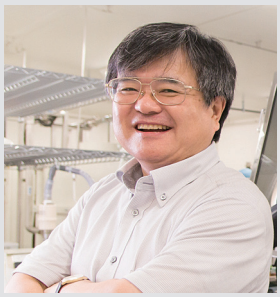
Organic Solar Cells

Department of Materials Molecular Science
Division of Molecular Functions



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Keywords Organic Semiconductors, Up-Conversion, Lateral Junction

Organic solar cells have been intensively studied due to many advantages like flexible, printable, light, low-cost, fashionable, *etc*. Followings are our recent results. (1) Photon up-conversion (UC) from near infrared (NIR) to visible yellow (Figure 1) by utilizing charge transfer (CT) states at donor/acceptor (D/A) interface of organic solar cells. (2) A novel concept of the structure of organic solar cell, namely, a lateral junction in which the photogenerated holes and electrons are laterally transported and extracted to the respective electrodes. Even 1.8 cm-length lateral cells (Figure 2) showed clear photovoltaic behavior. (3) The reduction of open-circuit voltage (V_{oc}) loss due to non-radiative recombination in organic solar cells by using high-mobility organic semiconductors. The V_{oc} reaching to thermodynamic (Shockley-Queisser) limit was observed. (4) The ppm-level doping effects in organic semiconductor films and organic single crystals for organic solar cells. So far, we have reported complete *pn*-control, doping sensitization, and the ppm-level doping effects using an ultra-slow deposition technique reaching 10^{-9} nm s^{-1} (Figure 3) in organic rubrene single crystals measured by the Hall effect, which shows a doping efficiency of 82% comparable to the B in Si. These results can be regarded as a foundation for the construction of highly efficient organic solar cells.

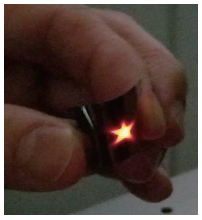

Figure 1. Up-converted (UC) yellow emission by star-shaped near infrared irradiation.

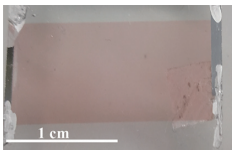

Figure 2. Picture of a 1.8 cm-length lateral cell.

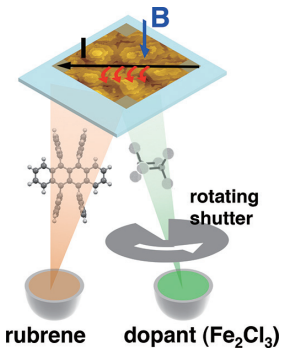

Figure 3. Ultra-slow co-deposition technique to produce the doped rubrene single crystal for Hall effect measurements.

Selected Publications

- M. Hiramoto, M. Kikuchi and S. Izawa, "Parts-per-Million-Level Doping Effects in Organic Semiconductor Films and Organic Single Crystals," *Adv. Mater.* **30**, 1801236 (15 pages) (2018).
- S. Izawa and M. Hiramoto, "Efficient Solid-State Photon Upconversion by Spin Inversion at Organic Semiconductor Interface," *Nat. Photonics* **15**, 895–900 (2021).
- J. P. Ithikkal, A. Girault, M. Kikuchi, Y. Yabara, S. Izawa and M. Hiramoto, "Photovoltaic Behavior of Centimeter-Long Lateral Organic Junctions," *Appl. Phys Express* **14**, 094001 (2021).
- *Organic Solar Cells—Energetic and Nanostructural Design*, M. Hiramoto and S. Izawa, Eds., Springer Nature Singapore Pte Ltd. (2021).
- M. Hiramoto, "Organic Photocurrent Multiplication," in *the Series of Electronic Materials: Science & Technology*, Springer Nature Singapore Pte Ltd. (2023).

1. Blue Organic Light-Emitting Diode with a Turn-On Voltage at 1.47 V[1)]

Blue light is vital for light-emitting devices, lighting applications, as well as smartphone screens and large screen displays. However, it is challenging to develop efficient blue organic light-emitting diodes (OLEDs) owing to the high applied voltage required for their function. Conventional blue OLEDs typically require around 4 V for a luminance of 100 cd/m^2; this is higher than the industrial target of 3.7 V—the voltage of lithium-ion batteries commonly used in smartphones. Therefore, there is an urgent need to develop novel blue OLEDs that can operate at lower voltages.

We reported a novel OLED device with a remarkable ultralow turn-on voltage of 1.47 V for blue emission and a peak wavelength at 462 nm (2.68 eV) (Figure 4).[1)] This OLED operates via a mechanism called upconversion (UC). Herein, holes and electrons are injected into donor (emitter) and acceptor (electron transport) layers, respectively. They recombine at the donor/acceptor (D/A) interface to form a charge transfer (CT) state. Subsequently, the energy of the CT state is selectively transferred to the low-energy first triplet excited states of the emitter, which results in blue light emission through the formation of a high-energy first singlet excited state by triplet–triplet annihilation (TTA). As the energy of the CT state is much lower than the emitter's bandgap energy, the UC mechanism with TTA significantly decreases the applied voltage required for exciting the emitter. As a result, this UC-OLED reaches a luminance of 100 cd/m^2, equivalent to that of a commercial display, at just 1.97 V.

Figure 4. Lighting up a blue organic LED with a single 1.5 V battery.

2. Exciton-Free Carrier Generation in Doped Rubrene Single Crystals[2)]

We combined the rubrene organic single crystal growth technique with our original ultra-slow deposition technique of 10^{-9} nm s^{-1} and have succeeded in producing the 1 ppm doped organic single crystal and in detecting the Hall effect signal (Figure 3).

Doping efficiencies of 82 and 60% for hole creation were observed for the rubrene single crystal by doping with the organic acceptors F4-TCNQ and HAT-CN, respectively. Corresponding activation energies (ΔE_A) are 9 and 26 meV (Figure 5), which are smaller than the thermal energy of room temperature, So, the organic acceptor doping can be regarded as exciton-free carrier generation. The organic dopants also observed a slight decrease in Hall mobility. An interstitial doping model is proposed to avoid hole scattering and disturbance of hole delocalization. On the other hand, less efficient doping efficiencies of 37 and 8% by doping inorganic acceptors of Mo_3O_9 and Fe_2Cl_6 were observed. Corresponding ΔE_A are 51 and 144 meV (Figure 5). This can be regarded as carrier generation through the Wannier excitons. A significant decrease in Hall mobility was observed for the inorganic dopants. A substitutional doping model that considers both hole scattering and the disturbance of hole delocalization is proposed.

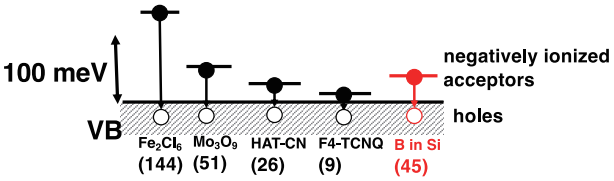

Figure 5. Energetic diagram of acceptor levels for Mo_3O_9, Fe_2Cl_6, F4-TCNQ, and HAT-CN in rubrene single crystal. That for B in Si is also shown for comparison. Numericals in the parentheses are activation energies in meV.

References

1) S. Izawa, M. Morimoto, K. Fujimoto, K. Banno, Y. Majima, M. Takahashi, S. Naka and M. Hiramoto, *Nat. Commun.* **14**, 5494 (2023).

2) M. Hiramoto, "Carrier generation in high-mobility organic semiconductors," Keynote lecture in 13th International Conference on Nano-Molecular Electronics, Tokyo Institute of Technology, Japan, 13, Dec. 2022.

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