





Public Relations Office, Kyushu University 744 Motooka Nishi-ku Fukuoka, 819-0395 TEL:092-802-2130 FAX:092-802-2139 MAIL:koho@jimu.kyushu-u.ac.jp URL:http://www.kyushu-u.ac.jp

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Exciton limits are meant to be broken: OLED surpasses 100% exciton production efficiency

Researchers at Kyushu University's Center for Organic Photonics and Electronics Research (OPERA) in Japan have demonstrated a way to split energy in organic light-emitting diodes (OLEDs) and surpass the 100% limit for exciton production, opening a promising new route for creating low-cost and high-intensity near-infrared light sources for sensing and communications applications.

OLEDs use layers of carbon-containing organic molecules to convert electrical charges into light. In normal OLEDs, one positive charge and one negative charge come together on a molecule to form a packet of energy called an exciton. One exciton can release its energy to create at most one beam of light, or photon.

When all charges form excitons that emit light, a maximum 100% internal quantum efficiency is achieved. However, the new technology uses a process called singlet fission to split the energy from an exciton into two, making it possible to exceed the 100% limit for the efficiency of converting charge pairs into excitons, also known as the exciton production efficiency

"Put simply, we incorporated molecules that act as change machines for excitons in OLEDs. Similar to a change machine that converts a \$10 bill into two \$5 bills, the molecules convert an expensive, high-energy exciton into two half-price, low-energy excitons," explains Hajime Nakanotani, associate professor at Kyushu University and co-author of the paper describing the new results.

Excitons come in two forms, singlets and triplets, and molecules can only receive singlets or triplets with certain energies. The researchers overcame the limit of one exciton per one pair of charges by using molecules that can accept a triplet exciton with an energy that is half of the energy of the molecule's singlet exciton.

In such molecules, the singlet can transfer half of its energy to a neighboring molecule while keeping half of the energy for itself, resulting in the creation of two triplets from one singlet. This process is called singlet fission.

The triplet excitons are then transferred to a second type of molecule that uses the energy to emit near-infrared light. In the present work, the researchers were able to convert the charge pairs into 100.8% triplets, indicating that 100% is no longer the limit. This is the first report of an OLED using singlet fission, though it has previously been observed in organic solar cells.

Furthermore, the researchers could easily evaluate the singlet fission efficiency, which is often difficult to estimate, based on comparison of the near-infrared emission and trace amounts of visible emission from remaining singlets when the device is exposed to various magnetic fields.

"Near-infrared light plays a key role in biological and medical applications along with communications technologies," says Chihaya Adachi, director of OPERA. "Now that we know singlet fission can be used in an OLED, we have a new path to potentially overcome the challenge of creating an efficient near-infrared OLED, which would find immediate practical use."

Overall efficiency is still relatively low in this early work because near-infrared emission from organic emitters is traditionally inefficient, and energy efficiency will, of course, always be limited to a maximum 100%. Nonetheless, this new method offers a way to increase efficiency and intensity without changing the emitter molecule, and the researchers are also looking into improving the emitter molecules themselves.

With further improvements, the researchers hope to get the exciton production efficiency up to 125%, which would be the next limit since electrical operation naturally leads to 25% singlets and 75% triplets. After that, they are considering ideas to convert triplets into singlets and possibly reach a quantum efficiency of 200%.

For more information about this research, see "<u>Exploiting singlet fission in organic light-emitting diodes</u>," Ryo Nagata, Hajime Nakanotani, William J. Potscavage Jr., and Chihaya Adachi, *Advanced Materials* (2018), DOI: 10.1002/adma.201801484.



Illustration of the singlet fission process used to boost the number of excitons in an OLED and break the 100% limit for exciton production efficiency. A singlet exciton, which is created when a positive charge and a negative charge combine on a rubrene molecule, can transfer half of its energy to a second rubrene molecule through the process of singlet fission, resulting in two triplet excitons. The triplet excitons then transfer to ErQ₃ molecules, and the exciton energy is released as near-infrared emission by ErQ₃.



Hajime Nakanotani (left), Ryo Nagata (center), and Chihaya Adachi (right) at Kyushu University's Center for Organic Photonics and Electronics Research (OPERA) reported a near-infrared OLED that uses singlet fission to boost the fraction of excitons created per pair of electrical charges to over 100%. Using the pictured electromagnetic, the researchers evaluated the efficiency of singlet fission based on changes in the OLED emission with different applied magnetic fields.

[Contact]

Chihaya Adachi, Director of OPERA

Hajime Nakanotani, Associate Professor

Center for Organic Photonics and Electronics Research (OPERA), Kyushu University

Tel: +81-92-802-6920/Fax: +81-92-802-6921

Email : nakanotani@cstf.kyushu-u.ac.jp