Analysis of *ab initio* Hamiltonians for molecular solid $(TMTTF)_2PF_6$ under pressure

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Organic conductors are widely studied due to the diverse physical properties that arise from the interactions of charge, spin, and lattice degrees of freedom. These interactions lead to various electronic states, including superconductivity, magnetic ordering, and charge ordering. In fact, crystals made of TMTTF molecules exhibit all these electronic states when external pressure is applied and have been the subject of active research for over 40 years. By combining these findings with results from related compounds made of TMTSF molecules, a unified pressure-temperature phase diagram has been discussed. However, there has not yet been a quantitative analysis of how external pressure impacts electron correlations in these systems, nor an understanding of what factors control the various phases under pressure.

In this project, we have performed a combined experimental and theoretical analysis of the pressure-dependent physical properties of the quasi-one-dimensional organic conductor $(TMTTF)_2PF_6$ [1]. We obtain the crystal structures under pressure by performing x-ray diffraction measurements for single crystals of $(TMTTF)_2PF_6$ with a diamond anvil cell up to 8 GPa. Based on the obtained crystal structures, we first derive the low-energy effective Hamiltonians for $(TMTTF)_2PF_6$ under pressure using Quantum ESPRESSO [2] and RESPACK [3]. We obtained the following characteristic features of the microscopic parameters in the low-energy effective Hamiltonians: (1) By applying the pressure, the transfer integrals increase, whereas the screened Coulomb interactions decrease, resulting in a drastic reduction of correlation effects. For example, the normalized onsite Coulomb interaction ($U/\bar{t}_a$) decreases from 12 at ambient pressure to 6 at 8 GPa. (2) The degree of dimerization in the intrachain transfer integrals, as the result of the decrease in structural dimerization together with the change in the intermolecular configuration, almost disappears above 4 GPa.

Then, we solve the low-energy effective Hamiltonians by using the many-variable variational Monte Carlo method [4, 5], which effectively treats electron correlation effects. We find that the charge ordering is substantially suppressed above 1 GPa while the spin ordering survives up to higher pressure. This theoretical result is consistent with the temperature dependence of the resistivity under pressure.

Changes in the electronic states of molecular solids under pressure have been studied as a typical example of pressure effects on strongly correlated electron systems. The present study demonstrates that recent advances in high-pressure experimental and computational tech-