Using Magneto-Electroluminescence As a Fingerprint to Identify the Carrier-to-Photon Conversion Process in Dye-Doped OLEDs

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ABSTRACT: Long-range Förster energy transfer (FET) and short-range charge trapping (CT) are two competing basic mechanisms in the carrier-to-photon process of dye-doped organic diode, but little is known about which of FET and CT governs the electroluminescence process for a given dye. Here we report that magneto-electroluminescence (MEL) response can serve to identify the fundamental issue. (1) In relative high magnetic field (>20 mT), dramatic decrease in MEL response implies that CT dominates carrier-to-photon process for a common fluorescent dye of 4-(dicyano-methylene)-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM1), whereas saturating of MEL response indicates that FET is dominant for another common fluorescent dye of Coumarin 540 (C540). (2) In low field (<15 mT), overlapping between MEL responses of DCM1-doped and control OLED shows that prior to being trapped by DCM1 molecule, electron and hole have formed electrostatically bound \( e^-h^+ \) polaron pair in adjacent Alq3 molecules. Both of the observations are finally confirmed by intermolecular correlated quantum calculation.

1. INTRODUCTION

Organic optoelectronic devices such as organic light-emitting devices (OLEDs),1,2 organic solar cells,3,4 and organic memory devices5 are being introduced into real world applications because of their distinctive advantages, that is, ease of fabrication, low cost, light weight, and great mechanical flexibility. The demand for high-efficiency and color-tunable OLED promotes the new architecture such as fluorescent and phosphorescent dye-doped diode.6,7 Two mechanisms, namely, Förster energy transfer (FET)8,9 and charge trapping (CT),10–12 are revealed to dominate the carrier-to-photon process of dye-doped OLEDs. FET is a spectroscopic process by which energy is passed nonradiatively between host molecule and guest molecule (namely, dye in this letter) over long distances (10–100 Å), which is much larger than the molecular diameter. Therefore, FET is a long-range interaction, whereas for CT, when current flows through a device, energy levels of dye in host molecules are deep enough to capture the flowing electrons or holes. Therefore, CT is a short-range interaction on the molecular length scale. The identification about which of FET and CT governs the carrier-to-photon conversion for a given fluorescent-dye-doped OLED is becoming of basic researching and technological importance, as the judgment can guide researchers to use dye properly for achieving higher performances13 or characteristic features.8 Several methods10,12–21 such as the analysis of energy diagrams,13 delayed electroluminescence (EL),14 time-of-flight (TOF),10 spectra overlap properties,21 and the photoluminescence in situ measured during fabrication20 have been explored to judge the issue.

The method of energy diagram provides a general judgment; however, CT can be inferred only from an extreme condition: the energy level (highest occupied molecular orbital or lowest unoccupied molecular orbital) of dye must be deep enough among the energy levels of host molecules. Therefore, the method will not work if the energy levels of dyes are of comparable magnitude to that of host molecules. As for other methods, only one of FET and CT is emphasized. For example, because the CT can influence charge transport in the doped region and TOF can reveal the mobility of carrier, TOF can be exclusively applied to CT. In contrast, photoluminescence in situ determining during fabrication is for FET. Considering FET and CT usually coexist for a given dye-doped OLED, the fundamental issue about which one of two mechanisms dominates is still inconclusive.17–21 There is therefore a need for experimental approaches able to probe the CT and FET simultaneously in the same device.

In fluorescent-dye-doped OLED, fluorescence occurs when the molecule returns to the electronic ground state, from the excited singlet state, by emission of a photon. Commonly, because of spin-forbidden transition of triplet exciton, only singlet excitons can emit through radiative transition. Carrier-to-photon conversion is therefore a spin-dependent process. Meanwhile, organic spintronics has been booming very recently22–27 for its prospect of both basic research on spin dynamics and applications. As one
main part of organic spintronics, sizable magnetic field effects (MFEs) in organic optoelectronic devices have been observed and have the potential applications for magnetically controlled optoelectronic devices and magnetic sensors. MFE comprises change of EL and current in response to the applied magnetic field, which are dubbed in the MFE literatures as magneto-electroluminescence (MEL) and magneto-current (MC), respectively. Here by experimentally and theoretically studying the MEL response of dye-doped OLED based on quantum correlated effect the fundamental issue of judgment about which of CT and FET dominates carrier-to-photon process can be well-identifiable through measuring MEL responses in different magnetic field ranges.

2. EXPERIMENTAL METHODS

Fabrication of Dye-Doped Devices. Figure 1a shows schematically the OLED structures driven at constant voltage in external magnetic field. Tris-(8-hydroxyquinoline) aluminum (Alq3) and N,N'-bis(l-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) function as the electron- and hole-transport layers, respectively. The emission layer is a 20 nm thick Alq3 doped with either C540 or DCM1 and sandwiched between the 40 nm hole transport layer and 80 nm electron transport layer. The evaporation rates of NPB, Alq3, LiF, and Al are 1.5, 1.5, 0.1, and 1.5 Å/s, respectively, measured by calibrated Sigma Instruments SQC 310 rate/thickness monitor. The emission area is approximately 4 mm\(^2\). Three devices were fabricated, which are the C540-doped, DCM1-doped and the nondoped control OLEDs. The volume concentrations of C540 and DCM1 in the doped OLEDs are both 1%.

Measurements and Characterization. The measurement of the device characteristics as shown in Figure 1a was taken in nitrogen environment for protection against oxygen and vapor. Alternating magnetic field was applied by switching on/off an electromagnet. Intensity of the magnetic field was controlled by the current from a high-power source applied to the electromagnet. The strength of the magnetic field was monitored by a Gauss meter with the detection accuracy of 0.1 mT. The photodiode was closely attached to the light emission side of OLEDs. The region with a uniformly distributed magnetic field was about 2 cm\(^2\). The area of each OLED was only 4 mm\(^2\) so that the magnetic field applied across OLED was uniformly distributed. The photodiode was magnetically shielded and was examined to be unaffected by an external magnetic field. Noticeably, MEL was insensitive to the direction of magnetic field. We verified this feature by measuring MEL at different directions of magnetic field. For example, by setting an identical magnetic field parallel or perpendicular to the OLED emitting surface, the measured MEL strength remained the same. Therefore, the response of MEL is sensitive to the magnetic field. Tris-(8-hydroxyquinoline) aluminum (Alq3) and N,N'-bis(l-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) function as the electron- and hole-transport layers, respectively. The emission layer is a 20 nm thick Alq3 doped with either C540 or DCM1 and sandwiched between the 40 nm hole transport layer and 80 nm electron transport layer. The evaporation rates of NPB, Alq3, LiF, and Al are 1.5, 1.5, 0.1, and 1.5 Å/s, respectively, measured by calibrated Sigma Instruments SQC 310 rate/thickness monitor. The emission area is approximately 4 mm\(^2\). Three devices were fabricated, which are the C540-doped, DCM1-doped and the nondoped control OLEDs. The volume concentrations of C540 and DCM1 in the doped OLEDs are both 1%.

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of B. Change of EL is defined as $\Delta EL = (EL(B) - EL(0))$, where $EL(B)$ and $EL(0)$ are the EL intensity with and without B, respectively. $\Delta EL$ increases with the low field (LF) $< 20$ mT, whereas in high field (HF) $> 20$ mT the $\Delta EL$ saturates. The feature is also the characteristic of the control OLED. It should be noted that $\Delta EL$ of the OLED is insensitive to the B direction. Figure 2b shows MEL response of the DCM1-doped OLED. The behavior of $\Delta EL$ in LF resembles that of C540-doped device in Figure 2a. However, in HF, especially for $B > 50$ mT, the $\Delta EL$ response to the field for DCM1-doped OLED dramatically decreases rather than saturates, as in the case of C540-doped OLED (see Figure 2a). Although both C540 and DCM1 are fluorescent dyes, the EL responses to HF are unambiguously different.

It should be noted that in the field of $> 100$ mT, there are sharp peaks existing at the field switching-on and -off edges in Figure 2b. As previously discussed, we found the increasing $\Delta EL$ in LF, followed by the suppressed $\Delta EL$ in HF. The strength of the growing $B$ at the switching-on side is still low in the initial stage, it is thus clear that the variation of the EL is large. However the EL drops as $B$ at the switching-on side grows further toward the high desired value. As a result, we can see a peak at the switching-on edge. The response is similar to the switching-off edge because the strength of the dropping $B$ becomes low and consequently the EL intensity increases and forms a sharp peak.

To quantify the MEL response, we evaluated MEL with the definition of $\Delta EL/EL(0)$ while sweeping $B$. Three devices are under driving condition of a constant initial brightness of 200 cd/m$^2$ at $B = 0$. Figure 3a shows MEL responses of the three devices. We note that for the C540-doped and control OLEDs the profile of MEL response (i.e., $\Delta EL/EL$) maintains the enhancement in LF, followed by saturation in HF. However, the MEL magnitude in C540-doped OLED is stronger than that in control OLED. As reported in ref 29, hyperfine interaction (determined by effective nuclear magnetic field) plays a dominant role in MEL. Because the effective nuclear magnetic field localizes at the H atoms in the host molecule, the hyperfine interaction is therefore a local intermolecular effect. Hopping between adjacent molecules is a feature of charge transport in organic semiconductors, which can determine the intermolecular interaction. The hopping in such a system is therefore a nonlocal intermolecular effect. The weakening of nonlocal hopping rate can increase the influence of localized hyperfine interaction and thus results in stronger intensity of MEL. It is a competition between the local and nonlocal effects that determines the intensity of MEL for many systems.22,28,29

Because of the higher current efficiency shown as the inset of Figure 1b, C540-doped OLED requires lower driving voltage to achieve the same brightness that the control OLED does. At the same time, a lower applied voltage leads to lower carrier mobility.

3. RESULTS

Figure 1b shows the brightness—current—voltage ($B$–$I$–$V$) characteristics of the three devices. The $I$–$V$ characteristics of the three devices are very similar to each other, and their turn-on voltage is around 5 V. However, the brightness of the C540-doped OLEDs is much higher than that of the control and DCM1-doped OLEDs. The current efficiency of the C540-doped OLED is almost two times higher than that of the control and DCM1-doped OLEDs. Figure 1c illustrates the EL spectra of three devices. The undoped control device has an emission from tris-(8-hydroxyquinoline) aluminum (Alq3) with the peak at 520 nm. The C540- and DCM1-doped OLEDs have an emission peak at 510 and 590 nm, respectively, which are correspondingly the characteristics of C540, DCM1 materials. The results indicate that ELs in our dye-doped devices are fully from the dye rather than host materials.

Figure 2a demonstrates the response of EL intensity for the C540-doped OLED to external magnetic field, $B$, driven at a constant current. By continuously switching the magnetic field $B$ on and off, the variations of EL intensity follow the on–off pattern strength but not the direction of the applied magnetic field. This also implied that the MEL studied here was not from the Hall effect, which was very sensitive to the direction of applied magnetic field.

Figure 3. MEL ($\Delta EL/EL(0)$) versus magnetic field of the control OLED and C540-doped and DCM1-doped OLEDs under (a) constant initial brightness of 200 cd/m$^2$ at no external magnetic field, (b) constant voltage driving of 7 V, and (c) constant current driving of 500 $\mu$A.
(ii) At LF (i.e., 0—12 mT), MEL responses for three devices are identical.

(iii) At HF, especially on the scale of >30 mT, the MEL response of the DCM1-doped OLED gradually reduces rather than saturates in CS40-doped and control OLEDs.

Besides investigating the above cases of constant initial brightness (Figure 3a) and constant applied bias (Figure 3b), we also study the response of EML to field under a constant current driving of 500 μA, as plotted in Figure 3c. The trends of the three OLEDs in this case are similar to the cases of constant initial brightness and constant applied bias that, for CS40-doped OLED and control OLED, the MEL consists of fast enhancement in low fields and saturation in high fields, whereas for DCM1-doped OLED, the MEL appears to be decaying in high fields. In addition, the results shown in Figure 3a—c provide clear evidence that MEL responses have LF and HF components.

4. DISCUSSION

To unravel the LF and HF components in MEL response, we must take into consideration four steps in the general carrier-to-photon process in control OLEDs.13,30

Step I. The carrier injection from nonmagnetic electrodes.

Step II. The formation of an electrostatically bound e-h polaron pair in adjacent Alq3 molecules.

Step III. The formation of an exciton through the hopping of the electron or hole in the e-h polaron pair from adjacent sites to one site.

Step IV. Luminescence through the radiative transition of an exciton from excited state to ground state.

Doping dyes at ultralow concentration of 1% into the emitting layer will not affect Step I because the control and two doped OLEDs have similar current—voltage characteristics, as shown in Figure 1b. Moreover, Step IV reflected by photoluminescence is non-magnetic-field dependent.29 Therefore, only Steps II and III possibly dominate MEL response in the studied OLEDs. For an in-depth understanding of roles of Steps II and III in MEL response, here we use the Hubbard model22,31 with some modifications to describe the MEL by considering Steps II and III together in the system.

The Hamiltonian of the Hubbard model is

\[ H = H_1 + H_2 \]

where \( H_1 \) includes hopping and Coulomb interaction of carriers as

\[ H_1 = - \sum_{\langle i,j \rangle, \sigma} (t_{ij}^e a_{i,\sigma}^e a_{j,\sigma}^e + t_{ij}^h c_{i,\sigma}^h c_{j,\sigma}^h + \text{h.c.}) + U \sum_{\langle i,j \rangle} (n_{i,e}^e n_{j,e}^e + n_{i,h}^h n_{j,h}^h) + V \]

where \( \bar{d}_{i,\sigma} (\bar{d}_{i,\sigma}^\dagger) \) and \( \bar{c}_{i,\sigma} (\bar{c}_{i,\sigma}^\dagger) \) are the creation (annihilation) operators of hole and electron, respectively. The hole and electron have spin \( \sigma \), which can be spin up (↑) or down (↓) in the \( i \)-th and \( j \)-th molecules. \( i \) and \( j \) denote the neighbors of the \( i \)-th and \( j \)-th molecule, respectively. \( t_{ij}^e \), \( t_{ij}^h \) is the hopping rate of hole between the \( i \)-th and \( j \)-th molecules (electron between \( j \)-th and \( i \)-th molecules) with units of meV. \( U \) is the Coulomb repulsive energy between two holes or two electrons with different spins at the same molecule. \( \bar{d}_{i,\sigma}^\dagger (\equiv a_{i,\sigma}^h a_{i,\sigma}^e) \) and \( \bar{c}_{i,\sigma}^\dagger (\equiv c_{i,\sigma}^h c_{i,\sigma}^e) \) are the corresponding creation and annihilation operators of the hole and electron, and \( V \) is the attractive interaction between the hole and electron at the same molecule.

\[ H_2 = g \mu_B \sum_{i,j} \left( \vec{B}_{\text{ext}}(i) \cdot \vec{S}_i^h + \vec{B}_{\text{hyp},i}(i) \cdot \vec{S}_i^e \right) \]

where \( g = 2.0 \) for organic materials,77,32 \( \mu_B \) is Bohr magneton, \( \vec{B}_{\text{ext}}(i) \) is the external magnetic field chosen to be along the \( z \) direction, and \( \vec{B}_{\text{hyp},i}(i) \) is the effective nuclear magnetic field of the \( i \)-th molecule, with the value \( ~1 \text{ mT} \).27,32 \( \vec{S}_i^h \) and \( \vec{S}_i^e \) are the classical spin vectors of the hole and electron in the \( i \)-th and \( j \)-th molecules, respectively. Hopping rates for electron and hole are, respectively, 6 and 4 μeV, within range of ref 33. Figure 4a shows the theoretical (using eq 1) and experimental MEL (i.e., ΔEL/EL) at various applied magnetic fields for control device, which coincide with each other. The detailed discussion about theory of the control device can be seen elsewhere.34

It should be noted that the Hamiltonian \( H \) in eq 1 takes into account a number of electrons and holes in organic molecules. For Step II, there are one electron and one hole in the space, and the total number of Alq3 molecules is three, denoted with the molecule indices \( i,j = 1, 2, 3 \), whereas for Step III, there are two electrons and two holes and the total number of molecules is 2. The procedure of our calculation is as follows. For Step II, we treat molecules 1 and 3 as the initial molecules containing the electron and hole. During the time evolution, they will hop to molecule 2 to form a polaron pair. We then calculate the evolution.
and average the values of correlation function for both singlet and triplet in molecule 2 over a time period from 0 to 10 ns for estimating the lifetime of a polaron pair. For step III, the procedure is all the same, except the initial state is that there are two molecules and each has one electron and one hole, that is, a bound polaron pair initially exists in each of the two molecules. We realize that from polaron pair to exciton the coherent hopping of polaron’s charge part does not influence the spin motion. Therefore, the hopping mechanism in this step has been neglected. During these two processes, hyperfine interaction and correlation between molecules play essential roles in MEL because they directly correspond to the spin degrees of freedom.

We found domination of magnetic-field-dependent Step II mainly in LF component (i.e., characteristic fast enhancement) (<25 mT), followed by domination of magnetic-field-dependent Step III in HF component (characteristic saturation) (>35 mT). Range from 20 to 35 mT is the merging region of LF and HF components. In the region, the theoretical results based on Steps II and III, respectively, show decaying and switch-on features. To stress and unravel roles of Steps II and III in LF and HF components, we do not give the theoretical result in the transition region here. One can see the whole LF component including the feature in transition regime in the following discussion about MEL response of DCM1-doped device.

Because LF/HF component is associated with magnetic-field-dependent Step II/III, it is ready to identify the carrier-to-photon conversion mechanism in dye-doped OLEDs by observing their MEL responses in Figure 3a and 3b. For observation (i) provided by Figure 3b, LF and HF components in C540-doped OLED are the same as those in control device. It can be thus concluded that the doping C540 cannot influence Steps II and III in host Alq3 molecules. In other words, the exciton preferentially forms at Alq3 molecule in both devices. In addition, because the EL spectrum of the C540-doped OLED is fully from C540 (Figure 1c), it indicates that after exciton formation in Alq3 the exciton transfers its energy to C540 molecule to excite an exciton, as schematically shown in Figure 5a. The carrier-to-photon processes in the control and C540-doped OLEDs in energy levels are, respectively, summarized in Figure 5c,d. This provides evidence that FET is dominant in carrier-to-photon conversion for the C540-doped OLED. For the observation (ii), LF component of DCM1-doped OLED coincides with that of control OLED, which signifies that Step II in Alq3 is unaffected. For the observation (iii), the MEL response for DCM1-doped OLED in HF dramatically decreases; namely, HF component is removed. To confirm further that LF component can be unraveled by doping DCM1, Figure 4b shows both theoretical and experimental results based on eq 1. Differently, only Step II is considered. The theoretical result of MEL response shows the same feature as the experimental result, namely, fast increase, followed by dramatic decrease. Noticeably, the dramatic decrease in theory is just the missing part of LF component in the transition region shown in Figure 4a. The well-fitting by using the one-step model further confirms that Step III that is associated with HF component is diminished due to the DCM1 doping. Moreover, the EL spectrum of the DCM1-doped OLED is mainly from DCM1 (Figure 1c), and electron and hole in the preferentially formed e-h polaron pair residing in Alq3 molecules are directly trapped by the adjacent DCM1 molecule, followed by the formation of exciton in DCM1 instead of Alq3, as schematically shown in Figure 5b. The whole process in the energy level is summarized in Figure 5e. Consequently, the exciton in DCM1 is generated by trapping electron and hole simultaneously from the e-h polaron pair in Alq3. On the basis of the above results, it is remarkable that MEL responses of the three OLEDs can serve as a fingerprint to identify the process of carrier-to-photon conversion in dye-doped OLEDs.

5. CONCLUSIONS

In conclusion, we systematically studied the MEL response of dye-doped OLEDs including C540- and DCM1-doped devices. Coexistence of fast-increasing low-field and saturating high-field components are observed in MEL responses of C540-doped and control device. Low-field and high-field components are due to magnetic-field-dependent formation of e-h polaron pair and exciton in host molecule, respectively. Therefore, overlapping MEL responses between C540-doped device and control device are assigned to the domination of Förster energy transfer in carrier-to-photon process for C540. Whereas for the DCM1-doped OLED, low-field component is unraveled because of expelling of the exciton-forming step in the host, which implies that charge trapping dominates the carrier-to-photon process for DCM1. Theoretical studies based on the modified Hubbard model provide strong evidence of the above analysis. The study paves the way of using the magnetic field effects as a fingerprint for studying the conversion process of all other dye-doped
optoelectronic device and establishes a foundation for understanding the MEL response in the whole range of magnetic field.

**ASSOCIATED CONTENT**

Supporting Information. MEL of other dye-doped OLED is also briefly introduced in the text and figure. This material is available free of charge via the Internet at http://pubs.acs.org.

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**REFERENCES**