Excitons and polaritons in singlet fission materials: Photophysics, photochemistry, and optoelectronics

Oksana Ostroverkhova,* Winston Goldthwaite, and Roshell Lamug

Organic (opto)electronic materials have been explored in a variety of applications in electronics and photonics, driven by several advantages over traditional silicon technology, including low-cost processing, fabrication of large-area flexible devices, and widely tunable properties through functionalization of the molecules. Over the past decade, remarkable progress has been achieved in understanding physical mechanisms and in developing guidelines for material design, which have boosted the performance of organic devices. However, further improvements in device performance are desirable, and challenges related to (photo)stability of organic devices need addressing. One of the major thrusts in developing new organic materials and device concepts has focused on materials exhibiting singlet fission, which is a charge-carrier multiplication process that could enable, for example, enhanced power-conversion efficiencies in solar cells. Nevertheless, fundamental questions pertaining to exciton physics in singlet fission materials, and how it can be manipulated by material design and external parameters, remain. Strong exciton–photon coupling that occurs when an organic film is placed in a microcavity, enabling formation of a light–matter hybrid state (polariton), presents a largely unexplored opportunity to control photophysics, photochemistry, and optoelectronic characteristics in singlet fission materials and devices using polaritons. In this article, we review the key requirements for singlet fission materials and promising advances toward controlling their properties using polaritons.

Introduction

Organic (opto)electronic materials are of interest due to their low-cost and tunable properties; a broad range of their applications, from photovoltaics to display technologies, has been demonstrated and commercialized.[1] Over the past decade, remarkable enhancement in performance of organic devices has been achieved, such as hole mobility of 20 $cm^2/(Vs)$ in thin-film transistors (TFTs) and solar-cell power-conversion efficiencies (PCEs) of more than 19% in single-junction devices.[2] Nevertheless, performance of organic devices has not yet reached fundamental limits, and innovative solutions are necessary for further improvements. One exciting area is singlet fission (SF)-based optoelectronics, a carrier multiplication process in which one absorbed photon generates two pairs of mobile charge carriers. This process enables PCEs that surpass the Shockley–Queisser theoretical limit on solar-cell efficiency.[3,4] However, in spite of much improved understanding of the SF mechanisms,[5] utilizing SF in devices has been challenging due to a number of constraints on the molecular energies and photophysics, which limits the choices for the successful systems. One promising way to mitigate some of these limitations is by utilizing light–matter hybrid states (polaritons) to modify excited state energies and dynamics toward relaxing these constraints.

Incorporating organic films into photonic or plasmonic structures to induce strong interactions between molecular excitons and cavity photons or plasmons that create polaritons (**Figure 1**b) has emerged as an innovative approach to enhancing organic device characteristics across various device platforms. Organic polaritonics has advanced toward practical uses with demonstrations of polariton lasing, nonlinear polariton–polariton interactions, and polariton electroluminescence, as well as revealed fascinating physical phenomenon of polariton Bose–Einstein condensation (BEC).[6] From the fundamental physics standpoint, strong light–matter coupling signifies a regime where coherence

Oksana Ostroverkhova, Department of Physics, Oregon State University, Corvallis, USA; oksana@science.oregonstate.edu
Winston Goldthwaite, Department of Physics, Oregon State University, Corvallis, USA; goldthww@oregonstate.edu
Roshell Lamug, Department of Physics, Oregon State University, Corvallis, USA; lamugr@oregonstate.edu
*Corresponding author

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plays an important role. Polariton states have been demonstrated to possess high spatial and spatiotemporal coherence, even in disordered amorphous films, and to propagate over tens of microns distances. Polariton-mediated long-range energy transfer, charge transfer (CT), and SF yield and rates are desirable for organic optoelectronics, but implementation of these processes in devices is at its inception. Although several polariton-assisted optoelectronic devices have been demonstrated, mechanisms that govern

polariton-mediated processes and how they are supported by material design are not well understood and are a subject of current research.

Another critical obstacle for commercializing organic electronics is the limited (photo)stability of organic materials. Polariton chemistry is an emerging area that aims to control (photo)chemical reactions, offering a way to enhance both performance and stability of organic (opto)electronics. In this article, we review ongoing efforts that could
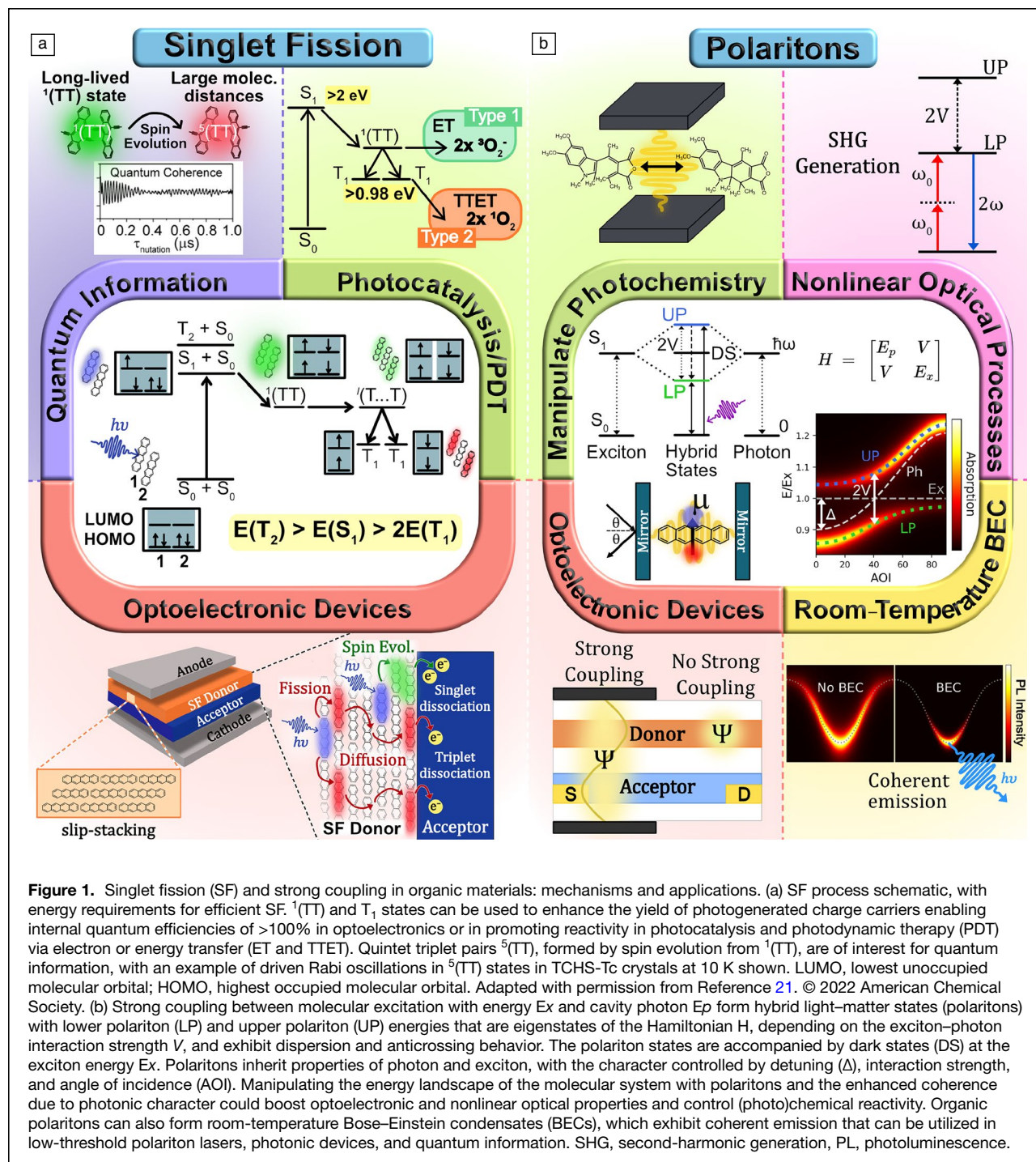

Figure 1. Singlet fission (SF) and strong coupling in organic materials: mechanisms and applications. (a) SF process schematic, with energy requirements for efficient SF. ^{1}(TT) and T_1 states can be used to enhance the yield of photogenerated charge carriers enabling internal quantum efficiencies of >100% in optoelectronics or in promoting reactivity in photocatalysis and photodynamic therapy (PDT) via electron or energy transfer (ET and TTET). Quintet triplet pairs ^{5}(TT), formed by spin evolution from ^{1}(TT), are of interest for quantum information, with an example of driven Rabi oscillations in ^{5}(TT) states in TCHS-Tc crystals at 10 K shown. LUMO, lowest unoccupied molecular orbital; HOMO, highest occupied molecular orbital. Adapted with permission from Reference 21. © 2022 American Chemical Society. (b) Strong coupling between molecular excitation with energy Ex and cavity photon Ep form hybrid light–matter states (polaritons) with lower polariton (LP) and upper polariton (UP) energies that are eigenstates of the Hamiltonian H, depending on the exciton–photon interaction strength V, and exhibit dispersion and anticrossing behavior. The polariton states are accompanied by dark states (DS) at the exciton energy Ex. Polaritons inherit properties of photon and exciton, with the character controlled by detuning (Δ), interaction strength, and angle of incidence (AOI). Manipulating the energy landscape of the molecular system with polaritons and the enhanced coherence due to photonic character could boost optoelectronic and nonlinear optical properties and control (photo)chemical reactivity. Organic polaritons can also form room-temperature Bose–Einstein condensates (BECs), which exhibit coherent emission that can be utilized in low-threshold polariton lasers, photonic devices, and quantum information. SHG, second-harmonic generation, PL, photoluminescence.

enhance the performance of existing SF materials and inspire novel material and device designs using polariton-enhanced processes.

SF: Processes, applications, challenges, and implications for photostability

SF

The SF process has been known since the 1960s, but it has attracted broad attention only in the 2000s when its potential in enhancing solar energy harvesting has been realized. SF starts by photoexcitation of one molecule to its S_1 singlet state, which interacts with a nearby molecule in the ground-state S_0 to form an entangled triplet pair state $^1(TT)$, serving as an intermediate state in SF (Figure 1a).[5] The $^1(TT)$ state then evolves into a more separated and spin-mixed state with contributions from $^l(TT)/^l(T…T)$ states ($l=1,3,5$) followed by formation of two free triplet excitons T_1. This is a spin-allowed process that occurs on femto- to nanosecond time scales. Efficient SF requires the energy of the singlet state $E(S_1)$ to be close to double the triplet state's energy $2E(T_1)$, with two scenarios: exothermic ($E(S_1)>2E(T_1)$) and endothermic ($E(S_1)\leq2E(T_1)$). The mechanisms and molecular designs that impact formation of the $^1(TT)$ states, their properties, and their evolution toward spatially separated $^l(T…T)$ states and splitting into uncorrelated T_1 excitons are still under debate.[5] Importantly, yield and dynamics of TT/T…T states depend on the energy difference $E(S_1)-2E(T_1)$ and entropic gain $\Delta S_{S1\to TT}=k_B \ln(\Omega_{TT}/\Omega_{S1})$ and $\Delta S_{TT\to T…T}=k_B \ln(\Omega_{T…T}/\Omega_{TT})$ (where k_B is the Boltzmann constant and Ω_i is the number of microstates available to the ith state),[7] which are dictated by molecular structure and packing. In exothermic materials, the energy difference $E(S_1)-2E(T_1)$ should be within ~0.1 eV to minimize energy dissipation via heat and competing processes. To prevent higher-energy triplet states via triplet–triplet annihilation, the energy of the higher triplet states ($E(T_2)$) must exceed twice the energy of the lowest lying triplet state: $E(T_2)\geq E(2T_1)$. Finally, to impede intersystem crossing (which creates only one T_1 triplet state from S_1), it is desirable to have $E(T_2)>E(S_1)$. Thus, the criterion for a promising SF chromophore can be summarized as $E(T_2)>E(S_1)\gtrsim 2E(T_1)$ (Figure 1a). Examples of classes of molecules meeting energy requirements for SF include acenes (such as pentacene (Pn), tetracene (Tc), and functionalized Pn or Tc [e.g., R-Pn or R-Tc with R = TIPS or TCHS]), rylenes and rylene diimides (such as perylene diimide [PDI]), biradicaloids, carotenoids, diketopyr-rolopyrroles, indigoids, and donor–acceptor polymers) (**Figure** 2a). Recent computational studies searching for molecular structures that satisfy the energy criteria revealed additional promising materials, with some examples shown in Figure 2a, which are awaiting synthesis and characterization.[8–10] Universal design principles for optimizing molecular packing for efficient SF are still under debate due to an intricate interplay between packing-dependent electronic coupling and nuclear dynamics, which impacts all processes participating in the

SF—$^1(TT)$ formation and $^1(TT)$ separation and spin relaxation into the $^l(T…T)$ and T_1+T_1—as well as the efficiency of competitive pathways (such as excimer formation) in an interconnected way. Nevertheless, slip-stacked geometries with 3–5 Å π-stacking distances and 3–6 Å longitudinal displacement have exhibited efficient SF in several material classes, with up to 200% yield of triplet states. The formation and dissociation of $^1(TT)$ states can also depend on external parameters such as excitation wavelength, temperature, and magnetic field, which are used in systematic studies of these processes and could potentially be utilized in applications.

SF-based optoelectronic devices

SF-based device implementations and quantitative assessment of the triplet-enhanced performance remain a challenge due to additional requirements for the materials to ensure that SF-produced triplet pairs can dissociate and enhance the photocurrent.[4] Nevertheless, several implementations have been reported with a principle of operation relying on SF-produced triplet excitons as a source of enhanced yield of (1) charge carriers via dissociation of triplet excitons and (2) down-converted photons via SF-photon multiplication (SF-PM) process that are then absorbed by a material capable of converting photons into charge carriers,[3,4,7] with photocurrent enhancement in both cases. Both device types rely on either charge or energy transfer from SF-produced triplet states in a SF donor to an acceptor material (Figure 2b). This imposes constraints on the design of donor–acceptor (D–A) combinations, due to the low energies of triplet states (which then require low LUMO energies of the acceptor to promote, for example, charge transfer from donor to acceptor, Figure 2d), and device geometries depending on the system, examples of which are given next and summarized in Figure 2.

All-organic SF donor–acceptor devices

In all-organic devices, SF donor molecule (e.g., pentacene [Pn] or tetracene [Tc]) is combined with an acceptor molecule (such as a fullerene or boron subphthalocyanine) to form either a planar heterojunction (HJ) or bulk heterojunction (BHJ) (Figure 2).[4] In both cases, the SF-generated triplet excitons diffuse to the heterojunction, followed by formation and dissociation of CT states into mobile charge carriers followed by their collection at the electrodes. The internal quantum efficiency (IQE) (the number of collected electron–hole pairs per absorbed photon × 100%) of >100% was observed in several D–A systems (e.g., IQE = 160% was obtained in Pn/C_{60} planar HJ devices).[11] Planar HJs rely on efficient triplet diffusion to the interface, limiting the SF donor layer thickness and imposing requirements of high absorption and high SF rates. BHJs improve charge-separation efficiency by providing large-area D–A interface, but suffer from triplet exciton-charge annihilation losses that limit IQE.[12] In both planar HJ and BHJ, CT from the singlet state S_1 is undesirable as it competes with SF, whereas desirable CT from T_1 can be five orders of magnitude
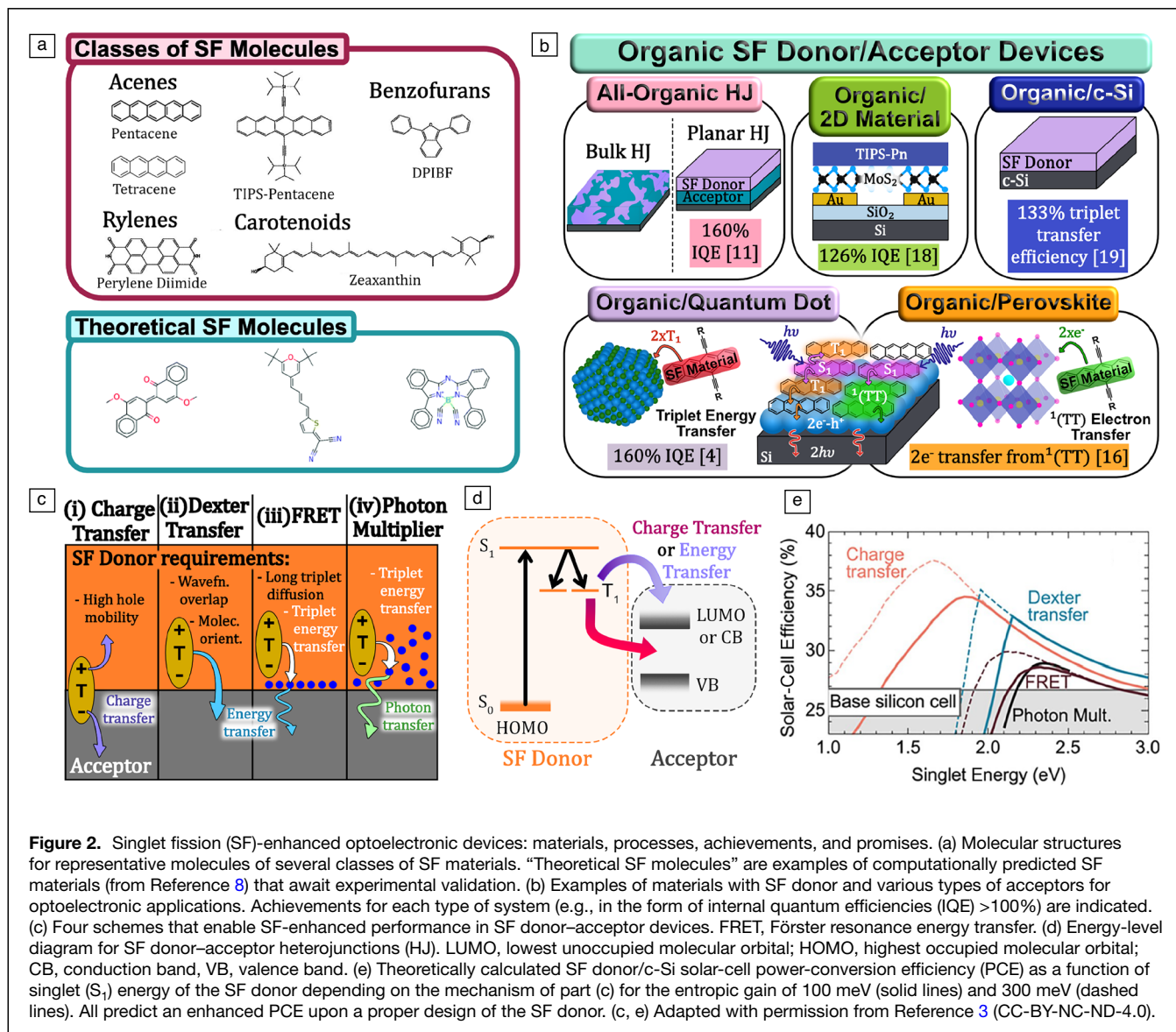
Figure 2. Singlet fission (SF)-enhanced optoelectronic devices: materials, processes, achievements, and promises. (a) Molecular structures for representative molecules of several classes of SF materials. "Theoretical SF molecules" are examples of computationally predicted SF materials (from Reference 8) that await experimental validation. (b) Examples of materials with SF donor and various types of acceptors for optoelectronic applications. Achievements for each type of system (e.g., in the form of internal quantum efficiencies (IQE) >100%) are indicated. (c) Four schemes that enable SF-enhanced performance in SF donor–acceptor devices. FRET, Förster resonance energy transfer. (d) Energy-level diagram for SF donor–acceptor heterojunctions (HJ). LUMO, lowest unoccupied molecular orbital; HOMO, highest occupied molecular orbital; CB, conduction band, VB, valence band. (e) Theoretically calculated SF donor/c-Si solar-cell power-conversion efficiency (PCE) as a function of singlet (S_1) energy of the SF donor depending on the mechanism of part (c) for the entropic gain of 100 meV (solid lines) and 300 meV (dashed lines). All predict an enhanced PCE upon a proper design of the SF donor. (c, e) Adapted with permission from Reference 3 (CC-BY-NC-ND-4.0).

slower than that from S_1 even with the optimized CT driving force (ΔG).[13] A promising line of inquiry is to utilize a two-electron ($2e^-$) transfer directly from the ^{1}(TT) state of the SF donor to the acceptor, which circumvents slow CT from T_1. This process has been observed in several acene/fullerene systems,[14] with current efforts aiming to develop molecular design principles to maximize efficiency of the $2e^-$ transfer process.[15]

SF donor with quantum dots or perovskite acceptors

With a principle of operation similar to that in all-organic devices, organic SF donor/quantum dot (QD) acceptor solar cells have been demonstrated. For example, in TIPS-Pn/PbS QD solar cells, IQE of 160% was achieved, although the overall PCE was only 4.8 percent.[4] In a SF donor-perovskite acceptor system TIPS-Pn/MAPbI$_3$, spectroscopic studies revealed the $2e^-$ transfer

from the ^{1}(TT) state of TIPS-Pn to MAPbI$_3$.[16] However, SF-sensitized perovskite devices have not yet been reported, and the need for increasing electronic interaction at the organic/perovskite interface (e.g., by forming chemical bonds) was emphasized to enhance triplet transfer across the interface.[17]

SF donor with 2D material acceptors

An example of such device is a SF donor (<20-nm crystalline TIPS-Pn) layer on MoS$_2$ monolayer incorporated in a photoTFT structure (Figure 2b).[18] Here, the triplet states T_1 are produced via SF at sub-100 fs time scales, which then undergo electron transfer from TIPS-Pn to MoS$_2$ in tens to hundreds of picoseconds. SF competes with ultrafast electron transfer from an excited singlet state S_1 of TIPS-Pn to MoS$_2$, which is the major source of losses, preventing 1/3 of all generated singlets from undergoing SF, and needs to be mitigated. Nevertheless,

IQE of 126% was reported and 150% was predicted by simulations when the SF layer thickness is optimized.

SF donor with c-Si acceptor

There is considerable interest in combining SF donors with crystalline silicon (c-Si) to enhance c-Si solar-cell performance. Four schemes of interfacing a SF donor with c-Si have been identified (Figure 2c),[3] each relying on different processes and imposing different constraints on the SF donor: (i) electron transfer from T_1 to Si, which relies on efficient transport of holes in the SF donor, thus requiring high-mobility SF material; (ii) Dexter energy transfer from SF donor to Si, where the triplet exciton (i.e., both hole and electron) transfers to Si, requiring efficient triplet diffusion in the SF donor and strong overlap of the donor and Si wave functions; (iii) triplet energy transfer from T_1 of the SF donor to a QD, which subsequently transfers energy via Förster resonant energy transfer (FRET) into Si, requiring long triplet diffusion in the SF donor and appropriate energy matching for the SF donor, QDs, and Si; and (iv) SF-PM process previously mentioned, where T_1 energy from the SF donor is transferred to a QD, which then emits a near-IR photon that is absorbed by Si. In all schemes, the most efficient SF donor would also have a strong entropic gain and broad absorption, with S_1 energies in the 1.85 eV (scheme [i])− 2.34 eV (scheme [iv]) range assuming 100-meV entropic gain. With such entropic gain, for a base c-Si solar cell with a~27% PCE, the predicted SF donor/c-Si efficiencies are 29% (schemes [iii] and [iv]), 33% (scheme [ii]), and 35% (scheme [i]). With a 300-meV entropic gain, these increase to 30%, 35%, and 38% (Figure 2e).[3] Entropic gains of 200 meV and 220 meV were obtained in Tc and PDI, respectively, which highlights the potential for derivatives of these materials to sensitize c-Si solar cells. Indeed, a 133% efficiency of triplet transfer from Tc to c-Si was observed via scheme (2), which is the most successful c-Si SF-sensitization to date.[19] Similar to organic-perovskite HJs, one of the critical factors that determines the device efficiency is the quality of the organic–Si interface.

SF-based quantum information

The SF-produced entangled triplet pairs, with four quantum entangled spins, can potentially be used as molecular qubits in quantum information applications. Unlike SF-based photovoltaics, in which the majority of experimental realizations rely on fast dissociation of ^{1}(TT) states into uncorrelated triplets T_1, these qubits require long-lived ^{1}(TT) states to favor spin evolution into quintet triplet pairs ^{5}(TT) before spin decoherence. The quintet states ^{5}(TT) fulfill the DiVincenzo criteria for a viable qubit:[20] they can be reproducibly generated by optical pumping of a scalable physical system, have long decoherence times, and can be controlled with pulsed microwaves that drive quantum gate operations. Current effort is in design of molecular structure and packing that supports efficient ^{1}(TT) formation, but minimizes triplet–triplet annihilation and triplet diffusion. In TCHS-Tc crystals, bulky TCHS side groups prevent tight packing of the Tc cores, resulting

in weak intermolecular coupling and suppressed triplet diffusion; coherence lifetimes T_2 of up to 10 μs, and population lifetimes T_{pop} of up to 130 μs were observed at 10 K and 5 K, respectively, and T_{pop} of 87 ns was obtained at room temperature.[21] An example of driven Rabi oscillations in ^{5}(TT) states in TCHS-Tc crystals at 10 K is shown on the upper left of Figure 1a; the spin-flip time, which is the time needed to perform a logic gate operation that flips the spin between the $m_s = 0$ and $m_s = \pm 1$ levels of ^{5}(TT), is ~11 ns, enabling >270 spin-flip operations before the decoherence occurs. The Rabi frequencies of ~50 MHz achieved in this model SF system are comparable to those achieved in hBN with spin defects; the microsecond spin coherence times are also considerably longer than, for example, those in perovskite QDs (<100 ps).[22] This highlights the potential of SF-based organic materials as an important class of quantum materials.

SF-based photocatalysis and photodynamic therapy

Another potential application of SF materials is in photosensitizing the generation of reactive oxygen species such as singlet oxygen $^1O_2$ and superoxide radical O^{2-} via Type II and Type I processes, respectively (Figure 1a). In the Type II process, the SF-generated triplets transfer energy to ground-state oxygen ($^3O_2$) to produce $^1O_2$. The process is efficient if the triplet state energy of the SF material is above the $^3O_2 - {}^1O_2$ energy gap of 0.98 eV. Prominent examples of such SF materials are Tc derivatives and covalently linked Tc dimers ($E(T_1) > 1.1$ eV); a $^1O_2$ yield of 148% was recently reported with a TIPS-Tc dimer linked with a biphenyl bridge as a photosensitizer.[23] The Type I process proceeds via electron transfer to the ground-state oxygen to form reactive superoxide O^{2-} (e.g., from the S_1 singlet state of the photosensitizer). Interestingly, in TIPS-Tc dimers, such electron transfer involves the ^{1}(TT) state instead of S_1, which highlights importance of these triplet pairs in chemical reactivity.[23] Both SF-generated $^1O_2$ and O^{2-} reactive species have been shown to catalyze the photooxidation reactions,[23] and SF-boosted superoxide O^{2-} formation enabled photodynamic therapy (PDT) treatment of tumors in mice.[24] The requirement of triplet energy $E(T_1) > 0.98$ eV for the SF photosensitizer to efficiently generate $^1O_2$ via Type II process also dictates that for efficient SF, the singlet energy $E(S_1)$ must be at $\gtrsim 2$ eV ($\lesssim 620$ nm). This limits its use in PDT for deep tumor treatment (which requires near-IR absorption in the 650–1350-nm biological transparency window). However, the energy requirement is less stringent for the Type I process, making SF materials with $E(S_1) < 1.9$ eV (>650 nm) and efficient two-electron transfer from the ^{1}(TT) states promising candidates for PDT photosensitizers.[24]

Photostability of SF materials

Relatively low (photo)stability of organic molecules is a key issue limiting their applications. Because of high triplet exciton densities in SF materials, they could be particularly vulnerable to singlet oxygen ($^1O_2$) generation via the Type II process discussed above, followed by the singlet oxygen causing chemical reaction leading to degradation. In acene derivatives

(Figure 2a), such reaction leads to the formation of endoperoxides (EPOs) which degrades (opto)electronic properties of the material and thus needs mitigation. In the absence of oxygen, acenes are considerably more stable, and the leading photodegradation pathway is photodimerization that, in addition to excited state dynamics, relies on the proximity of two molecules to form chemical bonds. This highlights the importance of considering both photochemical and photophysics aspects when designing a SF material, balancing the need for molecular proximity to form ^{1}(TT) states with the avoidance of tightly packed structures prone to photodimer formation.[25]

Strong light–matter coupling

Polariton formation

When a molecular system with a strong exciton resonance is placed in, for example, an optical microcavity, the exciton and photon mode can exchange energy at a rate faster than the decay rates of the exciton and the photon, enabling formation of hybrid light–matter states known as polaritons (Figure 1b). In organic materials, excitons are tightly bound and long lived, making them excellent candidates for room-temperature polaritonic devices. Polaritons are quasi-particles with dispersion characteristics consisting of anticrossed lower- and upper polariton branches (LP and UP, respectively) separated in energy by the Rabi splitting ($\hbar\Omega_R \approx 2V$, where V is the exciton–photon interaction energy, Figure 1b). The polariton character depends on detuning between the exciton and photon ($\Delta = E_p - E_x$, where E_p and E_x are cavity photon energy at normal incidence, or AOI = 0, and exciton energy, respectively); for example, at AOI = 0 and red detuning ($\Delta < 0$), the LP is more photonic and UP is more excitonic. Polaritons possess interesting physical phenomena (such as BEC and superfluidity) and enable novel applications, such as low-threshold polariton lasers, all-optical switches, and quantum information processing. Importantly, polaritons may alter the photophysics of "bare" (i.e., in the absence of strong coupling) molecules, potentially enabling control over processes, such as SF, energy transfer, and CT in (opto)electronic devices. However, the extent of such control and its utility in devices are currently under debate.

Dark states

For a molecule with one exciton resonance coupled to a single-mode microcavity, the interaction energy is $V = \mu \sqrt{\hbar\omega/2\varepsilon V_m}$ (where μ is the transition dipole moment of the molecule, $\hbar\omega$ is the photon energy, ε is the dielectric permittivity of the cavity-filling material, and V_m is the photon mode volume). However, for a microcavity containing an ensemble of N such molecules, the interaction energy V scales as $\sim\sqrt{N}$, enhancing the Rabi splitting. This system has $N+1$ degrees of freedom, resulting in two "bright" polariton states (UP and LP), and N-1 "dark" states decoupled from the light field, which form a long-lived exciton reservoir at the same energy as the uncoupled (bare) exciton states.[26] These are called dark states because they do not interact with the light field and as such optical transitions are forbidden, but they play a significant role in polariton relaxation dynamics, mediating decay to the LP state. In the presence of energetic disorder, the dark states become "gray" (or "subradiant") states, which are mixed exciton–photon states with varying photonic character, allowing them to be weakly optically active (**Figure 3**b).[26] These dark and gray states acquire properties of delocalized polariton states and their participation in promoting long-range energy transfer has been a subject of several investigations.[27,28] High density of dark states, however, may obscure polariton effects on both photophysics and photochemistry, and therefore, their contributions to the excited state dynamics have to be accounted for or avoided by the experimental design.[29]

Materials for strong coupling

Early research on organic polaritons mainly focused on J-aggregated dye molecules (which possessed characteristics important for achieving strong coupling: high oscillator strength, narrow absorption, and low Stokes shift) dispersed in a polymer matrix. More recently, however, the potential in utilizing strong coupling not only for photonic applications, but also for enhancing optoelectronic properties and controlling chemical reactions has been realized and so investigations into strong coupling were expanded to other classes of organic materials (such as organic semiconductors and photosynthetic systems), including SF materials.[25,30–35]

Strong coupling geometries

Interactions between molecular excitons and photonic or plasmonic states leading to polariton formation have been observed in various geometries (Figure 3a), including microcavities (with metal or dielectric Bragg reflector (DBR) mirrors), plasmonic nanostructures, HMM (hyperbolic metamaterial), dielectric slabs or microrings, and even in simpler, external cavity-free systems such as organic films on c-Si and ordered molecular assemblies. The cavity-free geometries of generating polaritons are especially attractive for implementation in organic devices, and simpler designs are important for scalability.

Polariton-mediated processes of interest for optoelectronic devices

Energy transfer in donor–acceptor systems

When different species (e.g., a donor and an acceptor molecule) simultaneously couple to the same cavity mode and undergo optically driven mixing, a variety of polaritonic states is formed. These entangled states provide an effective path for hybridization of different excitons in the molecular systems[36] and long-range energy transfer. The latter, extending to mesoscopic (micron-length) spatial scales (considerably beyond typical FRET length scales of <10 nm), has
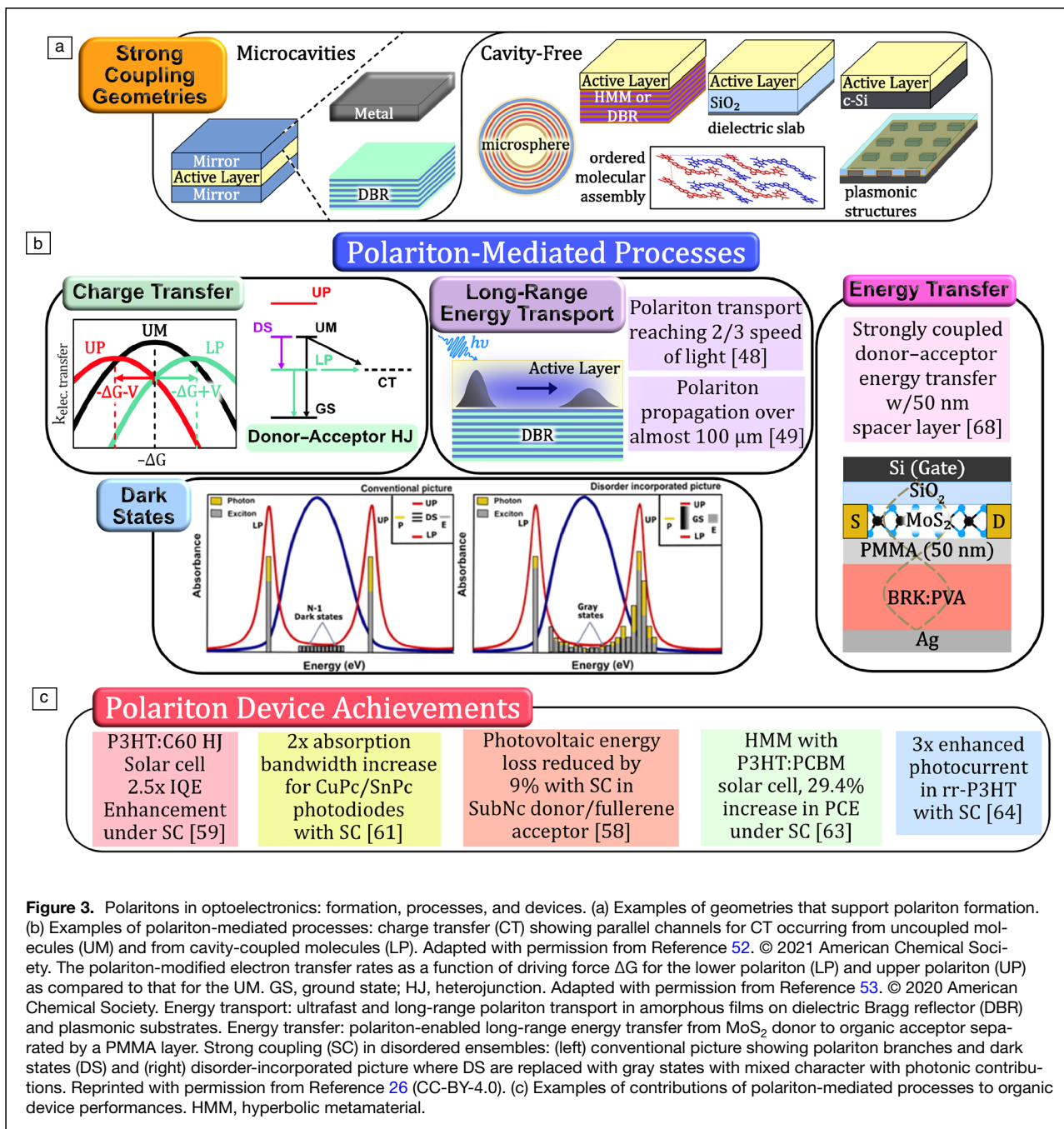
Figure 3. Polaritons in optoelectronics: formation, processes, and devices. (a) Examples of geometries that support polariton formation. (b) Examples of polariton-mediated processes: charge transfer (CT) showing parallel channels for CT occurring from uncoupled molecules (UM) and from cavity-coupled molecules (LP). Adapted with permission from Reference 52. © 2021 American Chemical Society. The polariton-modified electron transfer rates as a function of driving force ΔG for the lower polariton (LP) and upper polariton (UP) as compared to that for the UM. GS, ground state; HJ, heterojunction. Adapted with permission from Reference 53. © 2020 American Chemical Society. Energy transport: ultrafast and long-range polariton transport in amorphous films on dielectric Bragg reflector (DBR) and plasmonic substrates. Energy transfer: polariton-enabled long-range energy transfer from MoS$_2$ donor to organic acceptor separated by a PMMA layer. Strong coupling (SC) in disordered ensembles: (left) conventional picture showing polariton branches and dark states (DS) and (right) disorder-incorporated picture where DS are replaced with gray states with mixed character with photonic contributions. Reprinted with permission from Reference 26 (CC-BY-4.0). (c) Examples of contributions of polariton-mediated processes to organic device performances. HMM, hyperbolic metamaterial.

been demonstrated in several D–A systems using donor and acceptor layers separated by an inert spacer layer[37,38] in microcavities. In contrast to FRET, the polariton-assisted energy transfer does not depend on the distance if the strong coupling regime is achieved. Fundamentally, this behavior is rooted in the delocalization of polaritons, which results in enhancement, suppression, or even reversal (i.e., from acceptor to donor) of the energy transfer depending on whether the donor and/or acceptor molecules are coupled to the cavity photon.[27] Additional control over the energy flow can be achieved using photochromic donor molecules that undergo reversible photoisomerization, so that their absorption and emission properties reversibly switched by UV or visible light exposure. In this case, the long-range energy transfer can be turned "on" and "off" with light, which serves as an external gate.[39]

Long-range energy transport in pristine materials

Theoretical studies of polariton-assisted energy transport have predicted disorder immune,[40] dark states controlled,[41] and Q-factor-controlled[42] propagation. Experimentally, a combined real space and momentum (k-) space imaging has demonstrated energy transport over tens of microns using steady-state (e.g., PL imaging[43,44]) and time-resolved techniques (from ultrafast transient absorption or scattering microscopy[45,46] to ultrafast electron microscopy[47]). The PL imaging revealed polariton propagation lengths greater than 100 μm in an organic amorphous film on a DBR substrate (Figure 3b).[43] The time-resolved techniques probe not only the transport range, but also spatiotemporal dynamics of polariton transport, particularly in its coherent regime. These studies revealed long-range ballistic transport even for polaritons with large excitonic character[46] and cavity Q-factor-dependent propagation velocities.[28] Moreover, ballistic polariton transport with the velocity reaching 2/3 of the speed of light was demonstrated in organic films on a DBR substrate.[48] Toward utilizing these properties in devices, polariton propagation over almost 100 μm, coupled with an organic D–A HJ, was used in a photodetector that mimics a natural light-harvesting system.[49]

CT

Similar to energy transfer, polariton-mediated long-distance CT[50] and disorder-assisted charge transport[41] were also theoretically predicted. The polariton mediation of photo-induced CT and of related processes, such as charge-carrier photogeneration, has recently emerged as a promising direction[51,52] for manipulating CT rates and charge photogeneration mechanisms with cavity detuning and excitation wavelengths.[52] In particular, the CT rates from states modified by the presence of LP and UP polaritons can be enhanced or suppressed depending on the driving force (ΔG) and excitation (Figure 3b).[53] Additionally, a proposed mechanism involving second hybridization between polariton and dark states[54] could enhance ultrafast charge photogeneration by 50% under strong coupling conditions as compared to the "bare" molecular system.

Mitigation of unproductive states and disorder

Organic materials are prone to disorder and formation of long-lived excited states such as excimers, which serve as traps, potentially reducing SF efficiency by providing an alternative energy relaxation channel (**Figure** 4) and by promoting photodegradation. Because polariton formation modifies the energy landscape of the molecular system, it alters the excited state relaxation pathways, which can be used for manipulating photophysical and photochemical properties. Suppression of emission from trap excimer states due to excimer-LP energy transfer was observed in microcavities.[55] Enhanced photostability due to fast depopulation of states participating in processes leading to photodegradation via LP was also reported.[56,57] Theoretically predicted mitigation of negative impact of disorder on energy and charge transport[40,41] with polaritons is also a promising way of relaxing materials requirements for achieving high performance. Several experimental studies observed significant reduction in Urbach energy (which is a measure for energetic disorder) in strongly coupled organic solar cells;[58,59] however, clear design guidelines for systems efficiently mitigating disorder with polaritons have not yet been established.

Organic polaritonic devices

Several types of organic devices relying on polariton-modified optical absorption and emission spectra have been demonstrated, enabling tunable narrow emission in OLEFETs,[60] extended or enhanced wavelength responsivity in photodiodes,[61,62] and reduced losses in planar HJ solar cells.[58] However, development of polaritonic devices that efficiently utilize coherence of polariton states to promote energy or charge transfer is in its initial stage and polariton mediation of optoelectronic devices relying on the SF has not yet been demonstrated. Nevertheless, the polariton mediation of processes not directly related to the SF (such as energy transfer) is relevant for the SF-based devices as well. Selected examples of incorporating polaritons in (opto) electronic devices for mediating these processes are shown in Figure 3 and discussed below.

Solar cells

When benchmark organic photovoltaic polymer donor/fullerene acceptor (P3HT/C_{60}) planar HJ was incorporated in an all-metal microcavity under strong coupling conditions for the P3HT donor, up to a factor of 2.5 enhancement in IQE was observed at polariton energies.[59] The IQE enhancement factor increased with P3HT thickness and cavity Q-factor and was explained by efficient energy transport to the D/A interface facilitated by resonantly pumped polaritons. This study also noted signatures of lower disorder in the system via steepening of the absorption edge (reduction in Urbach energy), which was related to delocalized nature of polaritons and enhanced exciton–polariton diffusion.[59] Similar reduction in Urbach energy, as well as lowering of the driving force for electron transfer were observed in subphthalocyanine-based planar HJ organic solar cells with donor molecules strongly coupled to microcavities. However, this did not yield higher PCE than that from a reference cell without cavity, and no polariton-enhanced diffusion or delocalization were invoked.[58] Strong coupling was also explored in a benchmark organic D/A BHJ (P3HT:PCBM) on HMM substrates containing four Au(25 nm)/MoO_3(7 nm) pairs[63] where a ~11% enhancement in PCE was observed as compared to the reference cell with a fully reflective electrode instead of the HMM and a ~29% enhancement as compared to the cell with one Au/MoO_3 pair. The enhancement was attributed to high-k bulk plasmon polaritons contributing to enhanced exciton dissociation and charge transport at the organic/HMM interface.
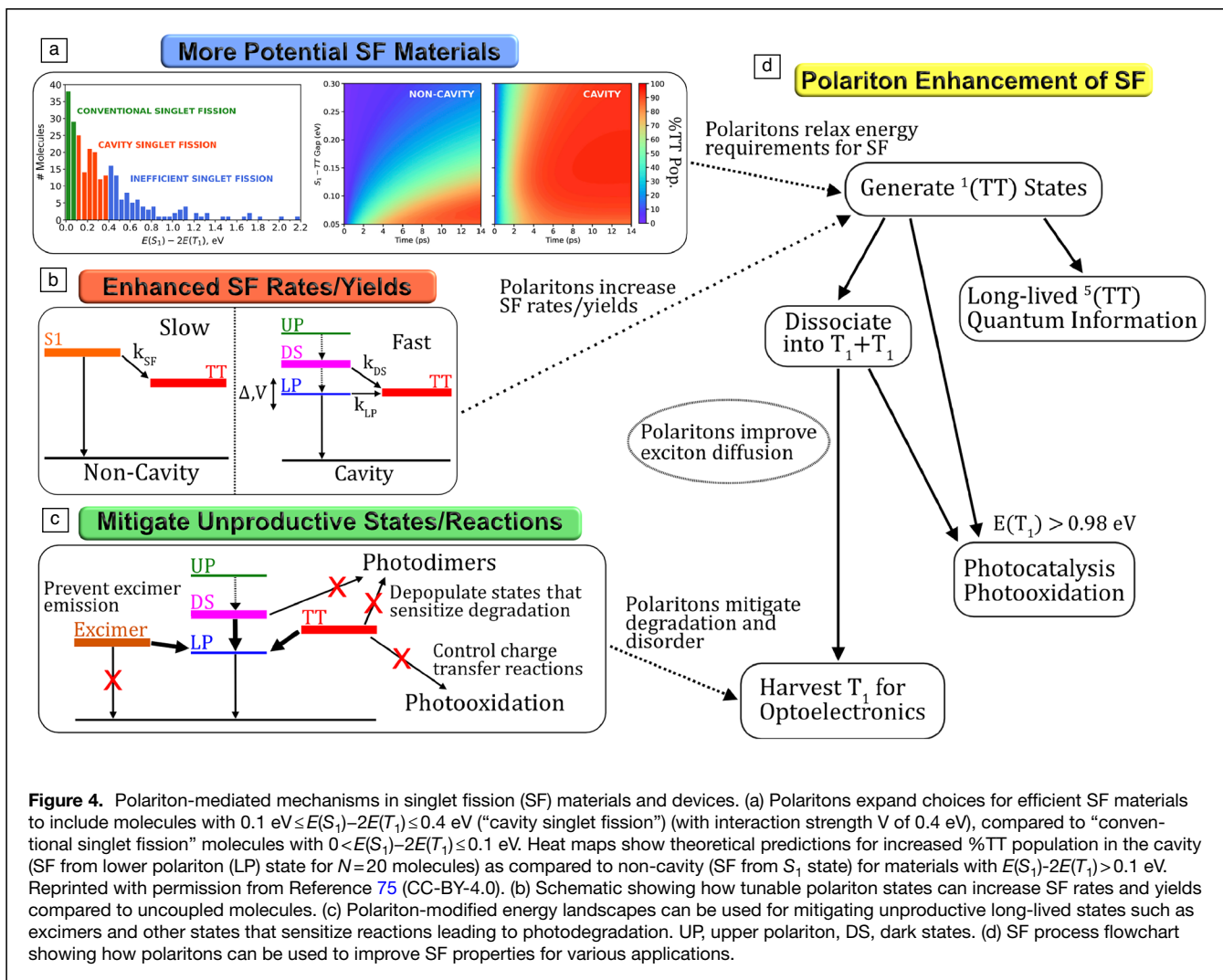
Figure 4. Polariton-mediated mechanisms in singlet fission (SF) materials and devices. (a) Polaritons expand choices for efficient SF materials to include molecules with $0.1\ eV \leq E(S_1)-2E(T_1) \leq 0.4\ eV$ ("cavity singlet fission") (with interaction strength V of 0.4 eV), compared to "conventional singlet fission" molecules with $0 < E(S_1)-2E(T_1) \leq 0.1\ eV$. Heat maps show theoretical predictions for increased %TT population in the cavity (SF from lower polariton (LP) state for $N = 20$ molecules) as compared to non-cavity (SF from S_1 state) for materials with $E(S_1)-2E(T_1) > 0.1\ eV$. Reprinted with permission from Reference 75 (CC-BY-4.0). (b) Schematic showing how tunable polariton states can increase SF rates and yields compared to uncoupled molecules. (c) Polariton-modified energy landscapes can be used for mitigating unproductive long-lived states such as excimers and other states that sensitize reactions leading to photodegradation. UP, upper polariton, DS, dark states. (d) SF process flowchart showing how polaritons can be used to improve SF properties for various applications.

(Photo)TFTs and other photoconductivity-based devices

Whether coherent ground-state charge transport can be achieved with polaritons is a subject of debate, with some studies reporting enhanced charge-carrier mobilities under strong coupling conditions[64–66] while others reporting no change.[60,67] However, there is agreement that polariton-mediated long-range energy transport,[49] D–A energy transfer,[68] and photo-induced D–A CT[51] can be realized in polaritonic devices relying on photoconductivity. For example, wavelength-dependent photocurrent characteristics measured in photoTFTs incorporated in a microcavity suggest that energy transfer from photoexcited MoS_2 donor to organic acceptor occurs even with a 50-nm PMMA spacer layer when the acceptor is strongly coupled.[68] In another study, polariton-enhanced electron transfer in strongly coupled doped organic films in microcavities was inferred from weaker electric-field dependence of charge photogeneration efficiency as compared to that due to exciton

dissociation in "bare" films,[51] with the mechanism attributed to polariton delocalization.

Applications of organic polariton BECs

BEC signifies the macroscopic occupation of a singlet coherent quantum state occurring when the de Broglie wavelength of bosons (e.g., polaritons) is comparable to the mean interparticle distance.[6] BEC has been demonstrated in a variety of organic materials incorporated in all-DBR or metal-DBR cavities and has been utilized in demonstrations of room-temperature polariton lasers,[69] ultrafast polariton transistors,[70] and polariton simulators[71] (Figure 1b). Development of material design principles for efficient BEC is an active current research area, and several novel applications of organic polariton BEC systems that use polaritons as qubits in quantum information, quantum computing, and neuromorphic polariton networks[72] are awaiting implementation.

What can polaritons do to remove bottlenecks in SF-based devices?

Polaritons to expand the range of SF-efficient materials

The effect of strong coupling on SF has been a subject of theoretical investigations, which suggested that the polaritons can enhance[73] or suppress[74] SF, depending on the SF mechanism and polariton properties. The most benefit of strong coupling for the yield of ^{1}(TT) states was predicted for exothermic SF materials with $E(S_1)-2E(T_1)>0.1$ eV where polaritons can help mitigate excessive exothermicity and its associated energy loss. In particular, polariton formation enables manipulation of the potential energy surface using cavity detuning and interaction strength, favoring ^{1}(TT) formation over relaxation to the dark singlet states,[73] and enhancing the rate of ^{1}(TT) by aligning the LP with the ^{1}(TT) energy (Figure 4b). Such alignment promotes mixing of the LP and ^{1}(TT) states, which boosts the LP→^{1}(TT) transition rate. Polariton-controlled SF expands the choice of materials suitable for SF-based devices by relaxing the criterion for desirable energies from $0<E(S_1)-2E(T_1)\leq0.1$ eV in "bare" materials to $0<E(S_1)-2E(T_1)\leq0.4$ eV under strong coupling conditions if an interaction strength of 0.4 eV is achieved (Figure 4a).[75] Another important requirement for the polariton-mediated SF is long (picoseconds) polariton lifetimes, which can be achieved in high-Q-factor cavities, but may present a limitation to SF polariton devices. The interplay of exciton–exciton and the exciton–photon coupling also influences the ^{1}(TT) formation and whether it would be enhanced or suppressed as a result of strong coupling,[76] so more work is needed to understand material design from the intermolecular interactions standpoint that enhances polariton-mediated SF. The experimental work probing SF rates under strong coupling conditions has been done only on a few systems (such as rubrene and TIPS-Pn)[30,34] and in low-Q-factor (all-metal) cavities that are not expected to produce significant polariton-induced changes.[75] Therefore, polariton-enhanced SF yields and rates predicted theoretically for exothermic materials are awaiting experimental validation.

Polaritons to manage triplet pairs

Triplet–triplet annihilation, which is the process reverse to the SF (i.e., $T_1+T_1\rightarrow{}^1$(TT)$\rightarrow S_1+S_0$), can also be manipulated by polaritons.[74,77] For example, harvesting of ^{l}(TT) populations with $l=1$ and 5 by LP was observed in TIPS-Tc films in cavities.[31] The mechanisms behind interactions between the triplet pairs and polariton states are still under investigation, but the initial results indicate the potential for polariton-controlled properties of triplet pairs in optoelectronic and quantum information devices.

Polaritons to relax requirements for SF donor–acceptor devices

For a successful SF D–A material, there is a long list of requirements,[4] which includes high SF yield and fast SF rate, as well as efficient exciton diffusion, for the SF donor, all of which can potentially be promoted using polaritons (Figure 4d). In particular, one of the bottlenecks for these devices is a limited triplet diffusion length, which in endothermic SF materials such as Tc can be enhanced via a singlet-mediated process.[78] An open question is whether this process can be further enhanced by strong coupling to the SF donor, taking advantage of a long-range energy transfer and improving photon harvesting using thicker films, well outside of a typical range of <100-nm thickness for organic solar cells. This would be especially beneficial for an endothermic SF donor, for which slower SF rates dictate the need for thicker films.[4] Another open question is whether it would be possible to manipulate two-electron transfer $2e^-$ directly from the ^{1}(TT) states with polaritons, to take advantage of both polariton-enhanced SF and polariton-mediated CT (Figure 3). In endothermic SF donor–acceptor materials, the polariton-suppressed CT may reduce the competition between the relatively slow SF and the undesirable one-electron CT from the donor to acceptor. Finally, given the importance of quality of D–A interfaces and associated disorder, another interesting question is whether the presence of delocalized polaritons at the SF D–A interfaces, and their reduced sensitivity to the presence of disorder, could mitigate the negative effects of disorder at D–A interfaces.

Polaritons to enhance photostability

Polaritons have the potential to both suppress and promote (photo)chemical reactivity.[79] However, what would be achievable with polariton chemistry is under debate. Among the open questions are the types of chemical reactions that can be effectively controlled with polaritons, including those relevant for (photo)degradation of SF materials. However, the design of polaritonic systems for achieving, for example, a specified CT driving force (e.g., to suppress the CT rate for an undesirable reaction) requires knowledge of oxidation and reduction potentials in polaritonic systems. Although the electrochemical control of strong coupling has been demonstrated,[80] this is only the first step toward polariton-controlled CT reactivity. For any other reaction, systematic knowledge of how to generate energy landscapes and incorporate them in devices that prevent the population of excited states involved in photogeneration or depopulate them via polariton states (Figure 4c), would help improve photostability of organic electronic materials and provide useful framework for polariton-controlled photochemistry in SF materials.

Summary and outlook

In the past decade, considerable progress has been achieved in understanding of physics of polaritons in organic materials, expanding the range of materials from J-aggregated dyes used in pioneering work to a variety of electronic and photonic materials, including SF materials. Moreover, polariton formation has been demonstrated in multiple geometries: cavities,

plasmonic strictures, HMMs, and cavity-free layered or ordered systems. This work laid foundations for the next steps for polariton-mediated SF materials and devices: (1) demonstrating theoretically predicted enhancement in SF rates and yield; (2) demonstrating polariton-mediated SF from novel materials that are not considered to be efficient SF materials in the absence of polaritons; (3) demonstrating polariton-enhanced SF-based device performance (that makes use of polariton-mediated SF yield, rate, and/or energy or charge transfer) and/or (photo)stability (Figure 4d); and (4) developing design guidelines for optoelectronic device structures that support polariton formation and efficiently utilize polariton-enhanced processes in a broad range of optoelectronic devices, with a systematic understanding of tradeoffs. Toward next-generation polariton-enhanced technology, it is necessary to understand how the processes relying on coherence of polariton states, observed by optical spectroscopy and ultrafast microscopy (e.g., long-range ballistic polariton transport), could be efficiently utilized in applications. This highlights the importance of comprehensive studies that combine material design with spectroscopy, theoretical modeling, and device studies.

Author contributions

All authors contributed to reviewing research articles and writing the manuscript.

Funding

Competing interests

The authors declare no competing interests.

References

1. O. Ostroverkhova, *Chem. Rev.* **116**, 13279 (2016)
2. L. Zhu, M. Zhang, J. Xu, C. Li, J. Yan, G. Zhou, Wi. Zhong, T. Hao, J. Song, X. Xue, Z. Zhou, R. Zeng, H. Zhu, C.C. Chen, R.C.I. MacKenzie, Y. Zou, J. Nelson, Y. Zhang, Y. Sun, F. Liu, *Nat. Mater.* **21**, 656 (2022)
3. B. Daiber, K. van den Hoven, M.H. Futscher, B. Ehrler, *ACS Energy Lett.* **6**, 2800 (2021)
4. A.J. Baldacchino, M.I. Collins, M.P. Nielsen, T.W. Schmidt, D.R. McCamey, M.J.Y. Tayebjee, *Chem. Phys. Rev.* **3**, 021304 (2022)
5. A.J. Musser, J. Clark, *Annu. Rev. Phys. Chem.* **70**, 323 (2019)
6. Z. Jiang, A. Ren, Y. Yan, J. Yao, Y.S. Zhao, *Adv. Mater.* **34**, 2106095 (2022)
7. A.J. Carrod, V. Gray, K. Borjesson, *Energy Environ. Sci.* **15**, 4982 (2022)
8. D. Padula, Ö.H. Omar, T. Nematiaram, A. Troisi, *Energy Environ. Sci.* **12**, 2412 (2019)
9. O. El Bakouri, J.R. Smith, H. Ottosson, *J. Am. Chem. Soc.* **142**, 5602 (2020)
10. T. Ullrich, D. Munz, D.M. Guldi, *Chem. Soc. Rev.* **50**, 3485 (2021)
11. D. Congreve, J. Lee, N.J. Thompson, E. Hontz, S. Yost, P. Reusswig, M. Bahlke, S. Reineke, T. Van Voorhis, M.A. Baldo, *Science* **340**, 334 (2013)
12. N.J. Thompson, E. Hontz, D. Congreve, M. Bahlke, S. Reineke, T. Van Voorhis, M.A. Baldo, *Adv. Mater.* **26**, 1366 (2014)
13. N.A. Pace, N. Korovina, T. Clikeman, S. Holliday, D. Granger, G. Carroll, S. Nanayakkara, J. Anthony, I. McCulloch, S. Strauss, O. Botalina, J. Johnson, G. Rumbles, O. Reid, *Nat. Chem.* **12**, 63 (2020)
14. C.K. Yong, A. Musser, S. Bayliss, S. Lukman, H. Tamura, O. Bubnova, R. Hallani, A. Meneau, R. Resel, M. Maruyama, S. Hotta, L. Herz, D. Beljonne, J. Anthony, J. Clark, H. Sirringhaus, *Nat. Commun.* **8**, 15953 (2017)
15. H. Kim, B. Keller, R. Ho-Wu, N. Abeyasinghe, R. Vazquez, T. Goodson III., P. Zimmerman, *J. Am. Chem. Soc.* **140**, 7760 (2018)
16. D. Guo, L. Ma, Z. Zhou, D. Li, C. Wang, X. Zhao, F. Zhang, J. Zhang, Z. Nie, *J. Mater. Chem. A* **8**, 5572 (2020)
17. A.R. Bowman, S.D. Stranks, B. Monserrat, *Chem. Mater.* **34**, 4865 (2022)
18. L. Ye, Y. Zhao, R. Xu, S. Li, C. Zhang, H. Lim, H. Zhu, *J. Am. Chem. Soc.* **145**, 26257 (2023)
19. M. Einzinger, T. Wu, J. Kompalla, H. Smith, C. Perkinson, L. Nienhaus, S. Wiegholed, D. Congreve, A. Kahn, M.G. Bawendi, M.A. Baldo, *Nature* **571**(7763), 90 (2019)
20. D.P. DiVincenzo, *Fortschr. Phys.* **48**, 771 (2000)
21. R.M. Jacobberger, Y. Qiu, M.L. Williams, M.D. Krzyaniak, M.R. Wasielewski, *J. Am. Chem. Soc.* **144**, 2276 (2022)
22. K. Gao, Y. Li, Y. Yang, Y. Liu, M. Liu, W. Liang, B. Zhang, L. Wang, J. Zhu, K. Wu, *Adv. Mater.* **36**, 2309420 (2024)
23. T. Wang, H. Liu, X. Wang, L. Tang, J. Zhou, X. Song, L. Lv, W. Chen, Y. Chen, X. Li, *ACS Catal.* **13**, 13902 (2023)
24. Y. Liu, J. Li, S. Gong, Y. Yu, Z. Zhu, C. Ji, Z. Zhao, X. Chen, G. Feng, B. Tang, *ACS Mater. Lett.* **6**, 896 (2024)
25. R. Puro, J.D.B. Van Schenck, R. Center, E.K. Holland, J.E. Anthony, O. Ostroverkhova, *J. Phys. Chem. C* **125**, 27072 (2021)
26. T. Khazanov, S. Gunasekaran, A. George, R. Lomlu, S. Mukherjee, A.J. Musser, *Chem. Phys. Rev.* **4**, 041305 (2023)
27. M. Du, L. Martinez-Martinez, R. Ribeiro, Z. Hu, V. Menon, J. Yuen-Zhou, *Chem. Sci.* **9**, 6659 (2018)
28. R. Pandya, A. Ashoka, K. Gerogiu, J. Sung, R. Jayaprakash, S. Renken, L. Gai, Z. Chen, A. Rao, A. Musser, *Adv. Sci.* **9**, 2105569 (2022)
29. E. Michail, K. Rashidi, B. Liu, G. He, V.M. Menon, M.Y. Sfeir, *Nano Lett.* **24**, 557 (2024)
30. B. Liu, V.M. Menon, M.Y. Sfeir, *ACS Photonics* **7**, 2292 (2020)
31. D. Polak, R. Jayaprakash, T. Lyons, L. Martinez-Martinez, A. Leventis, K. Fallon, H. Coulthard, D. Bossanyi, K. Georgiu, A. Petty, J. Anthony, H. Bronstein, J. Yuen-Zhou, A. Tartakovskii, J. Clark, A. Musser, *Chem. Sci.* **11**, 343 (2020)
32. J. Van Schenck, E. Tanyi, L.-J. Cheng, J. Anthony, O. Ostroverkhova, *MRS Commun.* **3**, 956 (2019)
33. J. Van Schenck, W.T. Goldthwaite, R. Puro, J.E. Anthony, O. Ostroverkhova, *J. Phys. Chem. C* **125**, 27381 (2021)
34. S. Takahashi, K. Watanabe, Y. Matsumoto, *J. Chem. Phys.* **151**, 074703 (2019)
35. C.P. Theurer, F. Laible, J. Tang, K. Broch, M. Fleischer, F. Schreiber, *Nanoscale* **15**, 11707 (2023)
36. M. Slootsky, X. Liu, V.M. Menon, S.R. Forrest, *Phys. Rev. Lett.* **112**, 076401 (2014)
37. M. Wang, M. Hertzog, K. Börjesson, *Nat. Commun.* **12**, 1874 (2021)
38. K. Georgiou, R. Jayaprakash, A. Othonos, D.G. Lidzey, *Angew. Chem. Int. Ed.* **60**, 16661 (2021)
39. A. Cargioli, M. Lednev, L. Lavista, A. Camposeo, D. Pisignano, A. Tredicucci, F. Garcia-Vidal, J. Feist, L. Persano, *Nanophotonics* **13**(14), 2541 (2024). https://doi.org/10.1515/nanoph-2023-0677
40. J. Feist, F.J. Garcia-Vidal, *Phys. Rev. Lett.* **114**, 196402 (2015)
41. G. Engelhardt, J. Cao, *Phys. Rev. B* **105**, 064205 (2022)
42. R.H. Tichauer, I. Sokolovskii, G. Groenhof, *Adv. Sci.* (Weinheim) **10**(33), 2302650 (2023)
43. S. Hou, M. Khatoniar, K. Ding, Y. Qu, A. Napolov, V. Menon, S.R. Forrest, *Adv. Mater.* **32**, 2002127 (2020)
44. A.M. Berghuis, R. Tichauer, L. de Jong, I. Sokolovskii, P. Bai, M. Ramezani, S. Murai, G. Groenhof, J. Rivas, *ACS Photonics* **9**, 2263 (2022)
45. G.G. Rozenman, K. Akulov, A. Golombek, T. Schwartz, *ACS Photonics* **5**, 105 (2018)
46. D. Xu, A. Mandal, J.M. Baxter, S.W. Cheng, I. Lee, H. Su, S. Liu, D.R. Reichman, M. Delor, *Nat. Commun.* **14**, 3881 (2023)
47. Y. Kurman, R. Dahan, H. Sheinfux, K. Wang, M. Yannai, Y. Adiv, O. Reinhardt, L. Tizei, S. Woo, J. Li, J. Edgar, M. Kociak, F. Koppens, I. Kaminer, *Science* **372**(6547), 1181 (2021)
48. M. Balasubrahmaniyam, A. Simkhovich, A. Golombek, G. Sandik, G. Ankonina, T. Schwartz, *Nat. Mater.* **22**, 338 (2023)
49. B. Liu, X. Huang, S. Hou, D. Fan, S.R. Forrest, *Optica* **9**, 1029 (2022)
50. C. Schäfer, M. Ruggenthaler, H. Appel, A. Rubio, *Proc. Natl. Acad. Sci. U.S.A.* **116**, 4883 (2019)
51. N. Krainova, A.J. Grede, D. Tsokkou, N. Banerji, N.C. Giebink, *Phys. Rev. Lett.* **124**, 177401 (2020)
52. C.A. Delpo, S.U.Z. Khan, K.H. Park, B. Kudisch, B.P. Rand, G.D. Scholes, *J. Phys. Chem. Lett.* **12**, 9774 (2021)
53. A. Mandal, T.D. Krauss, P. Huo, *J. Phys. Chem. B* **124**(29), 6321 (2020)
54. W. Wu, A.E. Sifain, C.A. Delpo, G.D. Scholes, *J. Chem. Phys.* **157**, 161102 (2022)
55. J. Mony, Y. Yu, C. Schäfer, S. Mallick, K. Kushwaha, K. Börjesson, *J. Phys. Chem. C* **126**, 7965 (2022)
56. B. Munkhbat, M. Wersäll, D.G. Baranov, T.J. Antosiewicz, T. Shegai, *Sci. Adv.* **4**, eaas9552 (2018)
57. V. Peters, M. Faruk, J. Asane, R. Alexander, D. Peters, S. Prayakarao, S. Rout, M. Noginov, *Optica* **6**, 318 (2019)
58. V.C. Nikolis, A. Mischok, B. Siegmund, J. Kublitski, X. Jia, J. Benduhn, U. Hormann, D. Neher, M. Gather, D. Spoltore, K. Vandewal, *Nat. Commun.* **10**, 3706 (2019)

59. L. M.A. de Jong, A.M. Berghuis, M.S. Abdelkhalik, T.P.A. van der Pol, M.M. Wienk, R.A.J. Janssen, J. Gómez Rivas, *Nanophotonics* **13**(14), 2531 (2024). https://doi.org/10.1515/nanoph-2023-0613

60. M. Held, A. Graf, Y. Zakharko, P. Chao, L. Tropf, M. Gather, J. Zaumseil, *Adv. Opt. Mater.* **6**, 1700962 (2018)

61. E. Eizner, J. Brodeur, F. Barachati, A. Sridharan, S. Kena-Cohen, *ACS Photonics* **5**, 2921 (2018)

62. L. Liu, Z. Wei, S.C.J. Meskers, *J. Am. Chem. Soc.* **145**, 2040 (2023)

63. Y.C. Chao, H. Lin, J. Lin, Y. Tsao, Y. Liao, F. Hsu, Y. Chen, *J. Mater. Chem. C* **11**, 2273 (2023)

64. K. Nagarajan, J. George, A. Thomas, E. Devaux, T. Chervy, S. Azzini, K. Joseph, A. Jouatti, M. Hossieni, A. Kumar, C. Genet, N. Bartolo, C. Ciuti, T. Ebbesen, *ACS Nano* **14**, 10219 (2020)

65. K. Kaur, B. Johns, P. Bhatt, J. George, *Adv. Funct. Mater.* **33**, 2306058 (2023)

66. T.W. Ebbesen, *Acc. Chem. Res.* **49**, 2403 (2016)

67. E.S.H. Kang, S. Chen, V. Derek, C. Hagglund, E. Glowacki, M. Jonsson, *J. Mater. Chem. C* **9**, 2368 (2021)

68. P. Bhatt, J. Dutta, K. Kaur, J. George, *Nano Lett.* **23**, 5004 (2023)

69. Y. Qu, S. Hou, S.R. Forrest, *ACS Photonics* **7**, 867 (2020)

70. A. Zasedataev, A. Baranikov, D. Urbonas, F. Scafirimuto, U. Scherf, T. Stoferle, R. Mahrt, P. Lagoudakis, *Nat. Photonics* **13**, 378 (2019)

71. M. Wei, W. Verstraelen, K. Orfanakis, A. Ruseckas, T. Liew, I. Samuel, G. Turnbull, H. Ohadi, *Nat. Commun.* **13**, 7191 (2022)

72. A. Kavokin, T. Liew, C. Schneider, P.G. Lagoudakis, S. Klembt, S. Hoefling, *Nat. Rev. Phys.* **4**, 435 (2022)

73. L.A. Martinez-Martinez, M. Du, R.F. Ribeiro, S. Kena-Cohen, J. Yuen-Zhou, *J. Phys. Chem. Lett.* **9**, 1951 (2018)

74. B. Gu, Y. Gu, V.Y. Chernyak, S. Mukamel, *Acc. Chem. Res.* **56**, 2753 (2023)

75. C. Climent, D. Casanova, J. Feist, F.J. Garcia-Vidal, *Cell Rep. Phys. Sci.* **3**, 100841 (2022)

76. B. Zhang, Y. Zhao, W.Z. Liang, *J. Phys. Chem. C* **125**, 1654 (2021)

77. C. Ye, S. Mallick, M. Hertzog, M. Kowalewski, K. Börjesson, *J. Am. Chem. Soc.* **143**, 7501 (2021)

78. Y. Wan, Z. Guo, T. Zhu, S. Yan, J. Johnson, L. Huang, *Nat. Chem.* **7**, 785 (2015)

79. B. Xiang, W. Xiong, *Chem. Rev.* **124**, 2512 (2024)

80. W. Ahn, B.S. Simpkins, *APL Photonics* **5**, 076107 (2020)   □

Publisher's note

Oksana Ostroverkhova is currently a full professor in the Department of Physics at Oregon State University. She received her diploma in physics and optical engineering from Kyiv Taras Shevchenko National University, Ukraine, in 1996, and her PhD degree in physics from Case Western Reserve University in 2001. After completing postdoctoral work at Stanford University and the Killam Memorial Fellowship at the University of Alberta, Canada, she joined Oregon State University in 2005. Her research interests focus on the optoelectronic and photonic properties of organic and two-dimensional materials spanning various time and spatial scales, in the development of sustainable materials, and in utilizing optical properties of materials in entomology. Ostroverkhova can be reached by email at oksana@science.oregonstate.edu.

Winston Goldthwaite is a doctoral candidate in the Organic Photonics and Electronics Group at Oregon State University. He received his BA degree in physics and astronomy from Carleton College in 2019. He then began his graduate studies at Oregon State University, and in 2021, received his MS degree in physics on the properties of strong exciton–photon coupling in organic semiconductors. His current research focuses on the photophysics and photochemistry of singlet fission materials. Goldthwaite can be reached by email at goldthww@oregonstate.edu.

Roshell Lamug is a doctoral candidate in the Department of Physics at Oregon State University. She received her bachelor's degree in physics from California State University, Long Beach, in 2020. She received her MS degree in physics from Oregon State University in 2022. Her research interests include organic optoelectronics, focusing on understanding exciton, polariton, and charge-carrier dynamics of singlet fission materials. Lamug can be reached by email at lamugr@oregonstate.edu.