Realization of triplet-triplet annihilation in planar heterojunction exciplex-based organic light-emitting diodes

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Abstract

Triplet-triplet annihilation (TTA) for enhancement of luminous efficiency occurs with difficulty in exciplex-based organic light-emitting devices (OLEDs) because it is an interaction among several neighboring donor and acceptor molecules. However, TTA has been realized in our planar-heterojunction (PHJ) exciplex-based OLEDs by using a thin recombination zone to enhance the interfacial density of the triplet states. The TTA process, which is characterized by a high-field decrease (HFD) in the magneto-electroluminescence from the PHJ OLEDs, appears at approximately 150 K and becomes stronger with decreasing temperature. At a given temperature, the higher the injected current is, the stronger HFD is observed. Additionally, we find that TTA could even happens at room temperature with appropriate selection of the donor molecule, which may be attributed to the favorable electron-donating ability of the methoxy group ($-\text{OCH}_3$) in the donor molecule and the matched overlaps of the intermolecular conformation of the donor and the acceptor.

Keywords: planar-heterojunction, exciplex, triplet-triplet annihilation, magneto-electroluminescence

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Enhancement of electroluminescence (EL) efficiency, which is dependent on the microscopic dynamics of the electron-hole (e-h) pairs in organic light-emitting devices (OLEDs), is a long-standing focus of research in the OLED field [1-12]. The e-h pairs in OLEDs commonly include excitons and exciplexes [13, 14]. An exciton is formed in a single molecule by the recombination of an electron and a hole, while an exciplex is usually generated at an organic/organic interface [15] via electron transfer from the lowest unoccupied molecular orbital (LUMO) of the acceptor to the highest occupied molecular orbital (HOMO) of the donor. While the EL efficiency in previously fabricated exciplex-based OLEDs is relatively low [14, 16, 17], exciplex emission has still received considerable attention because of its superior properties, including emission color tuning [18-20] and fabrication of white OLEDs [14]. In particular, after Adachi *et al.* reported very high external quantum efficiency in exciplex-based OLEDs using reverse intersystem crossing (RISC) [6-8], study of the enhancement of exciplex-emission efficiency has become a hot research topic. In addition to RISC, triplet-triplet annihilation (TTA) is another well-known and significant pathway to enhancement of EL efficiency that uses triplet states [1-5]. Although, the energy levels of singlet and triplet exciplex states are very close [8, 13, 21], that is, the total of twice the triplet energy is higher than the singlet energy, TTA still occurs with difficulty in exciplex-based OLEDs because it is an interaction among several neighboring molecules and the underlying mechanism of TTA remains unclear. Clearly, realization and exploration of TTA in exciplex-based OLEDs is very important for the understanding of this underlying mechanism and improvement of the EL efficiency of this type of device.

In general, two methods are used to detect TTA in the literature: the super-linear behavior of the luminance-current in OLEDs [22] and time-resolved photoluminescence (PL) or EL spectra [1, 2]. However, the former cannot probe TTA clearly in our device because the delayed fluorescence contribution from TTA is not very strong. It is also difficult to discern the RISC and TTA merely from time-resolved PL or EL spectra because of their complex but similar delayed fluorescence behavior [23]. However, magneto-EL (MEL), as a non-contact and non-destructive probing tool, is capable of distinguishing the two processes and has been used to study TTA in non-magnetic OLEDs [23-27]. This is because both RISC and TTA are

highly spin-related processes [23, 28, 29] that can generate different characteristic MEL responses (including their line-shapes and magnitudes) under application of an external magnetic field [23]. Thus, MEL could be used as a simple and effective method to explore TTA in OLEDs. In this letter, we used MEL to explore the TTA of exciplexes in planar-heterojunction (PHJ) OLEDs by appropriate selection of donor and acceptor materials to obtain large interfacial energy barriers. Clearly, the large interfacial LUMO-LUMO and HOMO-HOMO energy barriers between two materials could effectively confine holes and electrons within the donor and acceptor layers and produce high-density exciplex states in the vicinity of the hetero-interface. Results show that the MEL responses exhibit a rapid low-field increase followed by an obvious high-field decrease (HFD) at relatively low temperatures, which was understood on the basis of the magnetic-field mediated intersystem crossing process [13, 23] and TTA [23, 25]. Additionally, a HFD has been observed at room temperature in PHJ OLEDs with appropriately selected donors, reflecting the fact that room temperature TTA was realized in this kind of device. Our discoveries enrich the microscopic mechanisms and provide a feasible pathway to enhance EL efficiency by using the triplet states in exciplex-based OLEDs.

Six PHJ OLEDs and two bulk-heterojunction OLEDs (active area: $\sim 2 \times 2 \text{ mm}^2$) were fabricated by thermal evaporation under high vacuum ($\sim 10^{-6} \text{ Pa}$) conditions. The hole-transport materials used in these devices include 4,4',4''-Tris(N-3-methylphenyl-Nphenylamino)triphenylamine (*m*-MTDATA) and N1,N4-diphenyl-N1,N4-dim-tolylbenzene-1,4-diamine (TTP). Also, tris(8-hydroxyquinoline) aluminum (Alq_3), 1,3,5-Tri(1-phenyl-1H-benzo[d]imidazol-2-yl)phenyl (TPBi), and 2,8-bis(diphenylphosphoryl)dibenzo-[b,d]thiophene (PPT) were used as electron-transport materials. The device structures include ITO/*m*-MTDATA (or TTP) (80 nm)/ Alq_3 (80 nm)/LiF (1 nm)/Al (120 nm) and another four similar structures of *m*-MTDATA (or TTP) (80 nm)/TPBi (80 nm) and *m*-MTDATA (or TTP) (80 nm)/PPT (80 nm). **Four bulk-heterojunction device structures of ITO/*m*-MTDATA (40 nm)/25wt.% *m*-MTDATA:PPT (80 nm)/PPT (40 nm)/LiF (1 nm)/Al (120 nm) and ITO/ TTP ((160-x)/2 nm)/25wt.% TTP:PPT (x nm)/PPT ((160-x)/2 nm)/LiF (1 nm)/Al (120 nm) (x=80, 40, 10)were also fabricated.** The evaporation rates and the thicknesses of the deposited layers

were detected using a calibrated quartz crystal monitor (XTM/2). Immediately after preparation, the samples were mounted on the cold finger of a close-cycle cryostat (Janis: CCS-350S, temperature range: 10–325 K) located between the pole pieces of an electromagnet (Lakeshore EM647). A magnetic field with a range from –500 to 500 mT was applied parallel to the device surface and measured using a Hall probe gaussmeter when the sample was tested. The EL intensity was measured using a Si photodiode connected to a Keithley 2000 multimeter, and the PL or EL spectra were measured by a grating spectrograph (SpectraPro-2300i).

Fig. 1. Normalized EL spectra for six PHJ devices and two bulk heterojunction devices and the PL spectra of neat *m*-MTDATA (426 nm), TPBi (442 nm), PPT (361 nm), and PPT (392 nm) films at 20 K, respectively. The energy-level diagrams of the six PHJ devices are also displayed.

To achieve pure exciplex emission, we fabricated six PHJ devices that were composed of different materials with various energy levels. The LUMO and HOMO energies of the five organic materials used here were obtained from the literature [8, 13, 30, 31]. Figure 1 shows normalized EL spectra for six PHJ devices and two bulk heterojunction devices and the PL

spectra of neat films of the organic materials used in the devices at 20 K. Also the energy-level diagrams of the six PHJ devices are displayed. As shown on the right of Fig. 1a, the energy barriers between the LUMO levels and the HOMO levels of *m*-MTDATA and Alq₃ are 1.1 and 0.7 eV, respectively. Therefore, holes injected from the anode could overcome the relatively small energy barrier and enter the Alq₃ layer, leading to the formation of excitons with electrons injected from the cathode and the resulting fluorescence emission. This process can be seen in the spectra on the left of Fig. 1a, where the EL spectrum of the *m*-MTDATA/Alq₃ device is basically consistent with the typical radiative emission of Alq₃-based OLEDs [13]. However, the situation changes when Alq₃ is replaced with TPBi. As shown in Fig. 1b, the spectra show that the EL spectrum of the *m*-MTDATA/TPBi device has two peaks (at 420 and 531 nm), which indicates that composite states (excitons and exciplexes) are generated in this device. When combined with the energy-level and spectral diagrams shown in Fig. 1b, the weak peak is approximately in agreement with the PL spectrum peak of *m*-MTDATA (426 nm), thus reflecting that a small proportion of the electrons overcome the small interfacial energy barrier of 0.7 eV and then generate a small quantity of excitons in the *m*-MTDATA layer, leading to weak fluorescence emission. Additionally, the strong peak has a notable red-shift when compared with the PL peaks of *m*-MTDATA (426 nm) or TPBi (442 nm), which indicates that most of the electrons remain in the TPBi layer and form vast exciplex states in the vicinity of the *m*-MTDATA/TPBi hetero-interface by recombination of electrons and holes in the TPBi acceptor and the *m*-MTDATA donor, respectively. While the PPT has a slightly larger electron affinity than TPBi, similar results were also obtained in *m*-MTDATA/PPT devices because the interfacial energy barriers are not high enough, as shown in Fig. 1c.

Because pure exciplex emission could not be obtained in *m*-MTDATA/TPBi or *m*-MTDATA/PPT devices, we replaced *m*-MTDATA with TTP, which has a smaller ionization potential, to realize large interfacial energy barriers. Demonstrating consistency with the situations of *m*-MTDATA/Alq₃ and *m*-MTDATA/TPBi devices, pure excitons and composite states were generated in TTP/Alq₃ and TTP/TPBi devices, respectively, because the interfacial energy barrier is still not large enough, as shown in Fig. 1d and 1e. However, as shown in Fig. 1f, pure exciplex states were successfully realized in the TTP/PPT device

because the comparatively large interfacial energy barriers between TTP and PPT are 1.4 and 1.9 eV, respectively, which effectively confine the holes and the electrons within the donor- and acceptor-layers and produce exciplex states in the vicinity of the hetero-interface. This process can be reflected by the PL and EL spectra shown in Fig. 1f, where the pure exciplex emission from the TTP/PPT device shows an evident red-shift to 501 nm when compared with the respective PL peaks of 392 and 361 nm from the TTP and PPT films. In addition, this is approximately in line with the EL spectrum of the bulk-heterojunction TTP:PPT device. Thus, Fig. 1 demonstrates that the type of excited state is determined by the interfacial energy barriers in the PHJ devices, and pure exciplex emission would be realized when large interfacial energy barriers are obtained between two materials in these devices.

Fig. 2. MELs of seven devices under a constant bias voltage with current flow of 150 μ A over the temperature range from 20 to 300 K, where the temperatures from top to bottom are 300, 200, 150, 100, and 20 K. The insets in Fig. 2f and 2g present the MELs of the TTP:PPT and TTP/PPT device, respectively, under different injected currents at 20 K, where the injected currents from the top to the bottom are both 20, 50, 100, 150 μ A, respectively.

With the aim of investigating TTA in exciplex-based devices, we have measured the MEL of the TTP/PPT device, along with the MELs of four other PHJ devices and the two bulk-heterojunction devices for comparison. These MELs are measured under a constant bias voltage at a current flow of 150 μ A over a temperature range from 20 to 300 K. As shown in Fig. 2, each MEL of these devices presents both low-field and high-field components. The

former component shows a sharp increase, which can be understood based on the magnetic-field suppressed intersystem crossing process [13, 24, 25]. The latter component exhibits gradual variation, and its tendency is either positive or negative (HFD), depending on the evolution processes of the excited states at different temperatures in these devices. Previous research showed that the HFD in MEL could be regarded as a fingerprint of TTA [23, 25]. As shown in Fig. 2a, 2b, 2c, and 2d, the HFD in each MEL appears when temperature decreases to around 100 K in the four *m*-MTDATA (or TTP)/Alq₃ and *m*-MTDATA (or TTP)/TPBi devices. This is because a magnetic-field mediated TTA process occurs among the pure excitons [24, 26] or the composite states at relatively low temperatures in the devices. It was found that while there are only small numbers of excitons in the composite-state *m*-MTDATA (or TTP)/TPBi devices, we cannot exclude their contributions to the TTA and the resulting HFD in the MEL. Hence, in order to realize TTA process in pure exciplex-emission OLEDs, we therefore focused on another kind of device.

Nevertheless, TTA occurs with difficulty in pure exciplex-based devices, even at low temperatures. This can be demonstrated in many bulk-heterojunction exciplex-based OLEDs, as shown in Fig. 2e and 2f, where the high-field components of the MEL always present positive variations, even at 20 K. And this situation appears in all injected currents at 20 K, as seen in inset in Fig. 2f. Furthermore, in our trial experiments, no HFD is observed in the other four bulk-heterojunction exciplex-based OLEDs, which are composed of *m*-MTDATA (or TTP):Alq₃ and *m*-MTDATA (or TTP):TPBi (not shown here). However, this case changed remarkably in the PHJ exciplex-type device composed of TTP/PPT, as shown in Fig. 2g, where the HFD of the MELs appears at approximately 150 K. Additionally, stronger HFD is observed at lower temperatures, and the decreased intensity even falls below its initial zero-field value at 20 K. Therefore, these results indicate that TTA occurs in the pure exciplex-type device. Because the TTA is proportional to the density of the triplet states squared [25, 32], and the density of the triplet states is proportional to the injected current density [25], the MELs at different injected current levels have been measured at 20 K to confirm our conclusions. As shown in the inset of Fig. 2g, the stronger HFD of the MEL occurs when the injected current becomes slightly higher than 20 μ A, and as the injected current increases, then stronger HFD is observed, as expected. This observation further

confirms that TTA does indeed occur at relatively low temperatures in the PHJ exciplex-based TTP/PPT device.

Fig. 3. Schematics of the transport and recombination processes of electrons and holes in the two devices, where (a) is the planar-heterojunction structure of TTP/PPT, and (b) shows the bulk-heterojunction structure of TTP:PPT. MELs of TTP:PPT (40 nm) and TTP:PPT (10 nm) devices are also displayed under a constant bias voltage with current flow of 150 μ A over the temperature range from 20 to 300 K.

Obviously, it is necessary to reveal the mechanism for the different temperature-dependent high-field tendencies of the two devices that consist of the same materials of TTP and PPT but have different device structures (TTP/PPT and TTP:PPT). Figure 2 shows that the two devices have similar high-field tendencies at relatively high temperatures, but have the opposite tendency at relatively low temperatures. As shown in Fig. 3a, in the TTP/PPT PHJ device, the fairly large interfacial energy barriers between the TTP and PPT materials can lead to confinement of the holes and the electrons within the donor and acceptor layers, respectively, and thus produce high-density triplet states in the vicinity of the TTP/PPT hetero-interface. As reported in the literature [25, 26], the TTA process is strongly dependent on both the density and the lifetime of the triplet states in the devices. At relatively high temperatures, the lifetimes of the triplet states are significantly reduced by the fast phonon-assisted decay, which represents a tremendous weakness for TTA [25, 33]. However, when the temperature decreases, the effects of the phonon interaction become

weaker, and thus generate a prolonged lifetime for the triplet states. Therefore, the TTA process could occur and an obvious HFD can be observed in Fig. 2g. This argument was supported by the variation of MELs at 20 K versus various injected currents. As shown in the inset of Fig. 2g, when the injected current increases, the density of the triplet states becomes high and the TTA gets stronger, leading to a more obvious HFD. Nevertheless, there is another opposite situation in the bulk-heterojunction TTP:PPT (80 nm) OLEDs, as shown in Fig. 3b. The recombination of holes and electrons happens over a very wide blend layer (80 nm), leading to a low density of triplet states. Additionally, even at 20 K, while the triplet lifetime in the bulk-heterojunction OLEDs is prolonged, the TTA process can still hardly occur because of the low density of the triplet states in the wide blend layer. Thus, the HFD of the MEL was not observed from room temperature down to 20 K in bulk-heterojunction OLEDs, as shown in Fig. 2e and 2f. This argument was supported by the results of devices that reduce the thickness of blend layer, as seen in Fig. 3c and 3d. When the thickness of the blend layer is 40 nm, the HFD response still disappears, even at 20 K and 150 μ A. However, when the blend layer is reduced to 10 nm, the HFD responses appear just as the PHJ device of TTP /PPT.

Fig. 4 (a) The normalized EL spectra for two PHJ devices of MeO-TPD/TPBi and MeO-TPD/PPT. The insets in Fig. 4a display the energy-level diagrams of MeO-TPD and the normalized EL spectra of two bulk-heterojunction devices of MeO-TPD:TPBi and MeO-TPD:PPT. Fig. 4b shows the chemical structures of m-MTDATA, MeO-TPD and TTP molecules.

(c)–(f) show the MELs of two PHJ devices and two bulk-heterojunction devices under a constant bias voltage with current flow of 150 μ A over the temperature range from 20 to 300 K, where the temperatures from top to bottom are both 300, 200, 100, and 20 K.

Because OLEDs are mostly used at room temperature, the realization of room-temperature TTA would have significant importance for the practical application of these devices. We therefore choose N,N,N',N'-tetrakis(4-methoxyphenyl)-benzidine (MeO-TPD), which has a stronger ability to donate electrons as the electron-donor material, and is a possible candidate for realization of room-temperature TTA in exciplex-based devices. The similar device structures that are used are MeO-TPD (80 nm)/TPBi (80 nm), MeO-TPD (80 nm)/PPT (80 nm), MeO-TPD:TPBi (80 nm), and MeO-TPD:PPT (80 nm). Similar to m-MTDATA/TPBi (or PPT), there are composite states in the MeO-TPD/TPBi (or PPT) devices, as shown in Fig. 4a, where the spectra have a strong peak and a weak peak, respectively. And the inset in the Fig. 4a shows the spectra of MeO-TPD:TPBi and MeO-TPD:PPT devices. The strong peaks of the MeO-TPD/TPBi (or PPT) devices are approximately in line with the EL spectra of MeO-TPD:TPBi (or PPT) devices, respectively. As shown in Fig. 4c and 4d, the high-field components of the MEL always present positive variations even at 20 K and 150 μ A in bulk-heterojunction exciplex-based OLEDs of MeO-TPD:TPBi and MeO-TPD:PPT. However, in the PHJ devices of MeO-TPD/TPBi and MeO-TPD/PPT, as shown in Fig. 4e and 4f, a large HFD has been observed at 20 K, and the minimum magnitudes of the HFD are approximately -4.7% and -4.0% , respectively. Although the HFD tends to become slightly weak with increasing temperature, the apparent HFD still exists at room temperature, i.e., the TTA indeed occurs at room temperature. This is an unambiguously important achievement because TTA hardly ever occurs, even in pure exciton-based OLEDs at room temperature.

Similar to m-MTDATA, the LUMO/HOMO energy levels for MeO-TPD are 1.9/5.1 eV [34], and we cannot attribute this surprising discovery to the LUMO/HOMO energy levels of MeO-TPD. In fact, when compared with *m*-MTDATA or TTP, the chemical structure of MeO-TPD has a prominent difference in that the periphery of the MeO-TPD molecule has four methoxy groups ($-\text{OCH}_3$) [35, 36], as shown in Fig. 4b. In general, the methoxy group that has a strong electron donation ability presents an intense hole property, which is

beneficial for the generation of intermolecular excited states [37-39]. Therefore, it is easier for the devices that use MeO-TPD as a donor material to form exciplex states than those that use *m*-MTDATA or TTP donor materials. From another perspective, if we consider that the exciplex is formed between the donor and acceptor materials, the different chemical structures of *m*-MTDATA, TTP and MeO-TPD cause diverse intermolecular conformation overlaps with the acceptor molecules that could affect the generation efficiency of the exciplex states [40]. Therefore, we guess that the two devices with structures of MeO-TPD/TPBi and MeO-TPD/PPT may have more matched intermolecular conformation overlaps than the others, leading to the efficient generation of exciplex states and the occurrence of TTA at room temperature. It should certainly be noted that the thin recombination zone that is obtained from the application of the PHJ device structure is a prerequisite for the production of high-density triplet states. Obviously, the underlying mechanisms for the realization of TTA at room temperature from exciplex states should be explored further in future work.

In summary, using MEL as a tool, we found that TTA occurred in PHJ exciplex OLEDs at relatively low temperatures by a process of selecting suitable donor and acceptor materials that can generate large interfacial energy barriers to block the electrons and holes and form pure high-density exciplex states in the vicinity of the thin hetero-interface. Room-temperature TTA was also observed in PHJ OLEDs using MeO-TPD as a donor material, which can be attributed to the methoxy group ($-\text{OCH}_3$) of the donor molecule and the well-matched intermolecular conformation overlaps between the acceptor and the donor. These discoveries may offer a method to enhance the EL efficiency of exciplex-based OLEDs by using the triplet states.

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