Searching for Crystallographic Superstructures in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

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Abstract

To resolve a superstructure formation previously reported for the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, we present synchrotron x-ray diffraction experiments carried out at the MAGS beamline at BESSY, HZB. Surprisingly, in our low temperature (28K) experiments, when searching **k**-space at (h 0 3.5), h = 7, 8 and (h 0 0.5), h = 5, 7, for none of these spots we could detect scattering intensity associated to a superstructure formation, in contradiction to previous reports. Our data suggest that details of the structural properties of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br - such as superstructure formation - sensitively depend on sample handling, *e.g.*, cooling rates (in our case 4K/min), or thermal cycling. A direct relationship between superstructure formation and terminal ethylene group ordering cannot be verified, disproving proposals put forth previously.

Key words:

Due to its exotic superconducting- and normal-state properties, which resemble those of the high-T_c cuprates, the organic charge-transfer salt κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (abbreviated κ-Br hereafter) has been intensively studied in recent years [1, 2, 3]. Resulting from a layered crystal structure, the electronic properties of this material are quasi-twodimensional. It leads to electronic ground state properties which are commonly summarized within the framework of a bandwidth-controlled Mott transition [4].

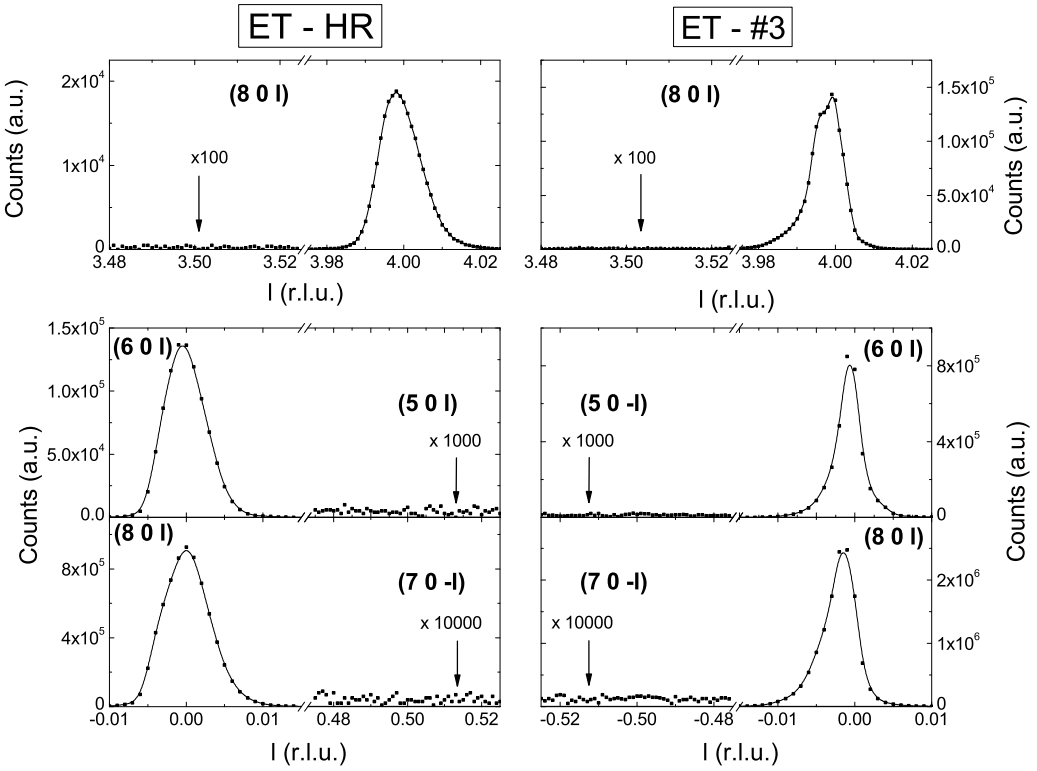

Figure 1: Peak intensity at superstructure positions in reciprocal space measured for two samples κ-Br at 28 K, together with structural reference peaks; for details see text.

There are various reports on superstructure formation occurring in κ-Br (see for instance Refs. [5, 6]). Tentatively, it has been associated to an ordering transition at T_{SS} of the terminal ethylene groups of the BEDT-TTF molecule [7]. Only, in a recent structural investigation we could not find evidence to support the notion that the terminal ethylene groups undergo an order-disorder transition [8].

Subsequently, we have set out to reexamine the claims of superstructure formation by studying two thoroughly characterized crystals κ-Br by means of synchrotron x-ray diffraction experiments carried out at the MAGS beamline at BESSY, HZB. The experiments have been performed in vertical scattering four-circle geometry at a photon energy of 12.398keV, and at low temperatures of 28 K (cooling rate 4K/min). To avoid irradiation damages the beam intensity was reduced using absorber foils. We have studied two

samples with different resistive characteristics, labelled as ET-HR and ET-#3 (see Ref. [3] for sample description). In Fig. 1 we summarize the essential result of our study by plotting the observed intensity distribution for some of the superstructure peak positions studied [9]. In the figure, we include the intensities from structural Bragg peaks close-by, which serve as a reference.

From the figure it can be seen that within experimental resolution we observe no intensity at those positions in **k**-space, where previously superstructure peaks were detected [5, 6]. It indicates that superstructure formation is not generic to κ-Br, but depends on external parameters like sample synthesis, handling and cooling, thermal cycling etc. Especially, the claims of a direct connection between superstructure formation and terminal ethylene group ordering, as proposed in Ref. [7], appears to be disproven by our data.

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[9] We have scanned **k**-space at (h 0 3.5), h = 7, 8 and (h 0 0.5), h = 5, 7.