【Grant-in-Aid for Transformative Research Areas（B）】

Multiply Programmed Layers: Advanced Functionalities in Ultrathin Films through Electronic and Lattice Degree of Freedom Control

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Project Information	Project Number : 25B205　Project Period (FY) : 2025-2027 Keywords : Ultrathin film, Charge-transfer complex, Molecule, Interface, Electronic function

Purpose and Background of the Research

●Outline of the Research

The properties of materials vary dramatically with changes in their size and shape. **Two-dimensional (2D) atomic layered materials** have emerged as a significant research area in modern science due to the discovery of unique phenomena that differ from those observed in three-dimensional (3D) crystals. **Charge-transfer (CT) complexes**, in which the donor and acceptor molecules are regularly arranged, exhibit diverse physical properties such as electrical conductivity and magnetism which depend on the molecular combinations and their arrangements.

In this study, we will create ultrathin films with nanometer-scale thickness, consisting of CT complexes and elucidate their physical properties. Exotic quantum phenomena resulting from the new ordered structures and the interfacial effects which arise from the reduction of dimensionality will be explored. By utilizing molecular designability and bottom-up assembly strategies, ultrathin films of **"Multiply Programmed Layers"** with the intra-layer and/or inter-layer molecular arrangements will be constructed. Precisely controlled **"Multiply Programmed Layers"** produce unconventional crystal structures and electronic states, leading to new functions together with excellent external field responsiveness attributed to their ultrathin state. We aim to realize exotic electronic phases, which normally appear only at very low temperatures and under high pressure, at ambient conditions. This will pave the way for the development of electronic devices based on new principles.

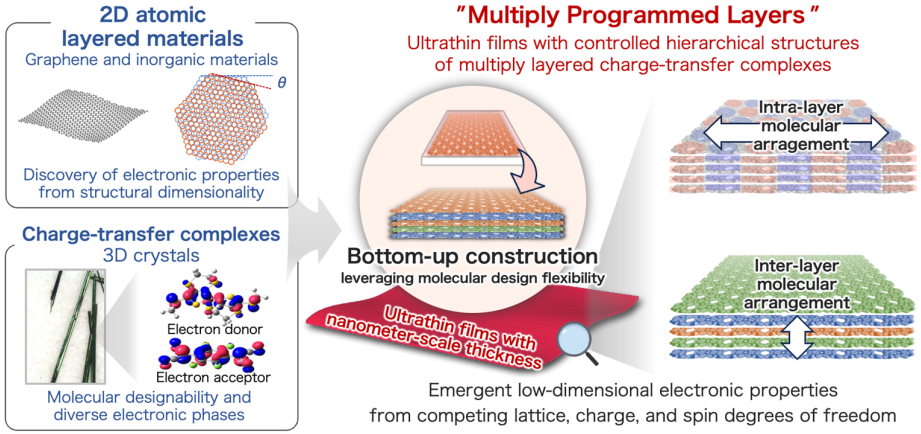

Figure 1. Project overview

●Significance of this study

Inorganic 2D materials like graphene, with strong intra-layer covalent bonds and weak inter-layer van der Waals forces, can be exfoliated into high-quality ultrathin films due to their structural anisotropy. In contrast, molecular crystals which typically form 3D structures held together by weak intermolecular forces are difficult to exfoliate while preserving order, highlighting the need for rational methods to construct highly ordered molecular ultrathin films.

Our research addresses this chagllenge using CT complexes, which offer high molecular designability. Through a bottom-up approach that enables precise control over molecular arrangements, we aim to create **"Multiply Programmed Layers"** ultrathin films that exhibit electronic structures unique to low-dimensional molecular materials.

Expected Research Achievements

The project aims to create **"Multiply Programmed Layers"** by sequentially stacking various molecular layers composed of CT complexes with a variety of electronic phases and clarify the correlation between their hierarchical structure and physical properties.

● **A01: Molecular Design and Synthesis:**
Tomoko Fujino (PI, YNU), **Yuya Haraguchi** (TUAT), **Kazuyoshi Yoshimi** (UTokyo)
Electronic structures using CT complexes will be designed by controlling molecular arrangements and inter-layer orbital hybridization. Heterostructures with inorganic layered materials will also be explored.

● **A02: Layered Structure Construction: Rie Makiura** (PI, OMU)
Ultrathin crystalline films will be synthesized at the air–liquid interface via self-assembly, and structure–property correlations will be examined.

● **A03: Functionaility Development:**
Toshiki Higashino (PI, AIST), **Satoru Inoue** (YGU), **Takeshi Fujita** (UTokyo)
Electronic phases in Multiply Programmed Layeres will be controlled using external stimuli to explore unique low-dimensional functionalities for device applications.

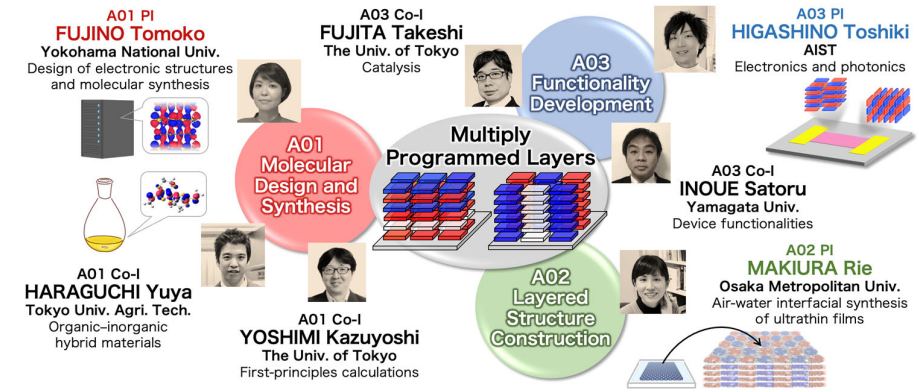

Figure 2. Research organization

Homepage Address, etc.	https://multiply.issp.u-tokyo.ac.jp/