

Computational Studies of Pressure Effects on Electronic Correlations and Superconductivity in Organic Conductors

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The objective of this research is to estimate detailed crystal structures under pressure and predict functional properties of strongly correlated materials using *ab-initio* calculations. Special attention is given to organic molecular solids, where pressure significantly affects electronic properties, including charge/spin ordering, superconductivity, and topological phenomena. Based on crystal structure data obtained from experimental groups, we derived low-energy effective models under pressure and quantitatively analyzed electronic correlation effects. We also performed first-principles calculations to optimize structures obtained under ambient pressure and verified consistency with experimental results. As a large number of systematic calculations were required for various pressure conditions, scripts were generated for bulk jobs using Moller [4] and exhaustive calculations were carried out for several pressure conditions in order to significantly improve the efficiency of the calculations.

In this study [5], single-crystal X-ray diffraction experiments on the organic conductor (TMTTF)₂PF₆ were performed under pressures up to 8 GPa, enabling precise structural determination. Using these structures, we conducted first-principles calculations with Quantum ESPRESSO [1] and derived low-energy effective models using RESPACK [2], based on the calculated band structures and Wannier functions. These models were then analyzed using mVMC [3], thoroughly evaluating

the stability of charge and spin ordering under pressure.

As a result of the significant decrease in crystal volume with increasing pressure, the conduction bandwidth is broadened and the screen-Coulomb interaction is dramatically reduced. Additionally, the degree of dimerization in the conduction bands sharply decreased and nearly vanished above 3 GPa, significantly diminishing the strength of electronic correlations. Furthermore, we confirmed a good agreement between experimental resistivity measurements and theoretical analysis. Specifically, the stability of spin-density wave (SDW) and charge ordering (CO), derived from theoretical calculations, was consistent with experimental resistivity data. The CO notably became unstable above approximately 1 GPa, while the SDW persisted at higher pressures. These outcomes clarify the differences between physical and chemical pressures [6] and deepen the understanding of unified phase diagrams.

Additionally, we performed X-ray structural analysis and electronic structure calculations under pressure on the organic superconductor β' -(ET)₂ICl₂ [7]. Our findings revealed a Lifshitz transition around 6 GPa, changing the Fermi surface from one-dimensional to two-dimensional. Concurrently, the on-site Coulomb interaction significantly decreased, reaching approximately half its ambient-pressure value around 10 GPa.

These results provide crucial insights into the mechanism of superconductivity under high pressure and elucidate the significant impact of pressure-induced electronic structure changes on superconductivity in organic materials.

References

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