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<http://molmat.kuicr.kyoto-u.ac.jp/index-e.html>



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Scope of Research

Our research goal is to develop high-performance organic electroluminescence devices, organic solar cells, and polymer materials. Toward this, we carry out syntheses, device fabrications, precise structure characterizations, and quantum chemical calculations for high functional organic materials. Along with exploring novel synthetic routes and novel devices, we perform detailed analyses of structures and dynamics, mainly by sophisticated solid-state NMR spectroscopy, in order to obtain structure–dynamics–property relationships.

KEYWORDS

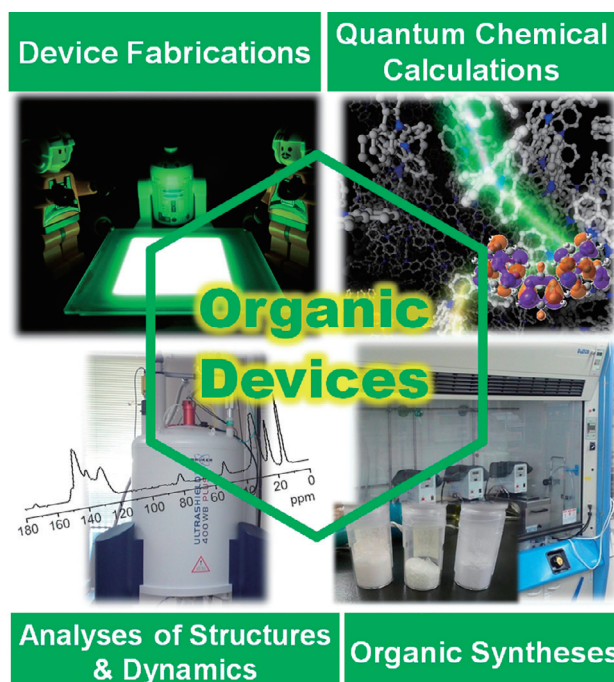
Organic Light-Emitting Diodes

Solid-State NMR

Quantum Chemical Calculation

Amorphous Materials

Dynamic Nuclear Polarization Enhanced NMR



Recent Selected Publications

Shizu, K.; Kaji, H., Theoretical Determination of Rate Constants from Excited-States: Application to Benzophenone, *J. Phys. Chem. A*, **125**, 9000-9010 (2021).

Ren, Y.; Wada, Y.; Suzuki, K.; Kusakabe, Y.; Geldsetzer, J.; Kaji, H., Efficient Blue Thermally Activated Delayed Fluorescence Emitters Showing Very Fast Reverse Intersystem Crossing, *Appl. Phys. Express*, **14**, [071003-1]-[071003-5] (2021).

Wada, Y.; Shizu, K.; Kaji, H., Molecular Vibration Accelerates Charge Transfer Emission in a Highly Twisted Blue Thermally Activated Delayed Fluorescence Material, *J. Phys. Chem. A*, **125**, 4534-4539 (2021).

Theoretical Determination of Rate Constants for Materials Screening

Theoretical prediction of rate constants has attracted great attention because of its relevance to materials chemistry. We report a cost-effective method of theoretically predicting electronic transition rate constants from the excited states of molecules. Our method is based on quantum chemical calculations of electronic states and the Fermi golden rule. We apply the method to the excited-state decay mechanism of photoexcited benzophenone and show that calculated rate constants, including radiative (fluorescence) and nonradiative decays from S_1 to S_0 , radiative decay (phosphorescence) from T_1 to S_0 , and intersystem crossing from S_1 to T_2 , are quantitatively in good agreement with the experimental ones. From the calculated population kinetics, a stepwise $S_1 \rightarrow T_2 \rightarrow T_1 \rightarrow S_0$ transition is found to be the predominant decay pathway. The direct $S_1 \rightarrow T_1$ transition is suppressed owing to a fast $S_1 \rightarrow T_2$ intersystem crossing. Our method of predicting rate constants is useful for accelerating materials screening.

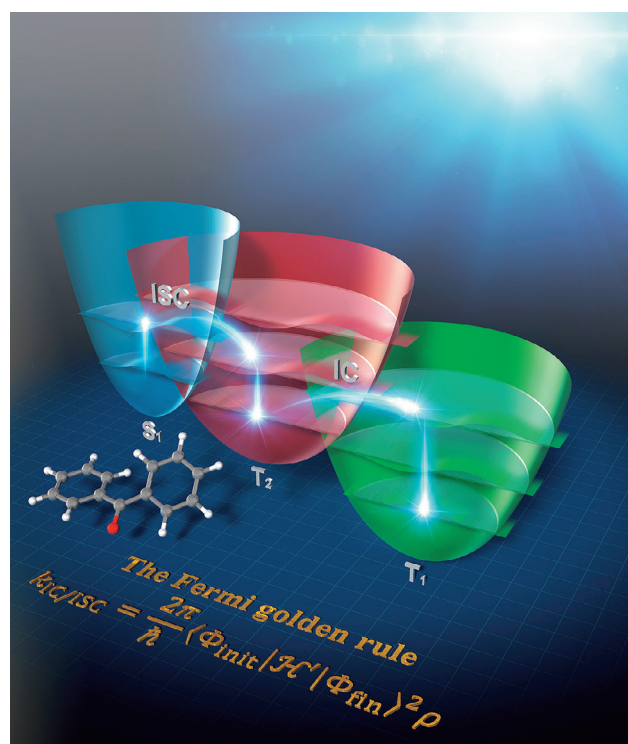


Figure 1. A stepwise $S_1 \rightarrow T_2 \rightarrow T_1$ decay mechanism of photoexcited benzophenone.

Thermally Activated Delayed Fluorescent Materials Showing Very Fast Reverse Intersystem Crossing

We report a robust molecular design, named “tilted Face-to-Face alignment with Optimal distance (tFFO),” for thermally activated delayed fluorescence materials showing very fast reverse intersystem crossing (RISC). tFFO design simultaneously realizes near-degenerate 1CT , 3CT and 3LE states (where 1CT and 3CT denote singlet and triplet charge transfer states, respectively and 3LE denotes triplet locally excited state) and notable spin-orbit coupling between them by controlling the through-space distance between the donor and acceptor segments in a molecule with tilted intersegment angles. Based on the concept, the first example molecule, TpAT-tFFO, realized very fast RISC with a rate constant of $1.2 \times 10^7 \text{ s}^{-1}$. In its application to organic light-emitting diodes, TpAT-tFFO exhibited high performance even at very high brightness owing to the ultrafast RISC. Our tFFO strategy can be versatily expanded to various types, numbers, and combinations of segments.

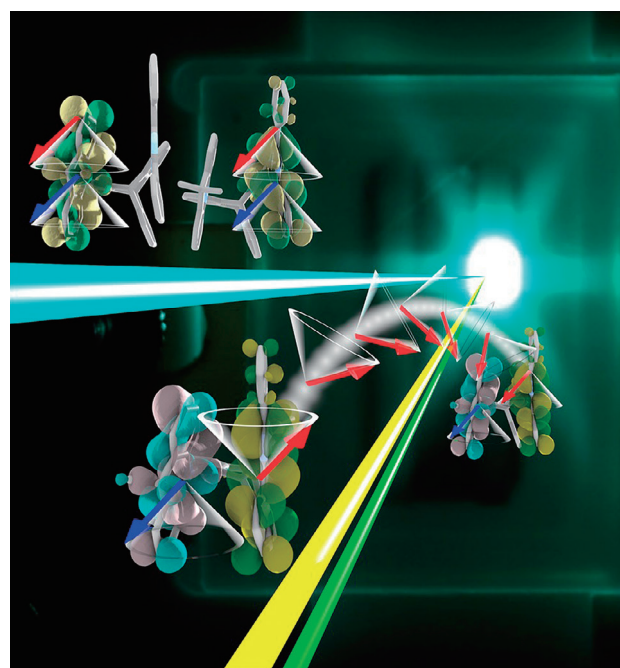


Figure 2. A thermally activated delayed fluorescence molecule showing very fast reverse intersystem crossing.