Microscopic understanding of exciton and carrier physics in functional molecules represents a great challenge, because of their complexity resulting from the strong electron-phonon coupling. It is of great importance to probe not only electron dynamics but also structural dynamics for gaining mechanistic details of their functions. The applicant has been working on several molecule-based optoelectronic materials: (1) hybrid lead-halide perovskites, and organic semiconductors which demonstrate (2) singlet fission (SF) or (3) thermally activated delayed fluorescence (TADF). The applicant has applied ultrafast spectroscopy to probe the structural/geometrical dynamics of functional materials associated with photo-induced electronic dynamics.

(1) Hybrid lead halide perovskites (LHPs) have become star materials for new-generation solar cells due to their exceptional tolerance to defects, which gives rise to long carrier lifetimes, long diffusion lengths, and low electron-hole recombination rates. Those remarkable optoelectronic properties can be explained by efficient charge screening along with polaron formation. The applicant detected the large polaron formation directly using time-resolved optical Kerr effect (TR-OKE). Dynamic responses from the inorganic sub-lattice detected in time domain uncovered that charge screening happens in less than 1 ps in both hybrid $CH_3NH_3PbBr_3$ and all-inorganic $CsPbBr_3$. Based on the unusual responses, the applicant proposed a new type of polaron: ferroelectric large polaron. Due to the enormous anharmonicity of the lattice responses, the lattice surrounding an injected carrier might undergo ferroelectric phase transition due to the electric field exerted from the carrier, which results in much stronger screening than conventional Fröhlich polaron picture.

(2) For SF materials, the applicant used coherent phonon spectroscopy as a tool to probe a new type of coherent SF. SF is the process where photo-excited singlet exciton splits into two triplet excitons in 100 fs ~ ps time scale. SF is getting popular because of its unique character as an exciton multiplier, which gives us a chance to enhance optoelectronic properties. To gain the microscopic mechanisms of SF, the applicant focused on rubrene single crystal as a model system, which is famous for showing thermally activated singlet fission. Unlike the conventional incoherent SF. The applicant discovered ultrafast coherent SF occurring even at low temperature. By analyzing coherent phonon oscillation on the transient absorption signal, the author succeeded in uncovering the dynamics of coherent SF activated by symmetry breaking phonons.

(3) Most recently, the applicant has been conducting systematic studies on TADF molecules using time-resolved infrared spectroscopy (TR-IR). TADF is gathering attention for its potential to boost the efficiency of organic light-emitting diodes without using precious metals, but the mechanistic details along intersystem crossing remain elusive. By probing the structural dynamics of the TADF materials, the applicant clarified the variation of the functions from the viewpoint of molecular geometry in the excited states. The findings clearly show that the geometrical configuration of TADF molecules in excited states is key to the efficiency of TADF materials.

As listed above, The applicant has consistently uncovered the complex electron/structural dynamics of emergent semiconductor materials from the viewpoint of ultrafast spectroscopy, tackling many of scientifically essential questions regarding optoelectronics.