Identify Triplet-charge Interaction in Rubrene-based Diodes Using Magneto-conductance: Coexistence of Dissociation and Scattering Channels

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ABSTRACT

Although it is known that triplet excitons can be quenched via triplet-charge interaction (TQI), it is still unclear how this process occurs in rubrene-based devices. We found that magneto-conductance (MC) can be used to probe the detailed mechanism of TQI in rubrene-based organic light-emitting diodes and observed the coexistence of negative and positive MC responses in the high-field region when holes and electrons were the dominant charged species at different interfaces adjoining the rubrene layer, respectively. Further analysis suggests that the negative MC response was originated from the dissociation of triplet excitons by holes, while the positive MC response was due to electron scattering by triplet excitons. The MC responses of the devices were examined under different injection currents and temperatures to confirm our hypothesis. This work gives significant insight into mechanisms of TQI in organic semiconductors, which will allow for the design of new and improved devices.

Keywords: triplet-charge interaction, dissociation channel, scattering channel, magneto-conductance

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1. Introduction

Rubrene has attracted great interests in the scientific community because of its unique optoelectronic properties that makes it desirable for use in organic semiconductor devices. For example, the energy of a singlet exciton within rubrene is resonant with that of two triplet excitons ($E_{S_1} \approx 2E_{T_1}$)[1-5], which facilitates bimolecular reactions, including singlet fission (STT) and triplet fusion (TTA), ($S_1 + S_0 \leftrightarrow T_1 + T_1$)[4, 6-8]. STT can lead to highly efficient organic solar cells (OSCs)[9], while TTA can increase the theoretical quantum efficiency of fluorescent organic light-emitting diodes (OLEDs) up to 62.5%[10, 11]. And the utilization of triplet excitons is especially important for these two processes in rubrene-based devices. However, the high densities of triplet excitons that are generated in rubrene-based devices through STT can be quenched via triplet-charge interaction (TQI)[4]. Therefore, it is important to understand TQI in triplet exciton-rich organic semiconducting devices.

TQI generally occurs via two magnetic field (B) sensitive channels: (i) the dissociation channel, in which the triplet excitons are dissociated into free electrons (e) and holes (h) when they react with free (C) or trapped (C_t) carriers ($T+C/C_t \rightarrow e+h+C$)[12-14] and (ii) the scattering channel, in which triplet excitons collide with free carriers ($T+C \rightarrow T+C$), resulting in a decrease of charge mobility[12, 15]. The presence of an external B can suppress their rate constants[16], which causes the generation of magneto-conductance (MC) signals with opposite signs. In 2007, Desai *et al.* proposed that a positive MC response was due to free carrier scattering by triplets while a negative MC response was caused by triplet dissociation[12]. Hu. *et al.* observed a negative MC response and attributed it to triplet dissociation that was caused by interactions with secondary charge carriers in the unbalanced charge injection conditions that were present[15]. Recently, we have observed that localized triplet excitons and triplet charge-transfer-complexes play crucial roles in the scattering and dissociation channels of TQI[17], respectively. However, the presence of TQI in rubrene-based devices is seldom reported, especially it is unclear whether the TQI process undergoes via dissociation or scattering channels.

In this work, the MC effects of four different rubrene-based devices were measured at a range of operating currents and temperatures. The MC curves were strongly affected by the polarity of the charges (electrons and holes) that had accumulated at different interfaces near rubrene layer existed in the devices and could be fitted to the sum of one Lorentzian and two non-Lorentzian empirical formulas. Further analysis indicated that the distinct changes that were observed in the high B range of the MC effects were caused by the dissociation and scattering channels of TQI that coexisted. The dissociation of triplet excitons by holes resulted a negative MC response while the scattering of electrons by triplet excitons generated a positive MC response. This work showed that MC can be used to identify the different TQI processes that exist in a device, and will enable the design and optimization of device structures that minimize the quenching of triplet excitons.

2. Experimental section

Rubrene was used as the fluorescent emissive layer, N,N' -di(naphthalene-1-yl)- N,N' -diphenyl-benzidine (NPB) was used as a hole-transporting material, 4,4',4''-tris(N -3-methylphenyl- N -phenylamino)triphenylamine (m -MTDATA) was used as an electron-blocking material, bathocuproine (BCP) was used as an electron-transporting and hole-blocking material, *tris*-(8-hydroxy-quinoline)aluminum (Alq_3) was used as an electron-transporting layer, and lithium fluoride (LiF) was used to improve the electron injection efficiency.

Four different device structures were used to investigate TQI by modifying the accumulation of charges at different interfaces near emissive layer within the devices. They had the following configurations: ITO/NPB (60 nm)/rubrene (30 nm)/BCP (50 nm)/LiF (1 nm)/Al (120 nm) (Dev.1), ITO/NPB (60 nm)/rubrene (30 nm)/ Alq_3 (50 nm)/LiF (1 nm)/Al (120 nm) (Dev.2), ITO/ m -MTDATA (60 nm)/rubrene (30 nm)/BCP (50 nm)/LiF (1 nm)/Al (120 nm) (Dev.3), and ITO/NPB (60 nm)/rubrene (30 nm)/BCP (50 nm)/Al (120 nm) (Dev.4). The devices contained indium tin oxide (ITO) and aluminum (Al) electrodes with active areas of $2 \text{ mm} \times 2 \text{ mm}$ that were deposited by thermal evaporation under vacuum ($\sim 10^{-6}$ Pa). The de-

vices were mounted to the cold finger of a closed-cycle cryostat (Janis: CCS-350S), which was located between the poles of an electromagnet (Lakeshore EM647). A B with a maximum strength of ± 500 mT was applied in a direction parallel to the device surface. A Keithley 2400 multimeter was used to provide the voltage bias and record the current signals, while a Keithley 2000 multimeter was used to record the EL intensity that was measured using a Si photodiode.

3. Results and discussion

Fig. 1. (a) MEL responses (the expansion shows the MEL up to 100 mT) and normalized EL spectra (top inset) of Dev.1 and 2 under an injection current of 100 μ A at R.T. (b) and (c) MC curves of Dev.1 and 2 with an injection current of 100 μ A at R.T.

The normalized electroluminescence (EL) spectra of Dev.1 and 2 are shown in the Fig. 1a (insert). The emission maxima (563 nm) and low energy shoulder peaks (600 nm) of the

two devices were the same, which was consistent with the EL emission of rubrene[4]. Dev.2 exhibited an additional high energy shoulder (~523 nm) that was consistent with the EL emission of Alq₃[18, 19]. The magneto-electroluminescence (MEL) curves of these devices that were subjected to an injection current of 100 μ A at room temperature (R.T.) are shown in Fig. 1a. MEL is defined as $MEL = [EL(B) - EL(0)]/EL(0) \times 100\%$, where $EL(B)$ and $EL(0)$ are the EL intensities with and without an external B , respectively. The MEL curves of Dev.1 and 2 exhibited similar line shapes at high B strength, which was consistent with the MEL fingerprint of a STT process[3]. However, as illustrated in the expansion of Fig. 1a, the MEL curve of Dev.1 shown a slight decrease within $0 < B < 20$ mT while that of Dev.2 increased sharply at low B strength and saturated at 5 mT. This typical increase in several mT can be attributed to the hyperfine interaction (HFI) model in which a B suppresses the hyperfine mixing of singlet and triplet polaron-pairs that occur on Alq₃ molecules[18, 20].

The MC of Dev.1 and 2 were measured at R.T. with an injection current of approximately 100 μ A, as shown in Fig. 1b and 1c, respectively. MC is defined as $MC = [I(B) - I(0)]/I(0) \times 100\%$, where $I(B)$ and $I(0)$ represent the currents in the presence and absence of an external B , respectively. The MC curve of Dev.1 increased sharply at low B strength ($|B| < 8$ mT) (low field MC, MC_L) and then dropped quickly as the B strength increased from 8 to 100 mT (intermediated field MC, MC_I), at last increased slowly at higher B strength ($|B| > 100$ mT) (high field MC, MC_H). Conversely, the MC curve of Dev.2 increased rapidly at low B strength ($|B| < 10$ mT) and then increased slowly ($|B| > 10$ mT) before it saturated at 500 mT. Further analysis of MC has shown that the response at low B strength can be attributed to the typical HFI model[21, 22]. However, it has been reported that MC caused by HFI gradually saturates at R.T. in the range $|B| > 50$ mT[21, 23, 24], which is clearly different from our experimental results. This suggests that a different mechanism may cause the observed MC effects at high fields in our devices. As stated previously, Dev.1 and 2 exhibited significantly different MC behavior, despite the similarity of their MEL curves at high B

strength. Therefore, the MC and MEL responses of these devices cannot be caused by the same mechanism (STT model).

Fig. 2. (a) Energy level diagrams of Dev.1 and 2. The electron-transporting layer of Dev.1 is BCP while that of Dev.2 is Alq₃. (b) Schematic diagram showing the scattering of electrons by triplet excitons that occurs in the rubrene-based devices (black arrows around electrons represent its direction of motion). (c) Schematic diagram showing triplet exciton dissociation by holes in the devices. The e, h and h...e represent electron, hole and triplet exciton respectively.

As neither the HFI nor the STT models can explain the shapes of MC curves that were observed over the entire B range, we proposed that the underlying mechanism may be TQI. This proposal can be explained using energy level diagrams of devices that contain rubrene. The energy-level structure of Dev.1 (Fig. 2a) allows for the accumulation of electrons in the rubrene layer near the NPB/rubrene interface because of the high electron injection barrier from rubrene to NPB (approximately 0.9 eV). Additionally, the high hole injection barrier for BCP (~1.1 eV) may induce the accumulation of holes in the rubrene layer near the rubrene/BCP interface. In contrast, Dev.2 contains Alq₃, which has a lower hole injection barrier for Alq₃ (only 0.3 eV). Thus, holes can diffuse across the rubrene/Alq₃ interface and then

recombine with electrons in Alq₃ layer, as demonstrated by the EL spectrum and MEL curve in Fig. 1a. Therefore, Dev.2 accumulates more electrons than holes in the rubrene layer.

Additionally, up to 90% of singlet excitons in rubrene-based devices can be converted into triplet excitons through the STT process at R.T.[4]. And as the STT process occurs on picosecond time scales[25] while the triplet states within rubrene have lifetimes of 100 ± 20 μ s[2], the number of triplet excitons that are generated is larger than those that are quenched. Hence, there is a high density of triplet excitons in rubrene-based devices at R.T. Moreover, triplet excitons within rubrene can diffuse distances of 2–8 μ m[26, 27], which is much larger than the thickness of the rubrene layer used in our devices (~ 30 nm). Thus, triplet excitons within rubrene could easily reach the NPB/rubrene and rubrene/BCP interfaces, which would lead to TQI[13, 14]. Although the TQI is weaker than other exciton deactivation routes such as STT and TTA, we can still identify detailed process of it by using MC as a powerful probing tool.

As mentioned previously, triplet excitons that are involved in the dissociation channel of TQI will decompose into free electrons and holes, which contribute to the current within a device[12-14]. However, triplet excitons that participate in the scattering channel of TQI collide with free charge carriers, which leads to a decrease in the mobility of the charge carriers[12, 15]. An applied B suppresses TQI[16], which results in a negative MC response if the dissociation channel occurs and a positive MC response if the scattering channel occurs. Hence, the sign of the MC effect is a simple and effective way to distinguish between the dissociation and scattering channels of TQI.

The polarity of charge carrier accumulated within Dev.1 and 2, and the corresponding MC curves of these devices indicated that electrons and holes indeed play important roles in TQI. The positive MC effect of Dev.1 and 2 was caused by the scattering of accumulated electrons by triplet excitons, as illustrated in Fig. 2b. In contrast, the accumulation of holes in Dev.1 caused the negative MC results because of triplet exciton dissociation (Fig. 2c). Thus,

triplet excitons can be quenched in the presence of excess holes, which can be minimized when designing devices.

Fig. 3. (a) The normalized EL spectra of Dev.3 and 4 at R.T. The left inset shows energy level diagram of Dev.3. (b) and (c) MC curves of Dev. 3 and 4 at an injection current of 100 μ A at R.T., respectively.

Devices in which holes could accumulate (Dev.3 and 4) were fabricated to further investigate our proposal. These devices had the following structures: ITO/*m*-MTDATA/rubrene/BCP/LiF/Al (Dev.3) and ITO/NPB/rubrene/BCP/Al (Dev.4). *m*-MTDATA was used in place of NPB in Dev.3 because it has a highest occupied molecular orbital (HOMO) level between the Fermi-level of ITO and the HOMO of rubrene (the inset of Fig. 3a), which creates a ladder-type energy structure in which hole injection and transport are improved[28]. This gave a device in which holes were dominant when compared with Dev.1. The LiF layer was not used in Dev.4 to lower the efficiency of electron injection[29], which would also led to a device in which holes were dominant compared with Dev.1.

The normalized EL spectra of Dev.3 and 4 are shown in Fig. 3a, which was consistent with the EL emission of Dev.1. The MC curves of Dev.3 and 4 are displayed in Fig. 3b and

3c, respectively. Both curves exhibited negative MC effects at high B strength, which indicated that the dissociation channel of TQI was dominant because of the increased proportion of holes. And effects of the scattering channel were negligible, which supported the proposal discussed in the previous section. Additionally, the maximum MC that was observed in Dev.3 at high B strength indicated that this device contained more holes than the others because of the ladder-type energy level structure.

Fig. 4. MC responses of Dev.1–4 with different injection currents at R.T.

Next, we investigated the dependence of the MC responses of the four devices on injection currents at R.T., as shown in Fig. 4. Lowering the current only made significant changes to the MC line shapes of Dev.1 (Fig. 4a), i.e., the MC_I ($|B| < 100$ mT, negative) and MC_H ($|B| > 100$ mT, positive) decreased when the current was reduced from 200 μ A to 5 μ A. To make a quantitative analysis on TQI, we tried to use an empirical formula that consisted of a Lorentzian component and two non-Lorentzian components to fit the MC data of Dev.1 in

which the two channels coexist. The Lorentzian function (first component, blue dashed lines in Fig. 5) models HFI (MC_L), while the two non-Lorentzian functions (last components, olive and magenta dashed lines in Fig. 5) model the TQI processes (the negative MC_I and positive MC_H , which are the dissociation and scattering channels, respectively)[11, 30-33]. The function used is shown in eq 1:

$$MC = a_1 * \frac{B^2}{(|B|^2 + B_{01}^2)} + a_2 * \frac{B^2}{(|B| + B_{02})^2} + a_3 * \frac{B^2}{(|B| + B_{03})^2} \quad (1),$$

where B is the applied magnetic field, a_1 , a_2 , and a_3 are pre-factors that represent the intensities of the three components and B_{01} , B_{02} and B_{03} are the characteristic magnetic fields of MC_L (3 ± 0.5 mT), MC_I (50 ± 5 mT) and MC_H (100 ± 3 mT), respectively. Notably, B_{01} is related to the strength of the hyperfine field, which was consistent with that reported in the literatures[34-36]. Importantly, the characteristic magnetic field parameter for the dissociation channel (B_{02}) was significantly different than that of the scattering channel (B_{03}), which implied that the hole-exciton and electron-exciton interactions were indeed different. This behavior between electrons and holes has also been observed by Bloom *et al* when a device was changed from unipolar to bipolar conduction conditions[33].

Fig. 5 The MC curves (black squares) and their fitted lines (red solid lines) obtained (a) under an injection current of 10 μA at R.T. and (b) under an injection current of 100 μA at 20 K. The dashed lines are the MC fingerprints for HFI model (blue), dissociation channel (olive) and scattering channel (magenta).

The final fitting curve (a representative data measured R.T.) is shown in Fig. 5a, which matches the experimental data well. The parameters a_1 , a_2 and a_3 that were used in fitting formula are summarized in Table 1, and give a clear indication as to the different mechanisms that were occurring at different injection currents. As the injection current was reduced the value of a_1 increased, which was consistent with the literature[23], while the values of a_2 and a_3 showed a monotonic rise.

Table 1

Fitting parameters for eq 1 from the MC responses of Dev.1 at R.T. with different injection currents.

	a_1	a_2	a_3
200 μA	0.0383	-0.9684	1.0355
100 μA	0.0752	-0.9010	0.9815
50 μA	0.0845	-0.8240	0.8860
10 μA	0.0805	-0.4880	0.4620
5 μA	0.0720	-0.3760	0.3275

This rise in the values of a_2 and a_3 indicated that the density of triplet excitons and charges modulated the TQI process. An increase in injection current would increase the density of both triplet excitons and charges within the devices, which would enhance their interactions. Similar, yet less obvious, trends were observed in the MC behavior of Dev.2 and 4 at higher B strength (Fig. 4b and 4d, respectively). The negative MC effect that was observed in Dev.3 increased initially and then decreased with the injection current (Fig. 4c). This may have been because the TQI process had saturated at a particular current, which caused a change in $I(0)$ that was greater than that of $[I(B)-I(0)]$.

Fig. 6. MC responses of Dev.1–4 at different temperatures with an injection current of 100 μ A.

We examined the MC behavior of the four devices at high B strength when the temperature was lowered because the lifetime and number of triplet excitons and the rate constant of TQI are all dependent on the temperature of the devices. All four devices exhibited MC responses that were significantly dependent upon the temperature at an injection current of 100 μ A (Fig. 6). At low B strength the MC curves did not show any significant differences as the temperature of the devices was decreased. However, at higher B strengths the MC response of Dev.1, 3 and 4 became increasingly negative as the temperature was lowered from R.T. to 20 K. The positive MC response of Dev.1 weakened with decreasing temperature and eventually disappeared, while that of Dev.2 increased initially and then began to decrease. The MC curves from Dev.1 at different temperatures were also fitted to eq 1 (a representative data is displayed in Fig. 5b). The parameters of the fits are summarized in Table 2.

Table 2

Fitting parameters for eq 1 from MC responses of Dev.1 at different temperatures with an injection current of 100 μA .

	a_1	a_2	a_3
R. T.	0.0752	-0.9010	0.9815
160 K	0.1433	-1.1750	0.8562
120 K	0.1601	-1.3985	0.8187
100 K	0.1650	-1.4408	0.3674
50 K	0.1690	-1.6863	0.1912
20 K	0.1703	-1.7700	0.0000

The results shown in Table 2 suggested that the dissociation channel of TQI was dominant at low temperatures. As stated previously, the MC at high B strength is closely correlated with temperature. When the temperature was decreased the value of a_2 increased, while the value of a_3 decreased to 0 at 20 K. The lifetime of triplet excitons is prolonged in the dissociation channel at low temperatures because of a reduction in the phonon-assisted decay[37]. Thus, the rate of interactions that lead to TQI is increased, which increases the probability of the dissociation channel occurring and leads to increased values of a_2 at low temperatures. This process also applies to Dev.3 and 4. However, the scattering channel of TQI is dependent upon the lifetime of the triplet excitons and the velocity of both the electrons and the triplet excitons. This is because the scattering channel involves a series of collisions that require a certain kinetic energy. Thus, lower temperatures will slow down the triplet excitons and electrons, which lead to a decrease in the rate of interactions and the value of a_3 . Note that the MC values of Dev.2 at high B strength did not decrease monotonically with decreasing temperatures, which may have been caused by the increased lifetime of the triplet excitons and a decrease in the velocity of the electrons and triplet excitons at low temperature.

4. Conclusions

In summary, we fabricated four rubrene-based OLEDs that contained an excess of either electrons or holes under operation to understand the differences between the dissociation and scattering channels of TQI. The two channels of TQI were found to coexist and were qualitatively analyzed by measuring the MC curves of the devices at different temperatures and injection currents. The dissociation of triplet excitons by holes generated a negative MC response and the scattering of electrons by triplet excitons produced a positive MC response. This work provides a deeper understanding of two mechanisms of TQI in organic semiconductors and will help optimize devices. However, it remains unclear why holes and electrons take part in different channels, and why they exhibit their characteristic magnetic fields widths. We speculate these might be because the spatial extent and localization of the holes and electrons themselves. And efforts are currently being undertaken to understand these important questions.

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(a)

Scattering channel

Dissociation channel

(b)