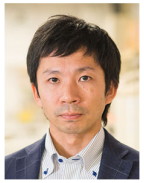
【Grant-in-Aid for Scientific Research (S)】

Advanced measurement for unveiling hierarchical quasiparticles towards renovating molecular materials research



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Project Information	Project Number : 23H05461 Project Period (FY) : 2023-2027 Keywords : photoemission, vibronic coupling, quasiparticle, wavefunction

Purpose and Background of the Research

●Outline of the Research

Phenomena challenging existing theories are being reported in molecular materials due to strong electron-phonon coupling. Our study presents a hierarchical understanding of the classical quasiparticle polaron, an electron-phonon system, offering a fresh perspective on molecular materials. Through advanced spectroscopic measurements and theoretical simulations on oriented molecular crystals, we analyze the symmetry and spatio-temporal variation of the quasiparticle wavefunction. This approach enables us to explore molecular functions beyond structures and orbitals, revolutionizing the design of molecules and their aggregates. Ultimately, it paves the way for innovative molecular materials with unprecedented control over physical properties, such as high-mobility and low-thermal-inactivation.

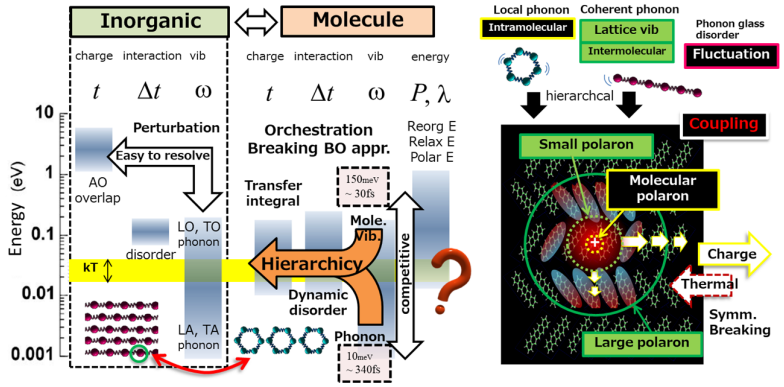

Figure 1. Differences between inorganic and molecular solids and hierarchy of quasiparticles.

●Fundamental Q: What is the "molecular shape"?

Molecular materials' sensitivity hinders understanding their macroscopic properties. Quasiparticle polaron undergoes unique temporal and spatial changes, affecting material properties. Electronic localization in molecules relies on orientation, alignment, bonding, and size within solids. Phonons, intramolecular and lattice vibrations, significantly impact localization. To comprehend polarons' hierarchy, we suggest using the "molecular shape" as a novel starting point. By capturing dynamic shape of quasiparticles, their functional properties can be revealed.

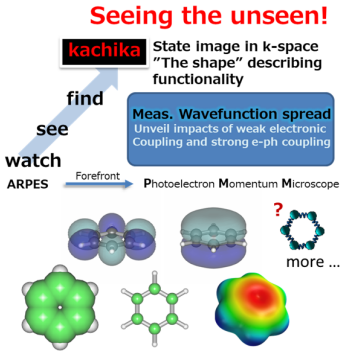

Figure 2. Open molecular function measurement

●The Power of Advanced Photoelectron Momentum Microscopy (PMM) to Record Hierarchical Shapes

PMM overcomes limitations of conventional spectroscopy, offering compatibility with small crystals and easy-to-damage molecules. It enables highly sensitive, multi-dimensional measurements. PMM's microscopic and imaging capabilities facilitate selective view of hierarchical units in real or reciprocal space. It provides a comprehensive overview of valence electron orbitals and spins, crucial for functional expression. PMM's key feature is its ability to measure valence band structure (energy, momentum, orbital, and spin) using a single technique. Photoelectron tomography images reconstruct wavefunctions, offering novel structural insights into molecules.

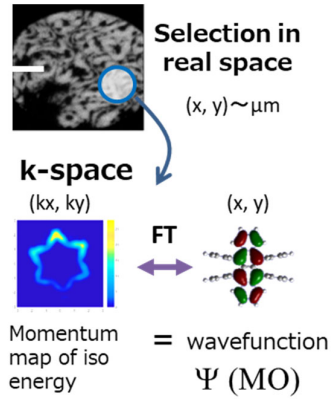

Figure 3. Hierarchical view by PMM

Expected Research Achievements

●STEP1: What does the 3D momentum image of photoemission observe?

PMM's 2D momentum map reconstructs wavefunctions and analyzes molecular orbital distributions based on group theory. High-resolution tomography reveals structural insights through quasiparticle wavefunction distortions and periodicity analysis. PMM redefines "molecular shape" based on distortion structures, characterizing molecular function transformations.

●STEP2: Can we control the many-body problem by understanding the concertation of electron correlation and electron-phonon couplings?

Prepare high-quality organic crystal samples for itinerant electrons. Use PMM's photoelectron tomography for wave-function measurement and advanced analytical methods. PMM's electronic structure imaging clarifies hierarchy and enables understanding of molecular solids' spatio-temporal variations and diverse functions.

●STEP3: What molecular shape governs physical properties?

Realization of the “shape" of the molecular materials will not only provide design guidelines for high-performance devices, but will also lead to the establishment of a new structural science that fuses molecular and electronic structures to explore the unexplored functions. We are aiming at the era of “molecular orbitronics” as a technology that uses the unique characteristics of molecules and is not a mere substitute for inorganic materials. If quasiparticle engineering is advanced based on the quantum concepts developed in hierarchical shape measurement, it will open the way to the creation of new values and markets.

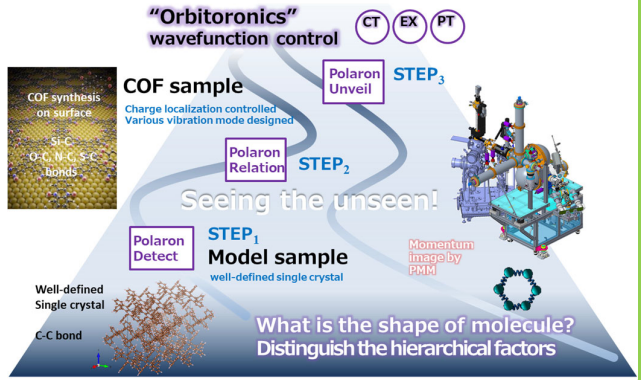

Figure 4. Three steps in research plan

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