Theoretical Classification of Two-Dimensional Organic Conductors

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Abstract

Several different polytypes have been found in the family of quasi-two-dimensional organic conductors designated as θ, κ, and so on, which exhibit characteristic ground states quite different from each other. We present a new unified scheme to classify these polytypes by use of the band structural parameters in an anisotropic triangular lattice model. Our theoretical calculations based on the above model including the on-site and inter-site Coulomb interactions indicate the actual relevance of these parameters to the ground state nature.

Key words: organic conductor; Mott insulator; band structure

A series of quasi-two-dimensional(Q2D) organic charge transfer salts are known for their rich electronic states, which are common to the strongly correlated electronic systems. Recent findings on the field-induced superconductivity in λ-BETS$_2$FeCl$_4$[1], or, the charge ordering phenomena (CO) in θ-ET salts[2] are only a fraction of the extensive and intensive studies made so far. However, the Q2D organic systems still lack the unified view of how to understand the ground state nature systematically, which is because their structural variety makes it difficult to deal all the materials on equal footing. Therefore, the classification of these organic systems becomes a confronted issue, which we are going to develop in this paper. Their band structure will be systematically characterized as an extension of the theoretical work by Kino and Fukuyama[3].

The systems we deal with have a layered structure, whose conducting molecular plane based mainly on ET or BETS molecules are charged with 3/4 electrons on an average. The materials are categorized by several molecular packing patterns (polytypes), which are assigned Greek letters such as α, β, θ, κ, and λ. In addition, the organic materials easily undergo structural changes by the external pressure or by the chemical replacements, and this flexibility together with a number of polytypes gives variety in their band structure. In the following, we give a new scheme to classify the band structure of representative polytypes and materials.

The schematic description of the molecular arrangements and band structures of four different polytypes, θ, β, κ, and λ, are shown in Fig. 1. The first step of classification is given by the number of molecules in the unit cell; θ and β have two molecules whereas κ and λ include four molecules. The latter group is characterized by the existence of a dimer (i.e. a pair of molecules connected by a large transfer integral), and can be transformed into the dimer-based system. After this process, both groups are mapped onto an anisotropic triangular lattice shown in the inset of Fig.1, where each lattice site corresponds to a molecule and a dimer, respectively. Then, the band structure of θ or β is straightforwardly related to that of this triangular lattice, and so as the upper two bands of κ or λ which are regarded as anti-bonding bands (the other two bonding bands situated far below the Fermi level are obliterated). Therefore the filling of bands becomes effectively 3/4-filled and 1/2-filled, respectively.

On the other hand, the classification within each group is given by the parameterization of the relative relationships between the upper and lower bands; the θ and κ bands have degeneracy along the Brillouin zone boundary (X-M-Y) whereas the β and λ does

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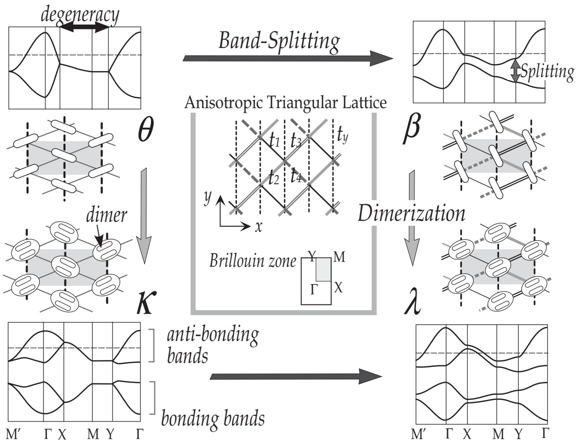

Fig. 1. Relationships among polytypes. Band structures and molecular arrangements are shown schematically.

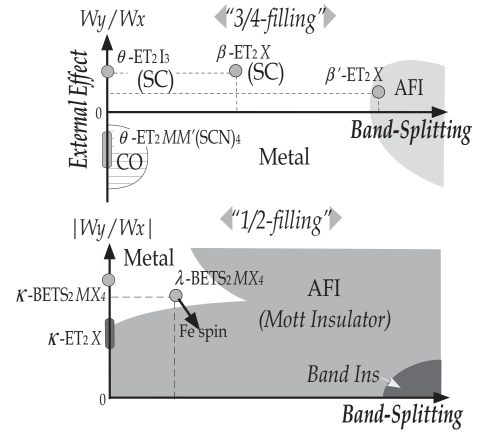

Fig. 2. Schematic ground state phase diagrams on the plane of two independent band parameters. The "3/4" and "1/2-filled" cases are adopted to θ, β and κ, λ-types, respectively.

not, which is classified as the degree of *"band splitting"*. Also the external effect such as pressure changes the detailed structure of bands, particularly, the ratio of dispersion along x and y-directions, W_y/W_x. This secondary band parameter also plays an important role in the classification of ground states.

We performed the mean-field calculation in an extended Hubbard model based on the anisotropic triangular lattice, which includes the on-site and the nearest neighbor site Coulomb interactions, U and V. The detail of the model and the results are presented elsewhere [4]. The schematic ground states as functions of band structural parameters are shown separately for both groups in Fig. 2; the axis of *band splitting* which discriminate θ and β or κ and λ is taken independently from the axis of W_x/W_y explained above. The interaction parameters are fixed to the appropriate values of the organic charge transfer salts, with U of the order of bandwidth and $V/U \sim$0.2-0.3. Here, U is transformed into that of the dimer orbital, $U_{\rm dimer}$, in the dimer-based latter group. Now, we see that the phase diagram of the former group is characterized by the CO phase which is confined in the particular region of small *band splitting*. Otherwise, the metallic phase (virtually including the SC state) is dominant which is transformed into the AF insulator (AFI) when *the splitting of two bands* become considerably large. On the other hand, the dimer-based latter group has the insulating phase except in the small *band splitting* region, whose Mott insulating character is expected to become a band insulator in the limit of large band splitting.

The actual materials are mapped on the phase diagram. The most general example of the 3/4-filled group is a series of θ-ET$_2MM'$(SCN)$_4$ (M=Tl,Rb, M'=Zn, Co, etc.), known as CO materials [2] whose parameter values fall exactly in the region of CO. Also the ground states of other β-type materials are well reproduced. As regards the 1/2-filled case, most of the intensively studied κ and λ materials have relatively small *band splitting* and are situated near the metal-insulator (MI) boundary. This is the reason why they could easily exhibit MI transitions by the external pressure or by chemical replacements, as we are familiar with κ-ET$_2X$. Finally, we should particularly emphasize that the λ-BETS$_2$FeCl$_4$ known to have the AFI ground state[5] is also discussed within our scheme; without the internal field (Fe spins) this material is placed in the metallic region very close to the Mott insulator. Then, the introduction of internal magnetic field (Fe) drives the system into the Mott insulator, as we have already discussed in Ref.[6], where we also noticed that κ and λ are classified by the degree of *band splitting*, which was a preliminary idea of the present work. Actually, the κ-BETS systems exhibit a robust metallic character in contrast to the insulating λ-analogue.

In summary, we presented a new theoretical scheme to classify the band structure of Q2D organic conductors in an anisotropic triangular lattice. The ground state phase diagram obtained by the mean-field calculation shows good relevance to the actual materials.

The author thanks Prof. Hidetoshi Fukuyama for his kind and useful discussions.

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