Our recent studies in vibronic coupling: simulation of optoelectronic materials, derivation of Jahn-Teller formalisms, and interpretation of photoelectron spectrum

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Singlet fission (SF) is a vibronic coupling process that converts one short-lived singlet exciton to two long-lived triplet excitons [1]. It can potentially enhance the power conversion efficiency in photovoltaic devices. In the new concept of intramolecular singlet fission (iSF), chromophore units are covalently bonded, so that it is easier to adjust inter-chromophore morphology to facilitate SF. In this seminar, our recent studies of SF materials are presented. Through using our new diabatization scheme [2] and performing quantum chemistry calculations and quantum dynamics simulations, we investigated effects of chromophore size in iSF and presented a step-by-step picture for the through-liner iSF [3,4]. With the new understanding, we proposed a strategy to enhance the iSF efficiency by making appropriate substitution on the linker. We also explored the significance of azaborine substitution in designing SF chromophores [5].

In the second half of the seminar, I will present our works on deriving general formalisms of Jahn-Teller (JT) and pseudo-Jahn-Teller (pJT) Hamiltonians. JT/pJT problems are induced by symmetry-related vibronic coupling and have far-reaching consequences in chemistry and physics. A growing number of studies reveal the limitations of the standard 2nd order expansions of JT/pJT Hamiltonians in vibrational coordinates. Although some case-specific high-order expansion formulas had been derived, general JT/pJT Hamiltonian formalisms up to arbitrary order were still unavailable. We endeavor to derive such general formalisms. Employing the root-branch approach and the modularized approach, we have successfully derived general expansion formulas for all trigonal [6], tetragonal [7], tetrahedral and octahedral [8,9] JT/pJT Hamiltonians. Spin-orbit JT/pJT expansion formulas for trigonal and tetragonal symmetries have also been derived [10]. A python program VHEGEN has been developed to generate the expansion formulas [11]. The trigonal formulas are used to simulate the photo-detachment spectrum of CO_3^-. New insight into this spectrum is obtained [12].

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