielectric response can be well fitted by the phenomenological Havriliak-Negami (HN) function

$$\varepsilon(\omega) - \varepsilon_{\rm HF} = \frac{\Delta\varepsilon}{1 + (i\omega\tau_0)^{1-\alpha}} \tag{1}$$

This function has been widely used to describe the non-Debye character of the relaxation processes in disordered systems [26,27]. τ_0 and $(1-\alpha)$ are the mean relaxation time and the shape parameter which describes the symmetric broadening of the relaxation time distribution function, respectively. The data were analyzed using two methods exposed recently [22]. The full lines in Figures 2, 3, 4 and 5 correspond to the calculated fits. The fit parameters as a function of inverse temperature are shown in Figure 6 and Figure 7 for the h_8/d_6 70%:30% and the d_8 system, respectively. Inspection of Figures 6 and 7 reveal significant similarities among the results for these two systems. First, at temperatures lower than $T_{\rm C1cool}$ ($T_{\rm C1cool} \approx$ 47 K and 77 K, for the h_8/d_6 70%:30% and the d_8 system, respectively) $\Delta\varepsilon$ which corresponds to the static dielectric susceptibility, is of the order of 10^4 - 10^3 and does not change with temperature. The gradual rise of $\Delta\varepsilon$ in the vicinity of $T_{\rm C1warm}$ observed for the h_8/d_6 70%:30% system, was not detected for the d_8 system possibly due to the very narrow $T_{\rm C1cool}$ - $T_{\rm C1warm}$ range. Further, the width of the relaxation time distribution, expressed by the $(1-\alpha)$ parameter, is large close to $T_{\rm C1warm}$, that is $(1-\alpha) \approx 0.5$. With lowering the temperature, the $(1-\alpha)$ parameter increases inside the region I. Outside the region I, the mode continues to narrow and $(1-\alpha)$ attains the value close to 1. Finally, the mean relaxation time $\tau_0 \approx 10^{-4} - 10^{-5}$ s for the both systems displays a saturated, temperature-independent behaviour in the whole temperature range measured.

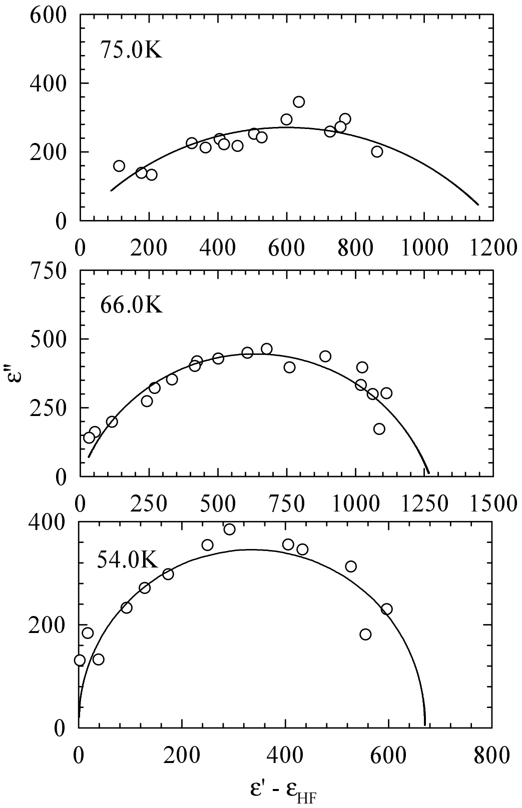

Fig. 5. Cole-Cole plots of the dielectric response at three selected temperatures in the d_8 system. Full lines are from fits to the HN form.

3 Discussion

Our dielectric response data have identified the following features of the relaxation process in the *insulating* phase of the h_8/d_6 70%:30% and the d_8 systems. The width is larger than for the Debye process in the hysteretic region (region I in Figure 1). At lower temperatures, that is in region II (see Figure 1), the process narrows and the width approaches the value typical for the Debye process. The relaxation strength is of the order of 10^4 - 10^5 at T_{C1warm} and decreases on lowering the temperature. At $T < T_{\mathrm{C1cool}}$, $\Delta\varepsilon$ levels off. The characteristic relaxation time is too long to be attributed to free carriers and shows the saturated behaviour in the whole temperature range covered.

Therefore, we are led to identify the origin of this relaxation as an intrinsic property of the insulating state. We propose a charged domain wall pair excitations from the $N = 3$ charge density wave, pinned by commensurability to the underlying lattice, as the relaxation entity. At temperatures below the hysteretic region (that is, in region II in Figure 1), the observed Debye form of relaxation indicates, in the framework of the uniform pinning (single-particle) model [18], that a commensurate pinning of CDW is established on the long range scale. On the other hand, in the hysteretic temperature range (region I in Figure 1) the insulating CDW phase exists only on the short range scale, since the nucleation of the metallic phase

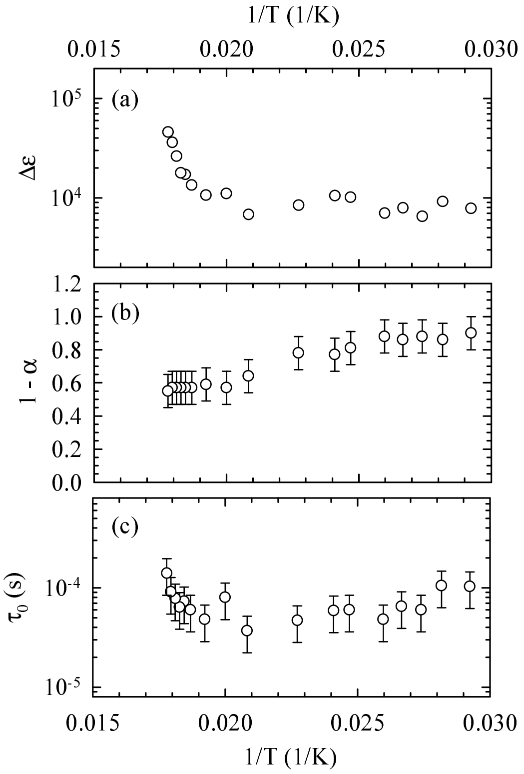

Fig. 6. (a) Relaxation strength ($\Delta\varepsilon$), (b) shape parameter (1-α) and (c) mean relaxation time (τ_0) *versus* inverse temperature in the h_8/d_6 70%:30% system.

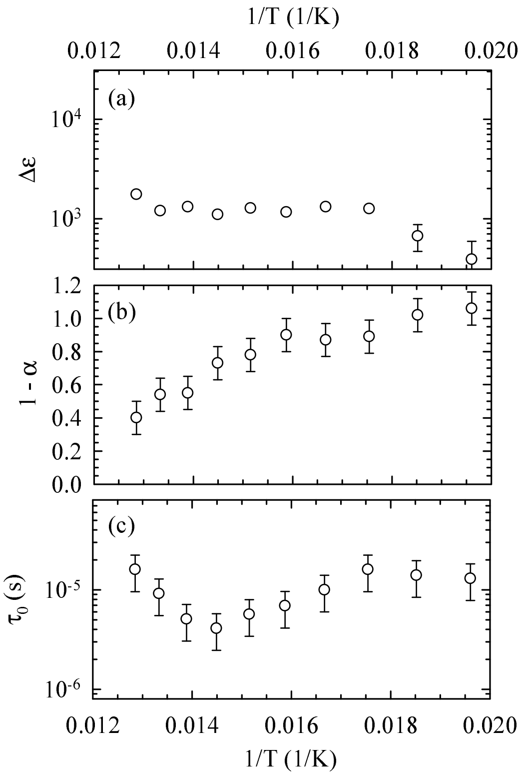

Fig. 7. (a) Relaxation strength ($\Delta\varepsilon$), (b) shape parameter (1-α) and (c) mean relaxation time (τ_0) *versus* inverse temperature in the d_8 system.

starts to grow up. Macroscopically, the system may be described as consisting of insulating (CDW ordered) and metallic domains with a high degree of randomness. Then, dynamically correlated CDW clusters would yield to a broad relaxation, as observed experimentally. This picture is supported by the existence of a percolation threshold for DC conduction found in combined ESR and DC transport measurements [8,23]. That is, these measurements have shown that as long as randomness is larger than a certain value (which scales with 20% of sample volume converted into the insulating phase at T_{C1}) electrons cannot percolate through a disordered lattice and the insulating phase dominates the DC electrical transport.

Further, the saturated, temperature-independent behaviour of the relaxation time is not a feature usually encountered at high temperatures close to the density wave transition temperature. That is, in that region the relaxation time is found to be thermally activated in a manner similar to the dc resistivity [18]. Such a behaviour is shown to be associated with the dominant resistive dissipation [28]. However, a crossover to a more smooth temperature dependence, that is to an almost saturated behaviour has been already reported in incommensurate charge and spin density wave relaxation processes [27,29]. The former has been recently proposed to be a signature of a new low temperature relaxation process attributed to defects [30], while the latter has been associated with a quantum tunneling and taken as an indication that the resistive dissipation mechanism becomes unimportant at lower temperatures [29]. Moreover, a crossover from the activated to the temperature-independent behaviour of the relaxation time at the phase transition temperature has been recently observed in the commensurate spin density wave phase with canted spins [22]. The fact that we do not find such a crossover in the deuterated DMe-DCNQI systems might be a consequence of a strong first order nature of the $N = 3$ CDW phase transition without precursor effects. We have already proposed [22,29] that one could expect such a behaviour if the electron density becomes smaller than the one electron per the CDW domain size L_{DW}. This condition is largely satisfied taking into account the resistance rise of at least about 6 orders of magnitude at the phase transition temperature [23,24]. This gives an effective one-electron length of about 1 mm, which is 1000 times larger than the lower limit of $L_{DW} \approx 0.1-1\mu$m. The latter is estimated from the measured value of the DC electric threshold field for non-linearity, as well as from the ESR measurements [9,31]. It is worth of noting that the observed temperature-independent behaviour of the relaxation time also demonstrates a negligibly small interaction between electrons and CDW excitations, associated with metallic islands and CDW ordered background, respectively. As far as the value of dielectric constant is concerned, the experimentally observed values are much smaller than the one usually encountered for IC CDW (10^7). This finding might not be surprising for $N = 3$ CDW, since the formation of CDW excitations like phase-vortex pairs or charge domain wall pairs should scale inversely with the order of commensurability N as predicted by Lee, Rice and Anderson (LRA) [32]. As far as the temperature behaviour of the relaxation strength is concerned, an observed gradual increase of $\Delta\varepsilon$ on approaching T_{C1warm} for the alloyed system reflects the fact that the importance of the LRA term diminishes in the hysteretic short range regime. On the other hand, a decrease of $\Delta\varepsilon$ observed below 54 K (See Figure 7(a)) for the pure system indicates that the LRA term starts to play a crucial role once the long range commensurate order is fully established in the whole bulk of the crystal. In our view, these results strongly suggest that the metallic islands act as domain walls to the CDW commensurate background.

Finally, we address briefly the subject of non-linearity observed above a finite threshold field. Theoretically, the collective DW transport by sliding above a finite threshold field is associated with the phason mode with a small gap, which exists in the incommensurate structure with randomly distributed impurities. When the commensurate phase is approached, the phason velocity tends to zero due to the substantial increase of the gap in the phason spectrum. However, the importance of this energy term strongly diminishes with increasing the order of commensurability N. Keeping that in mind, experimentally observed non-linearities which appear above large threshold fields in the range of 1-100 V/cm are in a rather good accordance with what we would expect for $N = 3$ CDW order. In particular, the important rise of E_T once the long range $N = 3$ CDW order is achieved in the whole bulk of the crystal might be compared with a similar effect in TTF-TCNQ when $N = 3$ is induced under pressure [33].

4 Conclusion

In conclusion, we have observed for the first time low-frequency dielectric relaxation in a novel insulating state of the organic π - inorganic d hybrid Cu-(DMe-DCNQI)$_2$ systems with different levels of deuteration. We identify broad relaxation as the intrinsic property of the $N = 3$ commensurate charge density wave below the Peierls-Mott phase transition and suggest that metallic islands act as charged domain walls in the random domain commensurate structure. We interpret the Debye relaxation observed at lower temperatures to be the manifestation of the $N = 3$ CDW long range order established outside the hysteretic region. An important rise of the threshold field attaining the values of the order of 10 - 100 V/cm appear to be also associated with the bulk 3D long range $N = 3$ charge density wave phase.

To our knowledge this might be the first opportunity to study experimentally the theoretically intriguing issue of the collective AC and DC CDW dynamics in the random domain commensurate structure in an organic π - inorganic d hybrid conductor. Further combined DC-AC transport measurements are planned to establish the mechanism of the observed non-linear effects and to correlate it to the low-frequency dynamics.

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