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Direct Observation of Resonant Energy Transfer in Lanthanide Complexes from Multidisciplinary Perspectives

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Abstract

Photosensitized energy transfer (EnT) phenomena occur frequently in a variety of photophysical and photochemical processes and have been treated traditionally with the donor-acceptor distance dependent Förster and Dexter models. However, in special case incorrect arguments and formulae were often employed by ignoring energy resonance conditions and selection rules of state-to-state transition, especially for the sensitive intramolecular EnT of lanthanide complexes bearing a uniform donor-acceptor distance. Herein, we proposed an innovative model of energy degeneracy crossing controlled EnT, which can be experimentally confirmed by a bi-exponential decay using the time-resolved photoluminescence measurements. The computationally determined energy resonance region provides the most effective channels to achieve the metal-to-ligand EnT beyond the distance-dependent model and sensitively bifurcates into the symmetry-allowed or forbidden channels for some representative europium antenna complexes. The outcomes of multidisciplinary treatment contribute a complementary EnT model that can be tuned by introducing a phosphorescence modulator through the mechanism-guided crystal engineering, and should motivate further development of mechanistic models and applications.

Keywords: Photosensitization mechanism; resonant energy transfer; light-harvesting complex; ab initio calculation; time-resolved spectroscopy; crystal engineering

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