BEDT-TTF molecular structural phases in dependence of doping molecule characteristics

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Abstract

In our report we show how the system demonstrates a transition from a parallel arranged molecular phase (β-phase) to a new equilibrium state where the molecules deflect at a certain angle (α- or θ-phase). This is due to the electrostatic and quadrupole interaction between BEDT-TTF and doping molecules. The modified potential of the Girifalco type is introduced and it depends on both intermolecular distance and the angle of the molecular rotation. It is found that the equilibrium distance between molecules in a stack increases and the deflection angle arises with an increase of the charge of doping molecules, which leads to new equilibrium states.

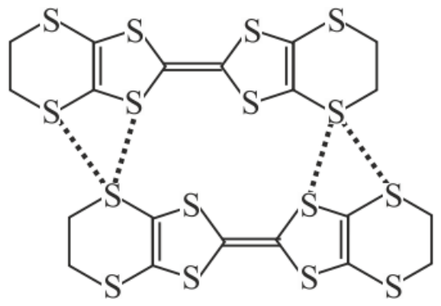

Fig. 1. Schematic representation of two BEDT-TTF molecules and their bonding between stacks [1].

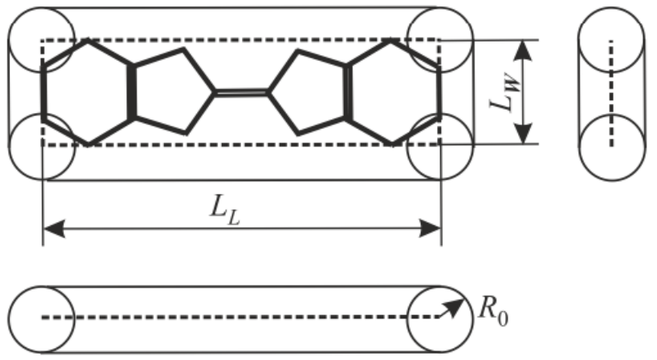

Fig. 2. A rectangular plate model of a molecule in the form of a rectangle parallelepiped superimposed on the real structure of BEDT-TTF molecules.

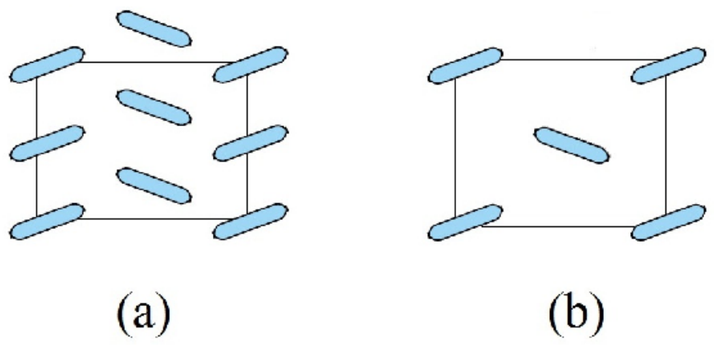

Fig. 3. (a) the α-phase; (b) the θ-phase [2].

Interaction between molecules includes the Girifalco potential, the quadrupolar and electrostatic potentials:

$$U(r) = U_{gir} + U_{quad} + U_{el}\text{, where}$$

$$U_{gir} = n_{\text{int}}\varepsilon_0\left[\left(\frac{r_0}{r}\right)^{12} - 2\left(\frac{r_0}{r}\right)^{6}\right]; \quad U_{el} = \frac{q^2}{r_0}\left(\frac{r_0}{r}\right);$$

$$U_{quad} = \frac{9Q^2}{4r^5}\left(1 - 10\cos^2\theta + \frac{35}{3}\cos^4\theta\right)$$

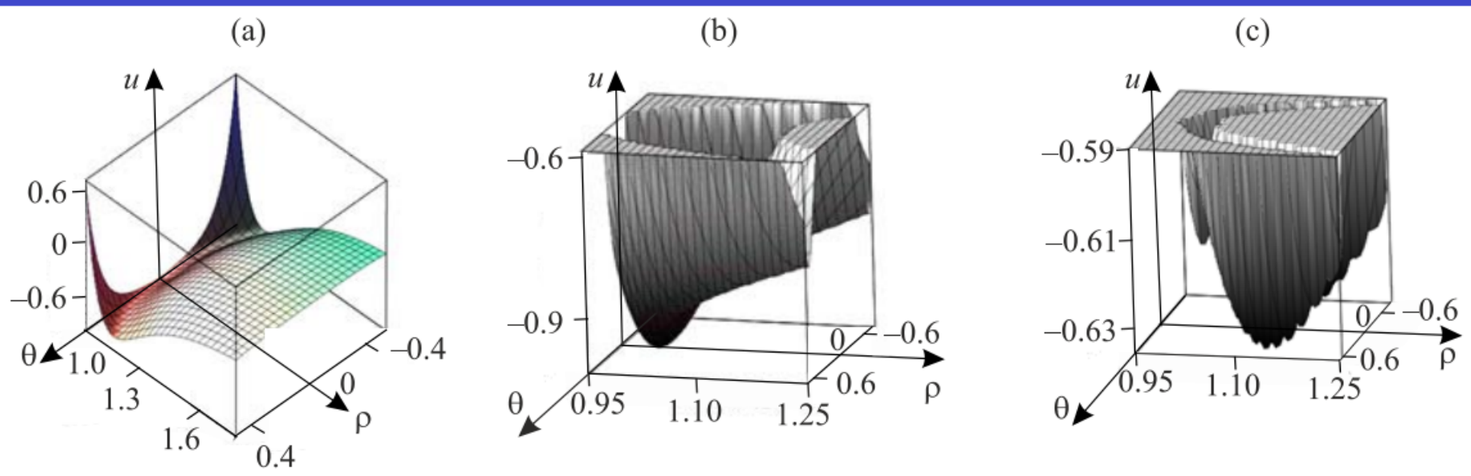

Fig.4. (a) General view of Lennard-Jones potential in form (11) with angular dependence on θ in the first term. (b) The similar enlarged image, with the minimum visible at ρ = 1, θ = 0. (c) The same fragment for the full potential (11), two new minima (new phase) are visible at ρ ≈1.13 , θ=±0.48 rad.

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2. B. Commeau, R. M. Geilhufe, G. W. Fernando, and A. V. Balatsky. Structural and electronic properties of α-(BEDTTTF)2I3, β-(BEDT-TTF)2I3, and κ-(BEDT-TTF)2X3 (X= I, F, Br, Cl) organic charge transfer salts. *Phys. Rev. B* **96**, 125135 (2017).