

Stretchy organic LED devices with an ‘exciplex’ state are highly efficient

Fully stretchable organic light-emitting diodes (OLEDs) have been limited by poor efficiency, in part because their elastic-polymer components hinder the harvesting of ‘triplet-state’ quasiparticles that can be converted into light-emitting states. Fully stretchable OLEDs with unprecedented efficiencies have been developed that recycle triplets in an elastomer-tolerant manner.

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The problem

Wearable displays designed for direct layering onto the skin demand fully stretchable components made of organic or polymer materials that keep their functional properties under mechanical strain¹. ‘Electroluminescent’ organic light-emitting diodes (OLEDs) emit light when an electric charge is injected and flows between two electrodes, but the efficiency of these devices is limited.

In these materials, electrons and their positively charged counterparts, called holes, recombine to form quasiparticles called excitons that, depending on their spin characteristics, can be classed as ‘singlets’ or ‘triplets’. In rigid fluorescent OLEDs, the ratio of singlets to triplets is 1:3, and the ratio of photons generated to the number of electrons injected (the external quantum efficiency; EQE) is theoretically limited to 5%. Harvesting non-radiative triplet excitons to convert them into radiative singlet excitons in stretchable fluorescent materials is a major challenge² (Fig. 1a).

Conventional strategies to enhance the stretchability of light-emitting materials include incorporating electronically insulating elastic polymers (elastomers) or flexible alkyl carbon-chain spacers. However, these negatively affect OLED function in various ways, such as by impeding exciton energy transfer in the light-emitting layer and hindering efforts to harvest triplets³.

Efficient charge injection under strain is also a challenge, and demands precise tuning of the electrical conductivity, mechanical compliance and alignment of energy bands in the active layer of stretchable electrodes⁴. These limitations have so far restricted the EQE of fully stretchable OLEDs⁵ to less than 10%.

The solution

To overcome long-standing limitations imposed by insulating elastomer matrices, we developed a stretchable phosphorescent layer that is assisted by charge-transfer complexes called exciplexes. This exciplex-assisted layer (ExciPh) comprises a phosphorescent ‘dopant’ compound (which contains a heavy-metal atom), co-hosted by electron-donating and electron-accepting materials, in an insulating elastomeric polyurethane matrix. The electron-donor and electron-acceptor co-host molecules form exciplexes that enable efficient conversion of triplets to singlets. Although short-range (within 1 nanometre) transfer of electrons between donors and acceptors is hindered by the insulating polyurethane

matrix, triplet excitons are converted to singlets in the exciplex host and the singlets are then transferred to the dopant through a longer-range transfer process called FRET (Förster resonance energy transfer; Fig. 1b). Overall, we call this process elastomer-tolerant triplet recycling. In conventional systems that lack exciplex formation, triplets are predominantly lost through non-radiative decay (Fig. 1a), but the exciplex-assisted mechanism in the polyurethane matrix enables ExciPh to harvest triplets effectively even under mechanical strain.

ExciPh films stayed crack-free when stretched to double their length, and demonstrated an EQE of 21.7% using the intrinsically stretchable light-emitting layer in rigid devices, surpassing the efficiency of previously reported stretchable emissive systems. To realize fully stretchable OLEDs, we integrated stretchable electrodes with contacts made from MXene (a 2D inorganic material). These have high mechanical durability and a tunable work function (the amount of energy needed to remove an electron) for electron and hole injection, and enable efficient charge injection into ExciPh. This architecture yielded fully stretchable OLEDs with a record-high EQE of 17% and negligible luminance loss under 60% tensile strain.

The implications

The intrinsic trade-off between mechanical stretchability and device efficiency remains a central challenge in the design of stretchable optoelectronic materials. Our system provides a strategy to overcome exciton energy-transfer limitations imposed by insulating elastomer matrices, establishing a versatile platform for stretchable semiconductors with broad applicability across diverse optoelectronic technologies.

Nonetheless, full commercialization of fully stretchable OLEDs remains an ambitious goal. Many key challenges persist, including long-term operational stability, mechanical durability under repeated deformation and, crucially, the development of a robust, fully stretchable encapsulation that can protect devices in real-world environments.

Future efforts will focus on enhancing device stability by integrating stable materials that simultaneously exhibit high stretchability and efficient charge transport. This should pave the way towards the practical deployment of next-generation stretchable optoelectronics.

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EXPERT OPINION

The authors have developed high-performance, fully stretchable OLEDs with an EQE of 21.7% and a stretchability exceeding 200%. The authors enhanced the efficiency of fully stretchable OLEDs by engineering the insulating elastomer matrix. The main strategy involves

enhancing the harvesting of triplets, a process facilitated by heavy-metal atoms in the phosphorescent materials, enabling high energy-transfer efficiency in the polyurethane matrix, even under strain.”

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FIGURE

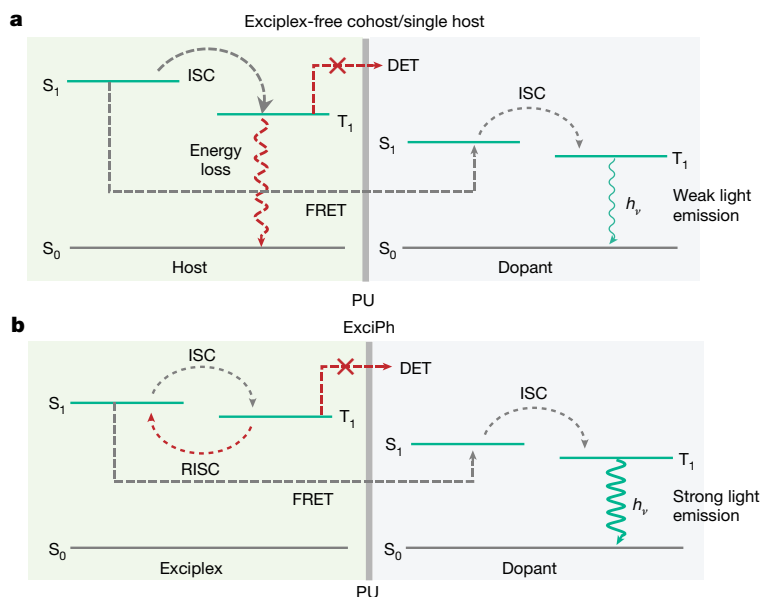


Figure 1 | Stretchable phosphorescent layer for highly efficient, fully stretchable organic light-emitting diodes (OLEDs). **a**, In conventional stretchy OLEDs, the recombination of electrons and their positively charged counterparts, called holes, in a host material forms exciton quasiparticles in a singlet (S_1) or triplet (T_1) state. Singlets are converted to triplets through a process called intersystem crossing (ISC). An elastic polyurethane (PU) matrix impedes the short-range transfer (DET) of excitons to a dopant compound, leading to triplet loss with weak light emission, and thus low OLED efficiency. **b**, Stretchable OLEDs containing a dopant compound that is co-hosted by materials that form an ‘exciplex’ (an intermolecular charge-transfer state) show much higher efficiency. Although DET is still impeded by the PU matrix, triplets are converted to singlets (through reverse intersystem crossing, or RISC), and are transferred to the dopant through a process called FRET (Förster resonance energy transfer). There, the triplets can emit light ($h\nu$). S_0 , ground state.

BEHIND THE PAPER

We started our stretchable-OLED research in 2018, building on our foundational work in flexible OLEDs. Our early efforts focused on fluorescent polymers that do not harvest triplets⁵; however, to improve device efficiency, we transitioned to phosphorescent emitters, which introduced other challenges related to triplet harvesting in the presence of insulating elastomers. This shift from fluorescent to phosphorescent emitters also imposed stricter demands on electrode design, because the light-emitting layers were thin (less than 50 nanometres) and so required exceptional surface uniformity and

low surface roughness for reliable operation. These accumulated challenges underscored a pivotal need for suitable stretchable electrodes and required the design of stretchable light-emitting layers through a framework grounded in photophysics and materials chemistry. Guided by mechanistic insights into device operation, we ultimately achieved high-efficiency devices.

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FROM THE EDITOR

The appeal of this paper is twofold. First, it shows how to combine two material platforms that are currently the focus of much attention for their application potential (the 2D electrically conducting MXenes and emissive organic semiconductors) in a way that harnesses the desirable functionalities of both. Second, it demonstrates that such a combination, when appropriately optimized, can yield substantial performance improvements for the target application — stretchable organic light-emitting diodes. **Karl Ziemelis**, Chief Applied and Physical Sciences Editor, *Nature*