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Charge-Transfer Complexes and Their Role in Exciplex Emission and Near-Infrared Photovoltaics

Tsz-Wai Ng, Ming-Fai Lo,* Man-Keung Fung, Wen-Jun Zhang, and Chun-Sing Lee*

Charge transfer and interactions at organic heterojunctions (OHJs) are known to have critical influences on various properties of organic electronic devices. In this Research News article, a short review is given from the electronic viewpoint on how the local molecular interactions and interfacial energetics at P/N OHJs contribute to the recombination/dissociation of electron-hole pairs. Very often, the P-type materials donate electrons to the N-type materials, giving rise to charge-transfer complexes (CTCs) with a $P^{\delta+}$–$N^{\delta-}$ configuration. A recently observed opposite charge-transfer direction in OHJs is also discussed (i.e., N-type material donates electrons to P-type material to form $P^{\delta-}$–$N^{\delta+}$). Recent studies on the electronic structures of CTC-forming material pairs are also summarized. The formation of $P^{\delta-}$–$N^{\delta+}$-type CTCs and their correlations with exciplex emission are examined. Furthermore, the potential applications of CTCs in NIR photovoltaic devices are reviewed.

1. Introduction

Development of organic optoelectronic devices including organic light-emitting devices (OLEDs) and organic photovoltaic (OPV) devices has shown rapid advances in recent years.[1,2] As these devices often utilize multiple layers of organic semiconductors for controlling carrier-injection/blocking, as well as electron–hole recombination/separation processes, a thorough understanding on the charge interaction at these organic–organic heterojunctions is important.

In early years, most organic semiconductors were observed to be non-interacting with each other, thus showing a negligible charge transfer with a flat energy-band structure sharing common vacuum levels (VLs) across the junctions (**Figure 1**a).[3,4] Until the past decade, OHJs formed by strongly interacting organic pairs showed attractive electronic/optoelectronic properties and drew much attention.[5–7] In all these cases, the P-type semiconductors donate electrons to the N-type semiconductors, giving rise to charge-transfer complexes (CTCs)

with a $P^{\delta+}$–$N^{\delta-}$ configuration, as shown in Figure 1b.[8–10] This results in the accumulation of electrons and holes, respectively, in the N- and P-type materials, leading to surplus majority carriers at the interface. This type of $P^{\delta+}$–$N^{\delta-}$ CTC is often realized in OPV devices[11] and organic field-effect transistors (OFETs) with high conductivities along the contacts.[12] Apart from their electronic properties, many CTCs have also been reported to show interesting long-wavelength optical absorption bands that are not observed in their constituting materials.[13–15] For example, Murata et al. reported that CTCs formed between MoO_3 and many hole-transporting materials (HTM) show absorption peaks in the near-infrared (NIR) region of 800 to 1600 nm.[13] Such CTC-related long-wavelength absorption has recently been exploited for applications in NIR photovoltaic devices.[16–18]

Until last year, OHJs with an opposite charge-transfer direction (i.e., N-type material donating electrons to P-type material to form $P^{\delta-}$–$N^{\delta+}$) had not been reported.[19] In such OHJs (Figure 1c), both the semiconductors lose their majority carriers in contrast to that of accumulation described in Figure 1b. The formation of this type of OHJ requires P- and N-type molecules with low-and high-lying Fermi levels, respectively. The rarity of these "depletion" OHJs can be understood by the fact that P- and N-type molecules are often characterized by, respectively, high-and low-lying energy levels instead. More interestingly, it was observed that such a special $P^{\delta-}$–$N^{\delta+}$ type of CTC is closely related to exciplex emission.[20]

In this Research News article, we summarize our recent studies on the electronic structures of interfaces of CTC-forming pairs of materials. We first discuss the formation of $P^{\delta-}$–$N^{\delta+}$-type CTCs and their relationship to exciplex emission. Furthermore, the applications of CTCs for NIR photovoltaic devices are reviewed.

Dr. T.-W. Ng, Dr. M.-F. Lo, Dr. M.-K. Fung,
Prof. W.-J. Zhang, Prof. C.-S. Lee
Center of Super-Diamond
and Advanced Films (COSDAF)
Department of Physics and Materials Science
City University of Hong Kong
Hong Kong SAR, P. R. China
E-mail: mingflo@cityu.edu.hk; apcslee@cityu.edu.hk
Dr. T.-W. Ng, Dr. M.-F. Lo, Dr. M.-K. Fung, Prof. C.-S. Lee
City University of Hong Kong Shenzhen Research Institute
Shenzhen, People's Republic of China

2. Organic Heterojunctions with Exciplex Emission

In general, emissions in OLEDs originate from radiative relaxation of excitons crossing energy gaps of single emitting materials. On the other hand, red-shifted and broadened emission can sometime be observed from the radiative decay of CTCs formed at OHJs; which is referred to as exciplex emission. The energy of this interfacial emission is always less than the
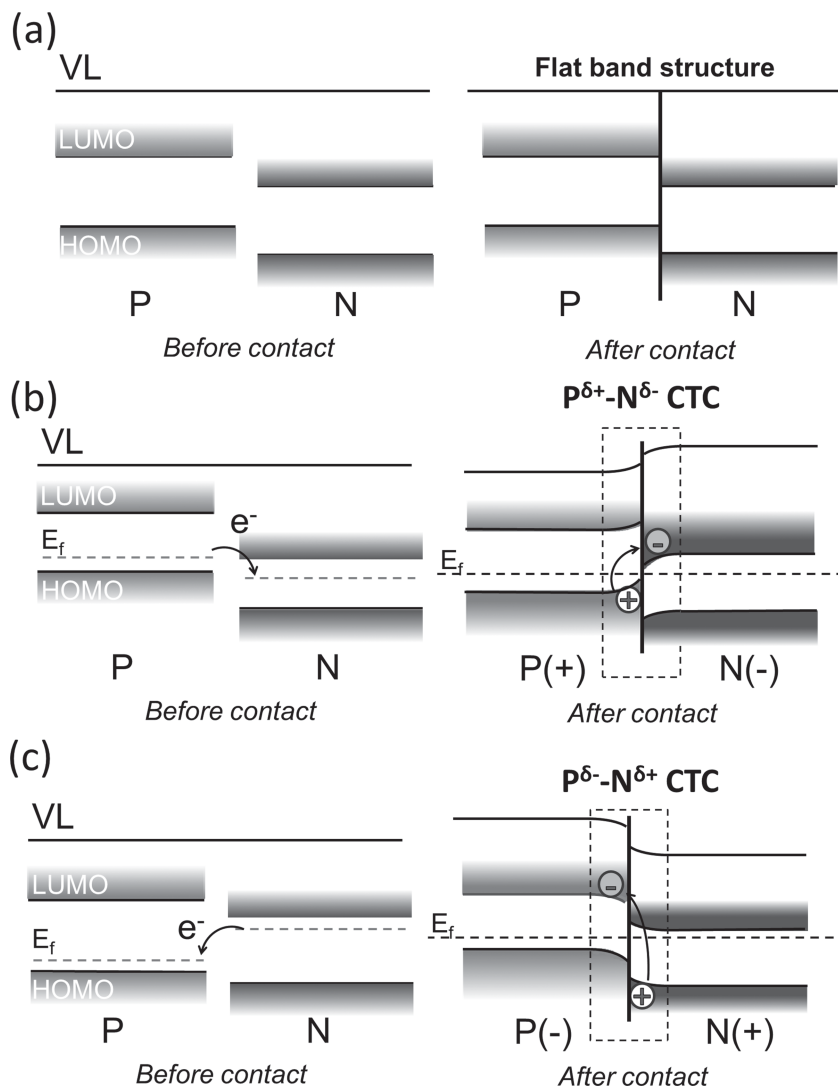
Figure 1. Schematic diagrams of D/A interface forming from P-type and N-type semiconductors. a) VL aligned HJ with little interaction; strong interactive intrinsic molecular pairs with (b) $P^{\delta+}-N^{\delta-}$ CTC; and (c) $P^{\delta-}-N^{\delta+}$ CTC formed at the P/N interfaces. The left and right panels show the respectively energy levels diagrams before contact and after contact formation. Reproduced with permission.[19] Copyright 2012, AIP Publishing LLC.

gap energies of the individual constituting molecules. In early studies, most exciplex-emission OLEDs are reported to have low efficiencies, and are therefore considered to be undesirable. Until recently, exciplex formation in OLEDs has attracted much attention from the viewpoint of spectral color tuning in white-light applications.[21,22] Another recent breakthrough is the application of an exciplex-forming co-host for obtaining thermally activated delayed fluorescent (TADF) OLEDs with high efficiency.[22,23]

In 2012, we used photoemission spectroscopy to study the electronic structures of the m-4,4,4-tris(3-methylphenyl-phenylamino) triphenylamine/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (MTDATA/BCP) junction which is known to give exciplex emission. It is interesting that the interface electronic structures show neither the flat energy-level feature, nor the $P^{\delta+}-N^{\delta-}$-type charge transfer. The m-MTDATA/

BCP junction shows a $P^{\delta-}-N^{\delta+}$-type charge transfer that had not reported before (Figure 1c).[20] This type of CTC is rare because its formation requires HTMs and electron-transporting materials (ETMs) with, respectively, low- and high-lying energy levels. Nevertheless, we have found another five examples of such $P^{\delta-}-N^{\delta+}$ junctions, rubrene/fullerene (C_{60}), 4,4′-bis(3-methylphenylphenyl-amino)biphenyl/bathophenanthroline (TPD/BPhen), TPD/tris 8-hydroxyquinoline aluminum (Alq_3), m-MTDATA/BPhen, and m-MTDATA/Alq_3, simply by focusing our attention on those material pairs reported to give exciplex emissions.

To further confirm the association of exciplex emission with $P^{\delta-}-N^{\delta+}$-type CTCs, we carried out an interfacial study of the rubrene/C_{60} junction. In this work, we made use of the fact that energy-level alignments of some OHJs are sensitive to the substrate work function.[24–26] We found that the energy-level alignment at the rubrene/C_{60} interface can switched between $P^{\delta-}-N^{\delta+}$ and the $P^{\delta+}-N^{\delta-}$ types by using a CuPc (**Figure 2**a) or an m-MTDATA interlayer (Figure 2b) on indium tin oxide (ITO).[20] Two OLEDs with the rubrene/C_{60} junction were then respectively fabricated on ITO/CuPc and ITO/m-MTDATA substrates. It was found that the rubrene/C_{60} interfaces with $P^{\delta-}-N^{\delta+}$- and $P^{\delta+}-N^{\delta-}$-type CTCs respectively give electroluminescence with and without exciplex emission at ca. 850 nm (Figure 2c) even under external bias from 4 to 7 V. This suggests that the observations are independent to external bias and the results are not due to the shift in electron–hole recombination zone in the forming devices. These results further confirm the correlation between exciplex emissions and the $P^{\delta-}-N^{\delta+}$-type OHJ. More experimental data and ultraviolet photoemission spectroscopy (UPS) results interpretation can be found in ref.[20] Also, it is noted that the EL emission of a device using an ITO/CuPc substrate at 600–700 nm is slightly weaker than that using an ITO/m-MTDATA substrate, due to the absorption properties of the CuPc layer in that region.

The question remaining is why such a majority-carrier "depletion" junction with $P^{\delta-}-N^{\delta+}$-type CTCs would give rise to exciplex emission. Figure 2d is a schematic diagram showing the $P^{\delta-}-N^{\delta+}$ CTC at the P–N junction. Upon formation of the CTC, the N-type molecule donates an electron to the P-type molecule to form a $P^{\delta-}-N^{\delta+}$ complex. It should be noted that both the hole at the N-type molecule and the electron at the P-type molecule participate in the CTC bonding, and are thus immobile (denoted by the circled charges in Figure 2d). Under external bias, electrons are driven towards the P/N junction. With the bounded CTCs localized at the interface,
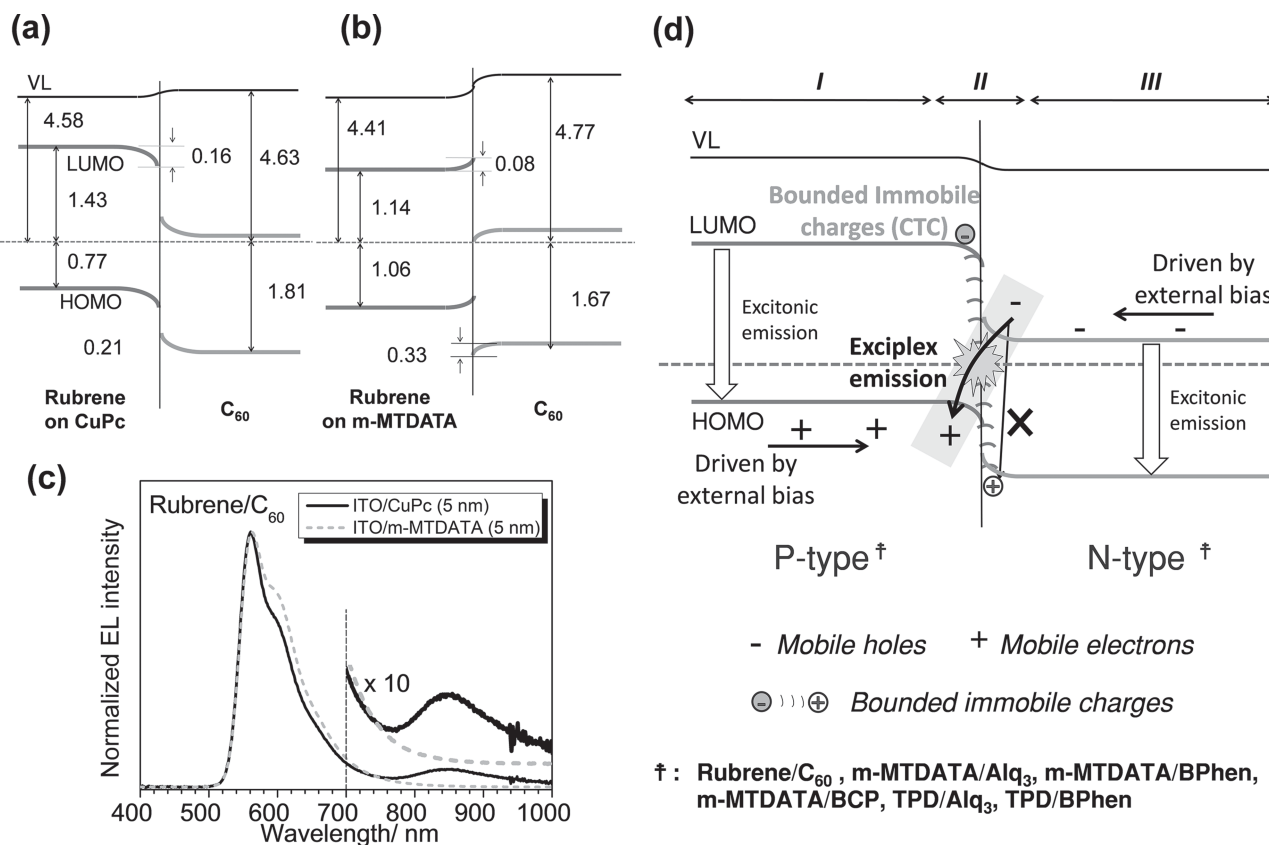
Figure 2. a,b) The electronic structure of rubrene/C_{60} interface formed on ITO/m-MTDATA (5 nm) and ITO/CuPc (5 nm) substrates, respectively. c) EL spectrum of rubrene/C_{60} devices fabricated on substrates of ITO/CuPc (5 nm) and ITO/m-MTDATA (5 nm). Device configurations: ITO/CuPc or m-MTDATA (5 nm)/rubrene (25 nm)/C_{60} (30 nm)/BCP (5 nm)/Al. d) Energy-level diagrams of exciplex-forming organic P/N heterojucntion.

electrons in the lowest unoccupied molecular orbit (LUMO) of N-type molecules cannot directly form excitons and recombine with holes in the highest occupied molecular orbit (HOMO) since these holes have already contributed to the CTC formation (the circled charges in regime II of Figure 2d). The same situation applies also for the incoming holes from the P-type molecules. Thus, instead of overcoming the energy barriers and freeing the bound charges in the CTCs to form excitons with the incoming mobile charges, it is energetically more favorable for the externally injected electrons (from the N-type molecules) to interact with the externally injected holes (from the P-type molecules) directly at the interface. Therefore, the charge-transfer characteristics at this junction favor the formation of exciplex emission. For those molecules far away from the interface (i.e., regimes I and III) with no CTC formation, an excitonic energy-gap emission can be obtained. Figure 2c provides the EL data, showing both excitonic and exciplex emission. This could be easily understood since our device is a simple bilayer device with a single rubrene/C_{60} contact interface. Therefore, depending on the current–voltage parameters, the light-emitting zone can be varied from regimes I to III (in Figure 2d) and gives rise to exciton and/or exciplex emissions.

This mechanism of exciplex emission can also be considered in another simple perspective. On the formation of a $P^{\delta-}$–$N^{\delta+}$

CTC, the P- and the N- molecules respectively use their $LUMO_P$ and $HOMO_N$ (the subscripts "P" and "N" used here indicate the orbitals are from the P- and the N- type molecules respectively) for bonding in the CTC. Upon CTC formation, the $LUMO_P$ and $HOMO_N$ orbitals obtain and lose electrons; that is, they are no longer being "unoccupied" and "occupied". For the CTC as a whole, its LUMO and HOMO are now respectively the $LUMO_N$ and the $HOMO_P$ of the original molecules. This explains why the emission wavelength of the CTC is determined by the energy offset of $LUMO_N - HOMO_P$.

Recently, high-efficiency OLEDs exploiting exciplex emission have attracted much attention.[22,23] Adachi et al. made a breakthrough on harvesting triplet energy via TADF without using any phosphorescence.[27] The key requirement for TADF is a small singlet-triplet energy-level split (<0.1 eV), which in turn means a wide separation of the LUMO and HOMO orbitals. Adachi et al. further propose a new approach of getting materials with such small singlet–triplet splits by mixing strongly interacting donor and acceptor molecules to form a CTC. As the LUMO and the HOMO of the CTC originate from two molecules, these orbital are inherently separated and result in a small singlet–triplet split in the CTC.[27] Kim et al. further extended this approach and reported OLEDs using an exciplex-forming co-host, that showed excellent performance with EQE of 29.1%, power efficiency of 124 lm/W and low turn-on voltage
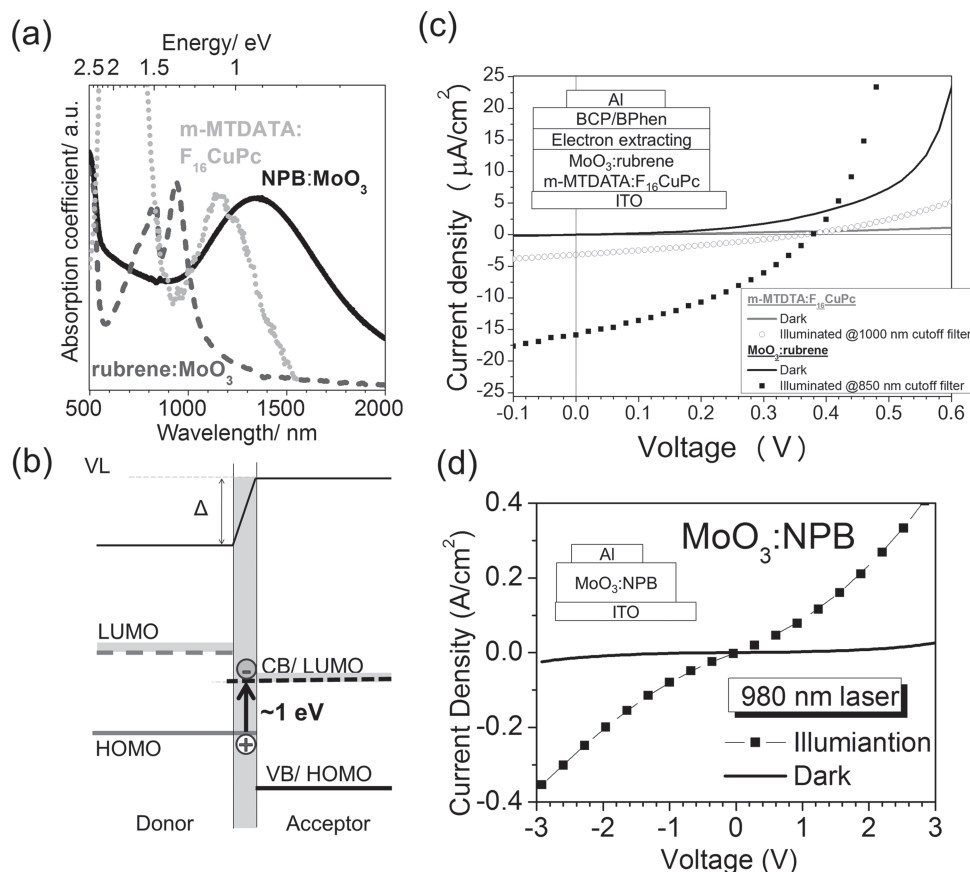
Figure 3. a) UV–vis–NIR absorption spectra of various charge interactive composites in NIR region. b) Interfacial energetics of donor/acceptor junction of NIR absorbing composite. c) Current density–voltage (J–V) characteristics of MoO$_3$:rubrene and m-MTDATA:F$_{16}$CuPc composite for NIR photocharge generation of devices under illumination with wavelength >850 nm and >1000 nm respectively and d) a MoO$_3$:NPB photodetector with NIR photoresponse.

at 2.4 V.[22] All these studies suggest that exciplex emissions are of both scientific and practical interest and further understanding of the physics of exciplex emission is highly desirable.

3. Bandgap Engineering Via Strong Charge Interactions: NIR Absorption

Organic photocharge generation from near IR (NIR) light is well-recognized as an important strategy for enhancing the performance of OPV devices. However, commercially available materials (e.g., SnPc, ClAlPc, PbPc) with a good IR response are rare. Burgeoning interest therefore has been focused on synthesizing narrow-energy-gap materials with an NIR response. Herein, an alternative approach for harvesting NIR light by exploiting CTCs formed by wide-energy-gap materials is highlighted. The interfacial CTCs induce intermediate stepping states within the original wide energy gaps, allowing the generation of charge-transfer (CT) excitons upon NIR excitation.

Applications of wide-bandgap transition-metal oxides in organic device have recently attracted much attention owning to their unique electronic properties of low-lying valence band (VB) levels and high electronegativities.[28,29] They show

strong interactions with many HTMs, such as rubrene,[16] N,N′-di(naphthalen-1-yl)-N,N′-diphenyl-benzidine (NPB),[14] and N,N′-diphenyl-N,N′-bis(1-naphtylphenyl)-1,1′-biphenyl-4,4′-diamine (α–NPD),[15] resulting in distinct enhancements in carrier density and conductivities over those of the individual constituents. These composites, therefore, have been widely used as charge-generating units in tandem OLEDs as well as the anode buffer layers for improving hole injection.[30]

Apart from the electronic charge transport, some of these composites exhibit unusual long-wavelength absorption bands as shown in **Figure 3**a. According to the energy gaps of MoO$_3$ (3.2 eV) and the organic materials (rubrene: 2.2 eV; m-MTDATA: 3.2 eV; copper hexadecafluorophthalocyanine F$_{16}$CuPc: 1.4 eV), the mixed composite should not have an absorption peak over the region of 800–1500 nm. Previous studies suggested that the emerging NIR absorption peaks are mainly attributed to the CTCs formed upon mixing.[13–15]

Due to the low-lying and non-overlapping energy levels, i.e., the HOMO and LUMO of the organic do not overlap the VB and the CB of the transition metal oxide, the estimated energy structure of MoO$_3$/organic interface following the VL aligned model suggests the type III junction with broken gap at the contact.[11] This kind of OHJ is charge non-interactive

in principle. In some special case studies, an interfacial dipole might formed at the contact, which might induce significant surface electronic states near the contact junction and accommodates a charge-exchange process at the junction.[25,26] However, the nature of the CT interaction between the mixing constituents (in Figure 3a) is unclear and its role on the NIR absorption is ambiguous.[31,32] Direct insight with regard to the CTC and its possibility of charge generation are thus of great interest.

Figure 3b illustrates the electronic structures of the donor/acceptor interface of the NIR absorbing composite measured by UPS. A significantly large interfacial dipole is observed at the contact showing considerable charge exchange at the interface. The formation of this surface dipole shifts the energy levels of the donor downwards, meanwhile inducing intermediate CT states within the original wide energy gap of the donor molecules. Similar evidence from the literature using photoluminescence (PL) spectroscopy also shows the existence of new electronic states within the original energy gaps of the organic molecules.[33,34]

CT excitons therefore can be generated directly via sub-energy-gap photoexcitation, by electron transition from the HOMO of the donor to the LUMO (or CB) of the acceptor with energy in accordance of the 'effective energy gap' of the CTC of the composite with a much smaller energy of ca. 1 eV (Figure 3b).[16,18] The exciton would subsequently be dissociated at the strongly interactive charge-transfer interface within the composite in the ground-state condition. OPV devices using charge-interactive composites can be found in ref.[16–18] The process of NIR photocharge generation is attributed to the interfacial interaction within the CTC-forming composite. In fact, similar NIR absorption studies have been reported recently by Murata et al. where they attributed the long-wavelength absorption to cationic state of the N,N′-diphenyl–N,N′-bis(1-naphthyl)-1,1′-biphenyl-4,4′-diamine (α–NPD) when it is deposited on top of the metal oxide such as MoO_2, MoO_3 and WO_3.[35]

Figure 3c and 3d illustrate the photoresponse of devices with NIR photosensitive composites. Two OPV devices with NIR-light-absorbing composites of MoO_3:rubrene and m-MTDATA:F_{16}CuPc were sandwiched between the electrodes with appropriate carrier-extraction layers. The current density–voltage (J–V) characteristics shown in Figure 3c clearly demonstrate the photoresponse with NIR power-generation devices upon illumination of NIR light of wavelength longer than 850 nm and 1000 nm respectively. Similar photoresponses upon variation of the external bias were observed when a single NIR-light-absorbing composite (MoO_3:NPB) is used as shown in Figure 3d.

Further studies on CTCs have found that they can play versatile roles in various organic devices.[33,34] In addition to the exciplex emission and NIR photovoltaic response discussed above, CTCs have been exploited for high-performance ambipolar field-effect transport,[5] as well as the connecting unit in tandem OLEDs.[30,36] Recent work on CTCs formed between single crystals further demonstrated the potential of CTCs for obtaining optoelectronic properties not available from typical organic semiconductors.[37–39] It is expected that further understanding and application of CTCs could open up new research windows for organic optoelectronic devices.

4. Conclusion and Outlook

This Research News article highlights recent advances regarding the understanding of local charge interactions and energetics at organic P/N junctions for various new applications. The strategy of exploiting interfacial CTCs with different charge-transfer interactions (i.e., $P^{\delta+}$–$N^{\delta-}$ and $P^{\delta-}$–$N^{\delta+}$) offers a simple and attractive alternative for obtaining novel optoelectronic properties that are not available in the individual constituting components. The formation of the $P^{\delta-}$–$N^{\delta+}$ CTC has been shown to have a close correlation with exciplex emission in OLEDs. On the other hand, it has been demonstrated that CTCs can be used for NIR photovoltaic devices via sub-energy-gap electronic transitions using wide-energy-gap semiconductors with strong charge coupling. Although these findings provide new insights for exploiting charge-interactive couples for new devices applications, more in-depth studies are needed in this area to establish a comprehensive roadmap for achieving new material properties and developing reliable contacts. Furthermore, the growing interest on CTCs in emerging concepts like hot-exciton photocharge generation and sensor applications urge further investigation of the microscopic charge-interaction behavior between donor and acceptor molecules. More studies on the energetics and kinetics of donor–acceptors, as well as the degree of charge-transfer process at electrical contacts, could lead to breakthroughs regarding the understanding of the physical processes in CTCs and rational design concepts of CTC-enabled optoelectronic devices.

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