**Theoretical Study of Electron Correlation in Molecular Crystals under Pressure by Combining Ab Initio and Effective Model Calculations**
**(**Parallel Session 1**, Oral)**

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To investigate physical properties and to explore novel states in molecular crystals, the application of pressure has been a predominant tool. Taking advantage of their mechanical flexibility, the controlling factors for interacting many-body problems can be varied in a drastic range of energy by external “*physical*” pressure. Moreover, “*chemical*” pressure, which is introduced via chemical substitution among each family of compounds without changing the lattice structure, has been widely used. These two methods have frequently provided unified phase diagrams combining them as a common pressure axis. However, systematic theoretical investigations at a quantitative level have been lacking, which are crucial in evaluating the determining factors and in elucidating the similarities and differences between chemical and physical pressure effects.
 For this purpose, we apply an *ab initio* scheme to a typical family of molecular conductors composed of TMTSF or TMTTF molecules forming 2∶1 salts with monovalent anions [1]. By deriving the extended Hubbard-type Hamiltonians from first-principles band calculations and evaluating not only the intermolecular transfer integrals but also the Coulomb parameters, we discuss their material dependence, i.e., the chemical pressure effect, in the unified phase diagram. We then apply the many-variable variational Monte Carlo method to accurately determine the symmetry-breaking phase transitions and show the development of the charge and spin orderings. Furthermore, we applied this scheme combined with the x-ray crystal structure analysis on TMTTF2PF6 under pressure up to 8 GPa [2], as a direct theoretical investigation of the physical pressure effect.
 Finally, we discuss a recent development of pressure control of magnetism, i.e., the piezomagnetic effect, in novel collinear-type antiferromagnets but with macroscopic time-reversal symmetry breaking [3], coined “altermagnets”, in a typical strongly-correlated molecular crystals, k-type BEDT-TTF compounds [4].

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[2] M. Itoi, K. Yoshimi, H. Ma, T. Misawa, T. Tsumuraya, D. Bhoi, T. Komatsu, H. Mori, Y. Uwatoko, and H. Seo, Phys. Rev. Research **6**, 043308 (14 pages) (2024).
[3] M. Naka, S. Hayami, H. Kusunose, Y. Yanagi, Y. Motome, and H. Seo, Nat. Comm.**10**, 4305 (2019); Phys. Rev. B **102**, 075112 (2020).
[4] M. Naka, Y. Motome, T. Miyazaki, and H. Seo, preprint arXiv:2505.07327.フォームの始まり

フォームの終わり