

Charge Ordering in (TMTTF)₂X: A Density Functional Theory Approach

Yukihiro Shimoi¹ and Kaoru Iwano²

¹Nanotechnology Research Institute (NRI),

National Institute of Advanced Industrial Science and Technology (AIST), Japan

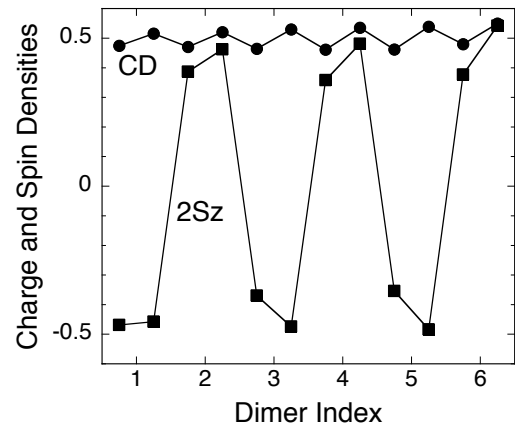
²Institute of Materials Structure Science, KEK, Japan

Email: y.shimoi@aist.go.jp

We investigate theoretically the charge ordering in (TMTTF)₂X, typical quarter-filled one-dimensional conductors. We argue this issue using density functional theory (DFT) calculations. This method enables us to discuss electronic properties based on actual molecular structure and arrangement and is complementary to an approach in terms of model Hamiltonians adopted in most previous theoretical works.

The cluster-based DFT calculations are augmented by point charges surrounding the cluster in order to incorporate the environmental effects in crystal. The degrees of point charges are determined self-consistently with charge densities at corresponding atoms in the cluster. We demonstrated that this approach successfully describes charge ordering and optical properties of (EDO-TTF)₂PF₆ and clarifies the mechanism of stabilizing the charge ordering in this material [1].

Figure shows the charge and spin densities at each molecule in a linear chain of six TMTTF dimers (12 molecules). The cluster is extracted from the crystal structure of (TMTTF)₂PF₆ at 20 K. We can see inhomogeneous distribution of charge and spin within each dimer, suggesting a weak charge ordering with the (0101) pattern. This feature is qualitatively consistent with NMR experiment [2]. Note that the disproportionation in spin is larger than that in charge. The spin is largely polarized at each molecule and aligns in the same direction within the dimer and in the opposite one between the neighboring dimers. Therefore, the results indicate the coexistence of the weak charge ordering with a dimer-Mott spin configuration.



[1] K. Iwano and Y. Shimoi, Phys. Rev. B 77 (2008) 075120.

[2] T. Nakamura, K. Furukawa, T. Hara, J. Phys. Soc. Jpn. 76 (2007) 064715.