ESRF	**Experiment title:** **CHARGE-TRANSFER MULTI-EXCITON STRINGS AT THE NEUTRAL-TO-IONIC TRANSITION**	**Experiment number**: 02 02 200
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Report:

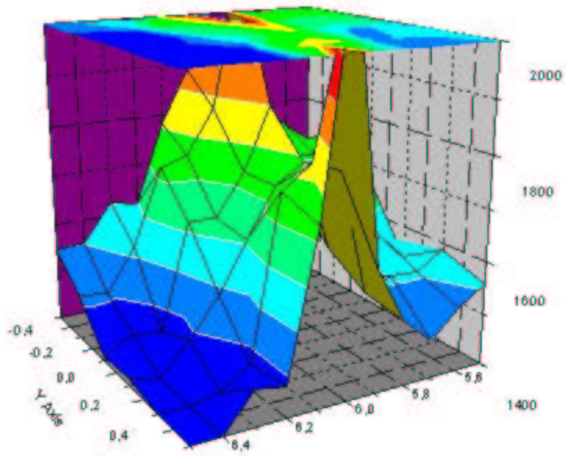
The possibility to tune between different ground states some organic materials is one of their most fascinating features. This situation especially occurs in some quasi-one-dimensional charge-transfer (CT) complexes with mixed-stack architecture, where the alternation of electron donor (D) and acceptor (A) molecules gives rise to chain multistability between a regular neutral (N) state …D°A°D°A°D°A°… and two degenerate dimerized ionic I_+ …$(D^+A^-)(D^+A^-)(D^+A^-)$… and I_- …$(A^-D^+)(A^-D^+)(A^-D^+)$… states. Electronic and structural aspects are especially closely coupled : changes of the electronic state only exist with large structural distortions, affecting both intra- and inter-molecular geometry. The thermal activation of lattice-relaxed CT exciton-strings is at the heart of N-I transition mechanism:

$$...D°A°D°A°\mathbf{(D^+A^-)(D^+A^-)(D^+A^-)}D°A°D°A°...$$

This type of non-linear excitations, specific of quasi-one-dimensional systems with strong electron-phonon coupling, can also be discussed in terms of self-trapped CT multi-excitons, called CT strings. Many intriguing physical properties are governed by these non-linear excitations. At finite temperature the thermal excitation of boundaries between "phases" destroys any long range order and the isolated chain becomes intrinsically inhomogeneous with the condensation of structurally relaxed CT strings. Inter-chain coupling in the crystal can drive phase transitions, and thus the so-called N-I transition may be induced by temperature, pressure and also light.

In addition to the thermally-induced fluctuations, CT excitations of quantum nature can be generated at low temperature and may be responsible for quantum paraelectric phenomena. This may be observed if no thermal transition occur, i.e. if the neutral phase is still the stable one at 0 K. Such restricting condition is realized in some complexes undergoing the N-I transition above the atmospheric pressure. Therefore we have investigated in the last experiment (Sept 2002) the (BEDT-TTF)-(ClMeTCNQ) system undergoing the N-I transition at 0 K at 2 kbars. Because of the well pronounced 1D physical properties of the complex, as the ones observed in previously investigated complexes undergoing the N-I transition, we expected to observe diffuse scattering signal associated with thermally induced CT excitations observed in the other complexes. As shown on fig 1, diffuse signal was observed at room temperature. This is an evidence of the intrinsic fluctuations in such systems associated with a large dielectric response.

Figure 1:
Diffuse planes associated with the 1D nature of the charge-transfer exciton-strings.



Once the diffuse scattering signal observed, we wanted to study the behaviour as the function of temperature. Unfortunately the cryostat was not the same as the one previously used. Some technical problems did not allow to correct the thermal contraction and the sample was not anymore at the centre of the 4 circles goniometer. In addition, the signal/noise ratio could not be optimised in order to study the weak diffuse scattering signal. Therefore we could not perform the precise studies.
However the observation of diffuse scattering in the (BEDT-TTF)(ClMeTCNQ) complex is an important result, it is necessary to perform a complete and precise investigation in the low temperature range in order to evidence the possibility of CT fluctuations of quantum nature.