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– Molecular Materials Chemistry –

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

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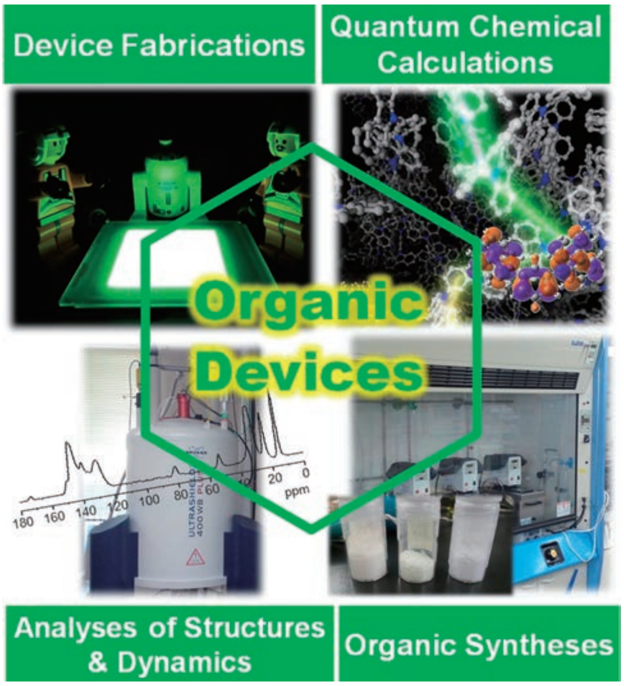
Scope of Research

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
Organic Solar Cell
Solid-State NMR
Quantum Chemical Calculation
Amorphous Materials

Selected Publications

Suzuki, K.; Kubo, S.; Aussenac, F.; Engelke, F.; Fukushima, T.; Kaji, H., Analysis of Molecular Orientation in Organic Semiconducting Thin Films Using Static Dynamic Nuclear Polarization Enhanced Solid-State NMR Spectroscopy, *Angew. Chem. Int. Ed.*, **56,** 14842 (2017).

Miwa, T.; Kubo, S.; Shizu, K.; Komino, T.; Adachi, C.; Kaji, H., Blue Organic Light-Emitting Diodes Realizing External Quantum Efficiency Over 25% Using Thermally Activated Delayed Fluorescence Emitters, *Sci. Rep.*, **7,** 284 (2017).

Moon, C.; Suzuki, K.; Shizu, K.; Adachi, C.; Kaji, H.; Kim, J.-J., Combined Inter- and Intramolecular Charge-Transfer Processes for Highly Efficient Fluorescent Organic Light-Emitting Diodes with Reduced Triplet Exciton Quenching, *Adv. Mater.*, **29,** 1606448 (2017).

Deep-blue Organic Light-emitting Diodes Realizing External Quantum Efficiency Over 25% Using Horizontally Oriented Thermally Activated Delayed Fluorescence Emitters

Organic light-emitting diodes (OLEDs) have attracted great interest as a promising technique for developing solid-state lighting source and flexible flat-panel displays. To date, much improvements to the electroluminescence efficiency of OLEDs have been done employing thermally activated delayed fluorescence (TADF). However, it remains challenging to realize highly efficient deep-blue OLEDs. We develop deep blue TADF emitters, 3-(9H-[3,9′-bicarbazol]-9-yl)-9H-xanthen-9-one (CCX-I) and 3-(9′H-[9,3′:6′,9″-tercarbazol]-9′-yl)-9H-xanthen-9-one (CCX-II). They exhibit highly efficient deep-blue emission and high triplet-to-singlet conversion efficiency. In addition, CCX-I and CCX-II orient parallel to the glass substrate, which leads to increasing light-outcoupling efficiency and improving electroluminescence efficiency. A CCX-II-based OLED shows a maximum external quantum efficiency (EQE) of 25.9% with International de L’Eclairage (CIE) coordinates of (0.15, 0.22). With an out-coupling sheet, the maximum EQE further increased to 33.3%.

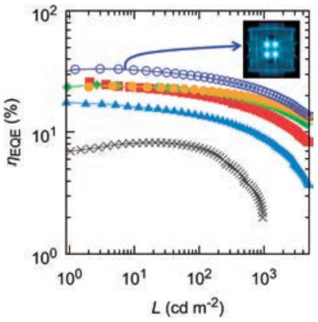


Figure 1. Luminance–EQE characteristics of the OLEDs containing CCX-II as an emitting dopant.

Charge Transport Simulations in Amorphous Thin Films for OLEDs

A fundamental understanding of charge transport properties of organic thin films is essential to improve OLED performance. Charge transport properties of organic molecules have been discussed in terms of their HOMO and LUMO energy levels and electronic couplings between molecules. The HOMO and LUMO can be calculated using quantum chemical calculations for isolated molecules, while the electronic couplings are considered to depend largely on an aggregated structure. We performed multi-scale charge transport simulations for amorphous structures of *N*,*N*′-dicarbazole-3,5-benzene (mCP) and 4,4′-bis(*N*-carbazolyl)-1,1′-biphenyl (CBP), which have been used as a host material for OLEDs. By explicitly considering organic molecules, we investigated the contribution of respective molecular pairs to the charge transport in amorphous thin films. The molecular-level analysis of the charge transport simulations showed that molecular pairs with large electronic couplings are not most important in charge transport processes. Charges were found to be transported effectively in the forward direction via other molecular pairs that do not have substantially large electronic couplings.

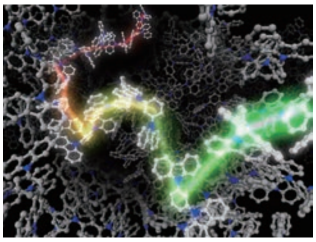


Figure 2. Image of charge transport in amorphous organic thin films.

Analysis of Molecular Orientation in Organic Semiconducting Thin Films Using Dynamic Nuclear Polarization Enhanced Solid-state NMR Spectroscopy

Molecular orientation in amorphous organic semiconducting thin-film devices is an important issue affecting device performance. However, to date it has not been possible to analyze the “distribution” of the orientations. Although solid-state NMR (ssNMR) spectroscopy can provide information on the “distribution” of molecular orientations, the technique is limited because of the small amount of sample in the device and the low sensitivity of ssNMR. Here, we report the first application of dynamic nuclear polarization enhanced ssNMR (DNP-ssNMR) spectroscopy for the orientational analysis of amorphous phenyldi(pyren-1-yl)phosphine oxide (POPy$_2$). The ^{31}P DNP-ssNMR spectra exhibited a sufficient signal-to-noise ratio to quantify the distribution of molecular orientations in amorphous films: the P=O axis of the vacuum-deposited and drop-cast POPy$_2$ shows anisotropic and iso-tropic distribution, respectively. The different molecular orientations reflect the molecular origin of the different charge transport behaviors.

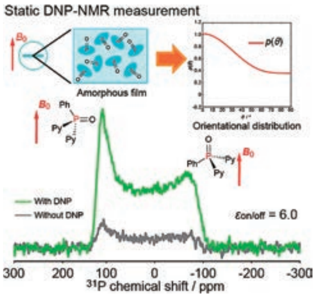


Figure 3. Analysis of molecular orientation using DNP-NMR.