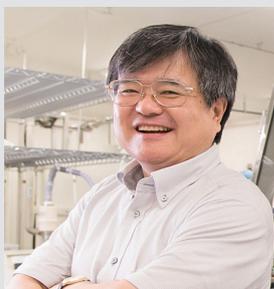


Organic Solar Cells

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Awards

2021 Outstanding Achievement Award, Molecular Electronics & Bioelectronics Division, Japan Society of Applied Physics
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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.

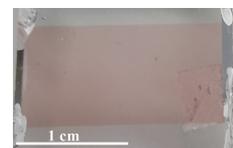


Figure 2. Picture of a 1.8 cm-length lateral cell.

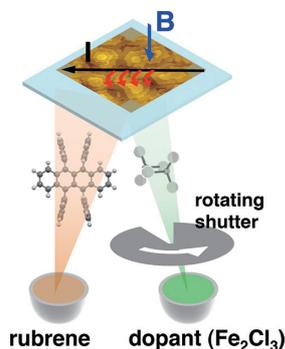


Figure 3. Ultra-slow co-deposition technique to produce the doped rubrene single crystal for Hall effect measurements.

Selected Publications

- S. Izawa and M. Hiramoto, "Efficient Solid-State Photon Up-conversion 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).
- S. Izawa, N. Shintaku, M. Kikuchi and M. Hiramoto, "Importance of Interfacial Crystallinity to Reduce Open-Circuit Voltage Loss in Organic Solar Cells," *Appl. Phys Lett.* **115**, 153301 (2019).
- 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).
- *Organic Solar Cells—Energetic and Nanostructural Design*, M. Hiramoto and S. Izawa, Eds., Springer Nature Singapore Pte Ltd. (2021).

1. Efficient Interfacial Up-Conversion Enabling Bright Emission with Extremely Low Driving Voltage in Organic Light-Emitting Diodes¹⁾

We reported the photon up-conversion (UC) from near infrared (NIR) to visible yellow (Figure 1).²⁾ The UC emission occurs by the combination of organic solar cell process and up-conversion (UC) process (Figure 4(b)). Double layer consists of rubrene donor (D) and non-fullerene acceptor (A) was used. In the solar cell process, the free charges are generated at D/A interface by near-infrared (NIR) excitation. In the UC process, the charge recombination at the D/A interface takes place and triplet states (T_1) are formed in the rubrene film through the CT states. Finally, the UC emission occurs by the T–T annihilation.

The UC process can also occur by the injected electrons and holes from the electrodes. So, we could fabricate the up-converted organic EL device. Red EL of 2.0 eV could be emitted by applied voltage of half energy of 1.0 V by utilizing up-conversion (Figure 4(a)). 100 cd/m² at 1.5 V and 1000 cd/m² at 2.5 V were obtained. A battery of 1.5 V was enough for EL operation. This is the world lowest operating voltage reported so far. Thus, the CT state, which is an essence of organic solar cell process, can be also utilized for up-converted organic EL.

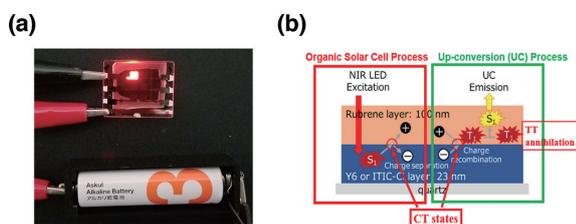


Figure 4. (a) Up-converted EL operated by a battery of 1.5 V. (b) Up-conversion (UC) mechanism.

2. Monolayer-Digitized Band Mapping for Doped Rubrene Single Crystals³⁾

The band mapping for doped rubrene single crystals by using Atomic/Kelvin force microscopy (AFM/KFM) was performed. The ppm-level doping technique using an ultra-

slow deposition (Figure 3) was used. Doped rubrene single crystal was obtained by the homoepitaxial growth on rubrene single crystal substrate. Mo₃O₉ (1,000 ppm) was used as an acceptor dopant. Thicknesses of doped homoepitaxial layers were varied from 0 to 20 nm.

Morphological and potential images of doped homoepitaxial rubrene are shown in Figures 5(a) and 5(b). The island image (AFM) (Figure 5(a)) and the simultaneously obtained potential image (KFM) (Figure 5(b)) are roughly coincided. The average work functions of 3 and 4 rubrene monolayers were determined to 4.18 and 4.22 eV, respectively. Thus, we could plot the relationship between work function and number of rubrene monolayer (Figure 5(c), blue dots).

For Mo₃O₉ (1,000 ppm) doped rubrene single crystal, the number of negatively ionized acceptors of $2.0 \times 10^{18} \text{ cm}^{-3}$ and the doping efficiency of 3.3% were calculated by the band mapping. Specific values of work function observed for each number of rubrene monolayer suggests that the built-in potentials are monolayer-digitized.

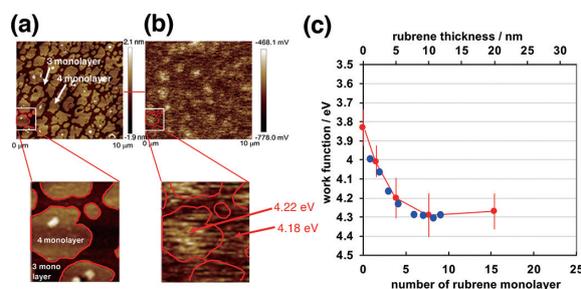


Figure 5. (a) Morphological image of Mo₃O₉ (1,000 ppm) doped homoepitaxial rubrene having the thickness of 5 nm. (b) Simultaneously obtained potential image. Magnified image (lower) of white rectangle area (upper) are also shown. Island shapes emphasized by red curves are overlaid in the potential image. (c) Monolayer-digitized band mapping.

References

- 1) S. Izawa, M. Morimoto, S. Naka and M. Hiramoto, *Adv. Opt. Mater.* **10**, 2101710 (8 pages) (2022).
- 2) S. Izawa and M. Hiramoto, *Nat. Photonics* **15**, 895–900 (2021).
- 3) M. Hiramoto, Y. Yabara, T. Minato and S. Izawa, *68th Spring meeting of Japan Society of Applied Physics*, 17p-Z23-2 (2021).

Awards

IZAWA, Seiichiro; NF Foundation R&D Encouragement Award (2021).

IZAWA, Seiichiro; The 11th Young Scientist Award of National Institutes of Natural Sciences (2022).

IZAWA, Seiichiro; The Morino Foundation for Molecular Science (2022).