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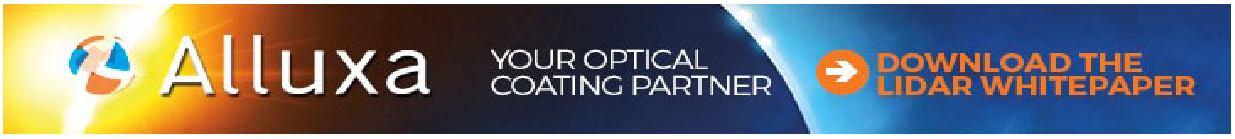

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ABSTRACT

The bias dependent behaviors in magnetic field effects (MFEs) of the current and the electroluminescence (EL) intensity in organic light emitting diodes (OLEDs) have been investigated from electrically-detected and EL-detected magnetic resonance (EDMR and ELDMR) techniques. An EDMR signal was not detected from the electron-only device, and the hole-only device gave only a much smaller EDMR signal than the OLED device. Both the EDMR and ELDMR signals observed from the OLED are concluded to primarily arise from the spin-dependent reaction of electron-hole (e-h) pairs. Both the normalized EDMR and ELDMR signal intensities decrease by increasing the operation bias of OLED, because the increased bias enhances the dissociation and recombination of e-h pairs beyond the increase in the pair-density by the bias. The bias-dependence curves of magneto-conductances and magneto-EL intensities are demonstrated to be very similar to those of the normalized EDMR and ELDMR, respectively. This similarity gives direct evidence that e-h pairs determine the MFEs of the present OLEDs at room temperature and that the MFEs are reduced by bias-dependent dissociation and recombination of e-h pairs. The bias-dependent EDMR and ELDMR experiments are thus effective as probing methods to examine the magnetic field properties via e-h pairs of OLEDs.

I. INTRODUCTION

It has often been reported that prominent magnetic field effects (MFEs) in the conductivity and the electroluminescence (EL) intensity are observed from organic light emitting diodes (OLEDs).[1–8] In the MFEs, various factors can be considered as a source of the magnetic properties. For instance, the operation of the OLED undergoes generation and dissipation processes of carriers, excitons, and electron-hole (e-h) pairs, some of which may be affected by the magnetic field. Also, the MFEs may occur via particular magneto-responsive species. Indeed, many species, such as an e-h pair (polaron pair),[3,4,6,8–15] a triplet exciton (TE),[16–18] a trap carrier, and a doubly-charged spinless bipolaron,[19–21] have been considered as the main species of MFEs. Due to such many factors to be considered, multiple mechanisms and explanations on MFEs have been proposed.[22,23] Also, as another reason for multiple models to exist, experiments of MFEs typically measure simply a change of the current and the EL intensity in the steady-state [magneto-conductance (MC) and magneto-electroluminescence (MEL), respectively], and they may allow several interpretations for the results. Thus, it would be difficult to evaluate the validity of a model only from the consistency of fitting curves for the MC or MEL intensity. It is therefore desirable to use other experimental techniques in parallel that can provide supplementary information in addition to the MC and MEL measurements to identify the main factor that induces the MFEs.

In the research of the magnetic properties, an electron spin resonance (ESR) technique is often regarded as effective because it allows electronic state evaluation of paramagnetic species. However, since a conventional ESR technique that measures microwave absorption is sensitive to all paramagnetic species, the ESR signal could be dominated by signals from carriers and defects generated under OLED operation, and thus, selective detection of magneto-responsive species would be difficult. Also, the conventional ESR method is not sensitive enough to detect signals from a small density of spins as produced in thin film OLEDs. By contrast, electrically-detected and EL-detected magnetic resonance (EDMR and ELDMR) techniques measure a change of the current and the EL-intensity, respectively, at the moment of ESR and enable sensitive and selective detection of ESR signals related to the current and EL. These techniques should thus be effective for the research of MFEs in OLEDs. In particular, whereas MFEs generally occur via Zeeman splitting between spin sublevels by applying a magnetic field, ESR induces a transition between the sublevels so as to reduce the population difference made by the Zeeman splitting. A strong correlation should thus be expected between the signals from MFEs and the ESR measurements.

In this article, properties of MC and MEL in OLEDs are explored from EDMR and ELDMR techniques. A simple OLED consisting of a single active layer was employed to simplify the discussion on magneto-dependent OLED processes. The MFEs and ESR characteristics are compared for the simple OLEDs fabricated in exactly the same conditions. We particularly focus on the bias voltage dependence of those properties. Recently, we showed that the normalized intensities of electrically-detected and electroluminescence (EL)-detected magnetic resonance (EDMR and ELDMR, respectively) signals of OLEDs, calculated by the EDMR and ELDMR intensities divided by the dark current and the EL intensity, respectively, are reduced by increasing the operation bias of OLED.[24] The signal-reduction was attributed to the dissociation of e-h pairs by the electric field.[24] Here, we discuss the bias-dependent ESR properties of OLED in detail through comparison of those with hole-only and electron-only devices, and thereby, we address the role of e-h pairs on the ESR signals and MFEs in both the current and EL properties. In particular, the results of the bias-dependence experiments indicate that both the efficiencies of MC and MEL are reduced by increasing the forward bias due to the dissociation of e-h pair. This provides direct experimental evidence that the bias-dependent pair-dissociation is an important factor determining the efficiency of MFEs in OLEDs and also indicates that the EDMR and ELDMR techniques give straightforward information on their magnetic properties.

II. EXPERIMENTS

Superyellow (SY) poly(-*para*-phenylene vinylene) (PPV) purchased from Merck was used for the active layer of OLED. The OLED structure was ITO/MoO_3/SY-PPV/PEI/Al, where ITO is the transparent indium–tin–oxide coated glass substrate and PEI is a polyethyleneimine layer. The SY-PPV layer was spin-casted from its chlorobenzene solution (5 mg/ml) on the MoO_3 layer vacuum-deposited on the ITO substrate (anode). The PEI layer was spin-casted on the SY-layer from the 1-propanol solution (0.5 mg/ml) and Al was vacuum-deposited on the PEI layer for the cathode (30 nm). The total thickness of the PPV layer including the PEI layer was about 100 nm. The devices with the structures of ITO/MoO_3/SY-PPV/MoO_3/Al and ITO/PEI/SY-PPV/PEI/Al were also fabricated as the hole-only and electron-only diodes. All device-fabrications were done in the nitrogen-filled glovebox. Each device was then loaded into a glass cell in the glovebox and used for ESR and MFE measurements under vacuum evacuation.

A partly-modified conventional ESR spectrometer (JES-FE1XG, JEOL) was used for all ESR measurements. Changes of the current density and the EL intensity induced by ESR were measured by recording their lock-in signals synchronized with the microwave modulation (140 mW, 1.1 kHz). The EL intensity was measured with a photodiode for the EL-output through an optical fiber inserted in the ESR cavity. The lock-in signals were measured with a dual-phase mode (SR-830, SRS) typically at a reference phase of 180°, by which the sign of the in-phase lock-in signal was ascertained to approximately match the actual sign of the signal identified from an oscilloscope in the measurement system used in this study. The bias-dependent EDMR and ELDMR measurements were performed simultaneously by recording the respective lock-in signals while sweeping the bias voltage. The ESR response from the off-resonance signal induced by the microwave modulation was eliminated. The magnetic field response of the current and the EL intensity was measured for the OLED located between the poles of an electromagnet. The magnetic field dependence of the current and the EL-intensity was obtained by measuring each response to the magnetic field modulation (2G, 80 Hz) while sweeping the magnetic field and integrating the obtained differential response over the magnetic field. For the bias-dependence of MC and MEL, responses of the current and EL intensity, respectively, for the ±20G-magnetic field modulation at 20G were recorded using a lock-in amplifier while sweeping the bias for the OLED. All measurements were performed at room temperature.

III. RESULTS AND DISCUSSIONS

A. ESR characteristics

Figures 1(a) and 1(b) show the EDMR and ELDMR spectra of SY-OLED under 3.5 V-bias measured simultaneously with a dual-phase lock-in technique. The EDMR signal has two phase components shown as the in-phase and quadrature components. The origin of the quadrature EDMR signal is discussed later. The in-phase EDMR and ELDMR spectra resemble each other. This spectral resemblance was also confirmed at other bias-voltages.[24] The EDMR and ELDMR signals are thus given by common ESR transitions regardless of the bias. EDMR measurements were also performed for the electron-only device (ITO/PEI/SY-PPV/PEI/Al) and the hole-only device (ITO/MoO_3/SY-PPV/MoO_3/Al) of SY-PPV [Fig. 1(c)]. The current-voltage characteristics of the devices are shown in the inset of Fig. 1(c). The EDMR signal was not detected from the electron-only device under the bias of 4 V. Also, the hole-only device gave only a much smaller EDMR signal than the OLED sample under 2.5 V where the current is comparable with that of the OLED under 3.5 V. The observed EDMR and ELDMR signals of OLED are thus not given by products from unipolar carriers, such as bipolarons, but by products containing electrons and holes. Such products are not only e-h pairs but could be trions consisting of a TE and a carrier or consisting of a bipolaron and its counter-carrier. In fact, the EDMR and ELDMR signals of OLEDs were previously explained by being due to the trions.[25,26] Contributions from the former trions can be examined from EDMR or ELDMR responses at the half

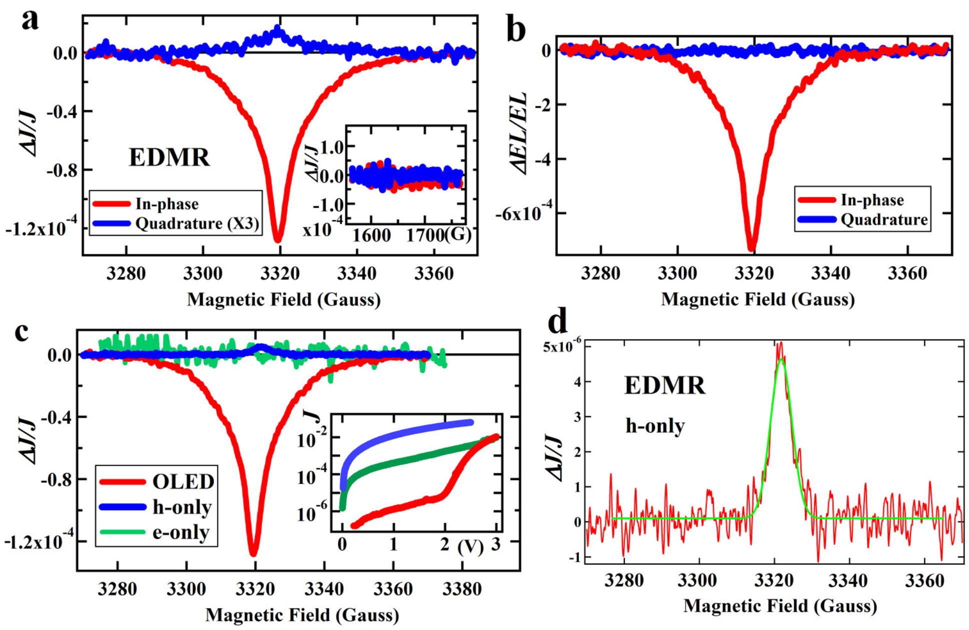

FIG. 1. The in-phase and quadrature-phase EDMR (a) and ELDMR (b) spectra measured under 3.5 V-bias for the OLED (ITO/MoO_3/SY-PPV/PEI/Al) by dual-phase lock-in techniques. The inset of (a) is the result of half-field EDMR measurement for the OLED under 4.0 V-bias. (c) Comparison of the EDMR spectrum of the OLED shown in (a) with those of the hole-only (h-only) device (ITO/MoO_3/SY-PPV/MoO_3/Al) under 2.5 V and the electron-only (e-only) device of SY-PPV (ITO/PEI/SY-PPV/PEI/Al) under 4 V. The inset of (c): Current density J (Acm^{-2})-voltage characteristics for each device. (d) The EDMR spectrum of the h-only device measured under 2.5 V-bias (Red). The green curve is the result of spectral fit using a single Gaussian curve with the full width half maximum (FWHM) of 4.0G.

magnetic field where the spin transition of $\Delta m_s = 2$ may be observed from TEs.[26,27] Indeed, a half-field signal was not detected from the present OLED. This result is consistent with the recent report that such half-field EDMR signals were observed only at low temperature in polymer OLEDs.[28,29]

Related to the trion generated via the bipolaron, the weak EDMR signal observed in the hole-only device could be a result of spin-dependent reaction of positive polaron carriers into the bipolaron. Then, if the EDMR and ELDMR signals of OLED occur via the trion, the trion should be formed by the spin-dependent reactions of the positive bipolaron and the negative countercarrier. However, since the bipolaron formation is presumed to be only slight judging from the EDMR signal of the hole-only device, the reaction of the trion-formation is unlikely to occur more frequently than the formation reaction from the singly-charged e-h pairs. We therefore conclude that both the observed EDMR and ELDMR signals are primarily given by the spin-dependent reaction of e-h pairs. This conclusion is also consistent with the conclusions drawn from pulsed EDMR and ELDMR measurements.[28,30] In this case, the EDMR and ELDMR spectra can be explained by the sum of spectral components from hole and electron carriers.[31,32] In reality, the observed spectra can be reproduced by the sum of two Gaussian curves with the full width half maximum (FWHM) of 3.1 G and 14.5 G.[24] Interestingly, the EDMR spectrum of the hole-only device was fitted well with a single Gaussian curve with a FWHM of 4.0 G: note that the FWHM is almost independent of the bias (see the supplementary material) and this width is relatively close to the smaller width of the OLED sample. Moreover, the feature of the smaller width in the hole carrier corresponds well with the recent reports that the MC curve of the hole-only device in polymer OLEDs is narrowed than that of the electron-only device.[33] We thus conclude that the smaller width component of the EDMR and ELDMR spectra in the OLED is given by the hole carrier. This is consistent with the conclusion of the recent report drawn from a comparison of the measured and simulated EDMR spectra in the high-magnetic field EDMR for the OLED of 2-methoxy, 5-(2′-ethyl-hexyloxy)-(MEH-) PPV.[34]

In the model of EDMR and ELDMR considering the spin-dependent reaction of e-h pairs,[35,36] an equilibrium relation is assumed to hold among free polaron carriers, e-h pairs, and excitons. An ESR transition occurring at sublevels of triplet e-h pair (TP) slightly changes the density-ratio between singlet e-h pair (SP) and TP through spin-mixing of S and T_0. The change of the pair-density leads to changing the densities of carriers and luminescent excitons, giving rise to EDMR and optically-detected magnetic resonance (ODMR) signals, respectively.[35–37] We observed negative ELDMR signals, indicating that the density of SP n_{SP} is reduced by the ESR transition. We also observed negative EDMR signals. Considering the ESR-induced reduction of n_{SP}, the negative EDMR signal indicates the relation of $d_{SP}\, k_{TPE} > d_{TP}\, k_{SPE}$, where d_{SP} and d_{TP} are the dissociation rates of SP and TP into carriers, respectively, and k_{SPE} and k_{TPE} are the transition rates of SP to singlet exciton and TP to TE, respectively: for detail, see the supplementary file of Ref. 24.

As an important characteristic of the e-h pair in an OLED, it has been revealed from the bias dependence of the photoluminescence-detected magnetic resonance (PLDMR) signal that the pair can be dissociated by external electric field even under low biases.[37] In relation to the property of e-h pair, Figs. 2(a) and 2(b), respectively, show the bias-dependence of the EDMR and ELDMR signal intensities (ΔJ and ΔI_{EL}, respectively) measured simultaneously at the resonance center (3325G), together with the bias dependence of the current density (J) and the EL intensity (I_{EL}) also measured simultaneously. Non-linear bias-dependent relations are found between J and ΔJ and between I_{EL} and ΔI_{EL}. Indeed, the normalized intensities $\Delta J/J$ and $\Delta I_{EL}/I_{EL}$ shown in Figs. 2(c) and 2(d) exhibit complicated behaviors: both quantities rise from around 2.2–2.3 V and

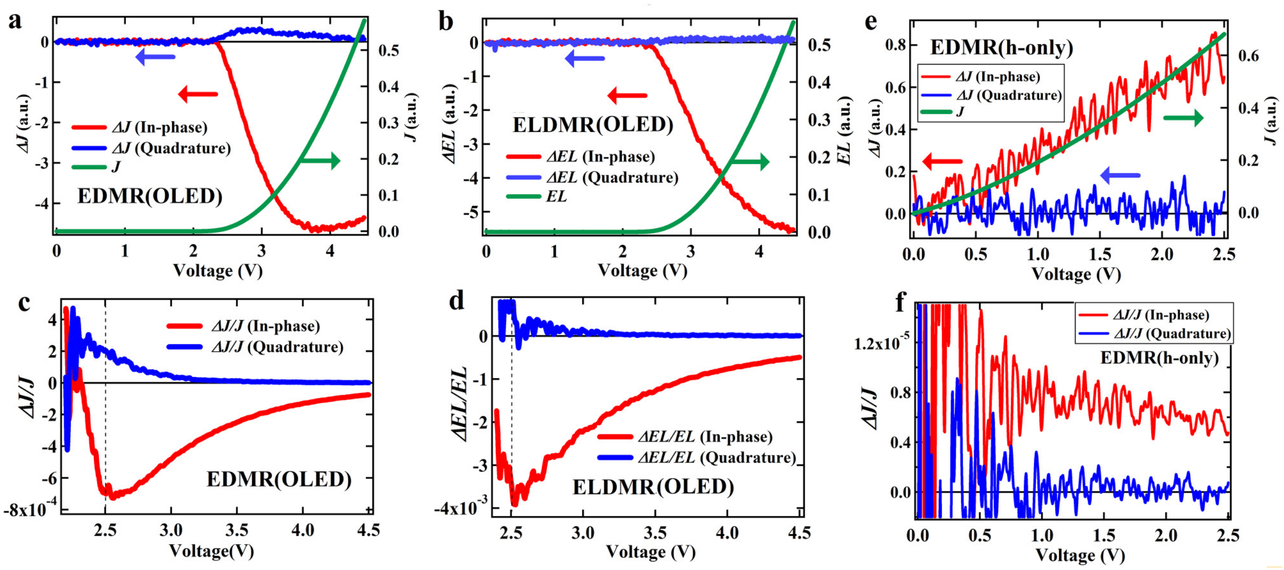

FIG. 2. (a) and (b) The bias-dependence of the EDMR (ΔJ) and ELDMR (ΔEL) intensities, respectively, measured simultaneously at the resonance center (3325G) by dual-phase lock-in techniques, together with the bias dependence of the current density (J: right axis) and the EL intensity (EL: right axis) also measured simultaneously. (c) and (d) Normalized EDMR ($\Delta J/J$) and ELDMR ($\Delta EL/EL$) intensities calculated from (a) and (b), respectively. (e) The bias-dependence of the EDMR signal and the current of the h-only device. (f) The bias-dependence of the normalized EDMR intensity in the h-only device.

then gradually decrease with a somewhat different bias-dependence. It was recently reported that the PLDMR signal of this OLED is reduced by increasing the bias in the positive bias region and almost disappears at around 4 V.[24] The characteristics of EDMR and ELDMR signals shown in Figs. 2(c) and 2(d) are thus explained as follows. Increasing the operating bias, the e-h pairs generated from the injected carriers increase, resulting in increasing the EDMR and ELDMR intensities. However, as the bias is further increased, the increase due to the field-induced dissociation to carriers and recombination to excitons of e-h pairs exceeds the increase of the e-h pair by bias.[24] The observed remarkable decreases in both the $\Delta J/J$ and $\Delta I_{EL}/I_{EL}$ are thus due to the dissociation and recombination of the pairs.

In addition to the in-phase EDMR and ELDMR signals, the quadrature component signal was also observed only from the EDMR signal [Fig. 1(a)]. Signals of such different phases suggest the presence of species with different origins from e-h pairs in the in-phase. The quadrature component indeed shows a rapider bias-dependent decrease compared with the in-phase component [Fig. 2(a)]. Although the lineshape and linewidth of the in-phase and quadrature signals are similar to each other, the difference in the bias-dependence is evidence that the quadrature component exists in the EDMR spectrum. Particularly, such quadrature signals were noticeably observed in the EDMR signal from the MEH-PPV light-emitting diode (LED) fabricated in an environment slightly exposed to air (see the supplementary material). This quadrature component is not directly related to EL because ELDMR signals did not give quadrature signals. This quadrature signal thus provides the evidence that non-emissive spin-dependent reactions are induced by the ESR transition. It has been suggested that electrons immobilized in trapping sites recombine with free holes and such trap-assisted recombination is non-emissive.[38] The non-emissive reactions thus probably occur between trapped electrons and free holes. Namely, spin-dependent reactions between trapped electrons and free holes are modulated by ESR, resulting in the change of carrier density. The rapid bias-dependent decrease in the quadrature signal thus indicates that the coupling strength of the trapped electrons and free holes is weaker than that of e-h pairs.

Figure 2(e) shows the bias-dependences of the EDMR signal (ΔJ) and the current density (J) in the hole-only device. Unlike the case in the OLED sample, the EDMR signal increases with approximately the same bias dependence as the current. Indeed, nearly constant bias-dependence is obtained from the normalized EDMR signal $\Delta J/J$ [Fig. 2(f)]. Thus, the EDMR signal from a unipolar device simply increases with the current, in contrast to the strongly bias-dependent properties of the EDMR signal of the OLED. As a possible explanation, the EDMR signal of the hole-only device could be due to a spin-dependent reaction of positive polaron carriers into a bipolaron: 2 polarons → bipolaron.[25] In this case, when the proportion of a polaron converted into a bipolaron is α, the density of the bipolaron is calculated to be $1/2\ \alpha\ n_p$ using the polaron density n_p. Assuming that the EDMR signal is obtained from a drift current, the change of the current density by the ESR transition is calculated as follows:

$$\Delta J = 1/2\, n_p\, \alpha (2e) \mu_{bp}\, F - n_p\, \alpha\, e \mu_p\, F = n_p\, \alpha\, e\, F(\mu_{bp} - \mu_p), \quad (1)$$

where e is the elementary charge, F is the electric field, and μ_{bp} and

μ_p are the mobilities of a bipolaron and a polaron, respectively. Since the polarons are the main carriers in this device, $\Delta J/J$ can be approximately calculated as follows:

$$\Delta J/J = \alpha(\mu_{bp} - \mu_p)/\mu_p. \tag{2}$$

Thus, when the bias-dependence of α and $(\mu_{bp} - \mu_p)/\mu_p$ is negligibly small, a bias-independent $\Delta J/J$ is obtained. Also, in this model, the obtained positive ΔJ in the hole-only device could be due to the relation of $\mu_{bp} > \mu_p$. For confirming the contribution of the bipolaron to EDMR, evidence that the bipolaron exists needs to be proved, for instance, from spectroscopic measurements.[39,40] Yet, the observed linear relation of $\Delta J/J$ in the hole-only device can be well-explained by the bipolaron model since α and $(\mu_{bp} - \mu_p)/\mu_p$ could be actually bias-independent. We note that, although the observed $\Delta J/J$ is small in the present hole-only device, this signal increases linearly with increasing the bias regardless of whether the signal is attributed to the bipolaron model. By contrast, the EDMR and ELDMR signals of OLED are remarkably reduced by increasing the bias. Therefore, under high bias, the EDMR signal of OLEDs could be dominated by this unipolar process depending on materials used for OLEDs.

B. Magnetic field effects

The characteristics of the MFEs, such as MC and MEL, were investigated for the OLED used for the ESR measurements. The normalized MC and MEL, defined by $\{J(H) - J(0)\}/J(0)$ and $\{I_{EL}(H) - I_{EL}(0)\}/I_{EL}(0)$ for the magnetic field strength H, were measured for the SY-OLED under the bias of 3.5 and 4 V [Figs. 3(a) and 3(b), respectively]. Both results exhibit a sharp increase over near 0G, a behavior typically observed in the MFEs of organic materials. The MC and MEL curves of organic materials have been shown to be well reproduced empirically by the non-Lorentzian function $\{H/(|H| + H_0)\}^2$ (H_0 is the quarter-saturation field)[2,41] typically in the magnetic field region much lower than a few thousand gauss, over which the contribution from Δg-dependent MC could be dominant (Δg is the difference of the g-factor between the hole and electron carriers).[42] Actually, the obtained MC and MEL curves were found to match the function using the following parameters: H_0 values for MC are 20G (3.5 V) and 18G (4 V) and H_0 values for MEL are 30G (3.5 V) and 35G (4 V), as shown in Figs. 3(a) and 3(b). We note here that, in the low magnetic field region typically below 50G, MC could include a low field effect indicated by the function of $H^2/(H^2 + H'^2_0)$ (H'_0 is a constant).[42,43] This effect is typically observed with the opposite sign to the contribution from the non-Lorentzian function. Yet, the MC in Fig. 3(a) showed only positive signals and the low field effect is probably small in the present OLED, being consistent with the previous report.[17]

MFEs of OLEDs have often been explained by the e-h pair (polaron pair) model,[3,4,6,8–15] the bipolaron model,[19,20] and the trion model.[17,18] Particularly, as evidence of the trion involved in MC, Cox *et al.* showed the result that the MC of organic devices decreases with increasing the bias.[17,18] In relation to the report, we measured the bias-dependence of MC and MEL for the SY-OLED at $H = 20$G using the ±20G-modulation. As shown in Figs. 3(c) and 3(d), both the MC and MEL signals increase to about 2.6 V and then turn to decrease. Although the magnitude of MC is lower than that of the previous report,[17] a similar bias-dependent MC-curve was obtained. In the reference, it was explained that a quartet trion (S = 3/2) to hinder carrier transport is formed between a TE and a trap carrier and the trion density becomes saturated in increasing a bias due to the finite number of trap sites, and the results of fit for the bias-dependent MC-curve based on the model seemed to be appropriate.[17] However, we note that the bias-dependence curves of MC and MEL resemble those of the normalized EDMR and ELDMR shown in Figs. 2(c) and 2(d), although their signal signs are opposite to each other: almost the same peak positions around at 2.6 V in all curves and the same zero crossover position at 2.3 V in EDMR and

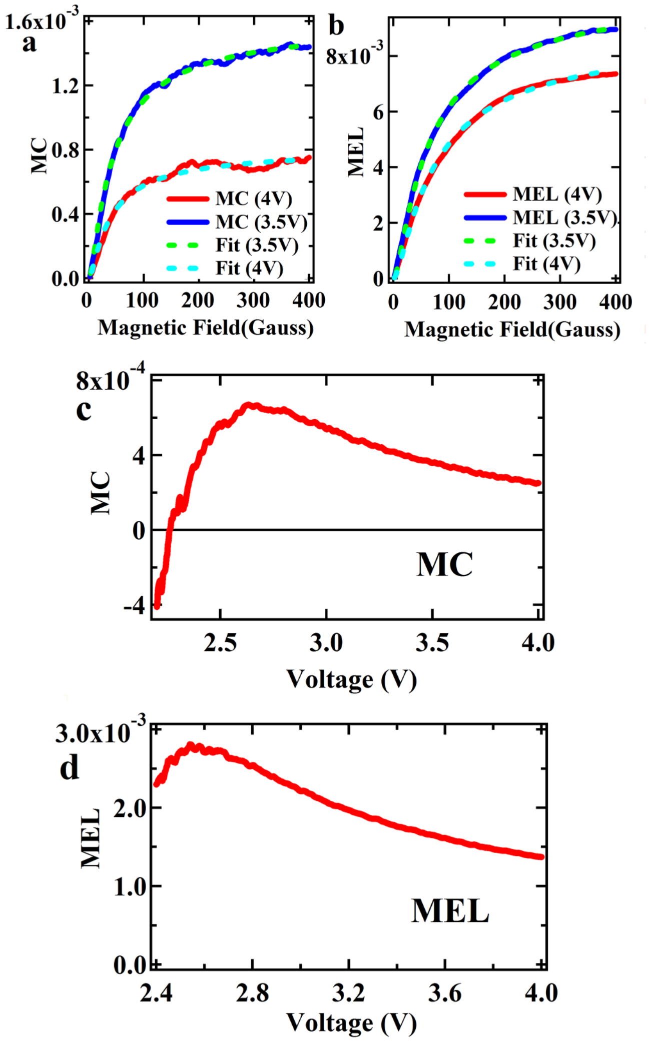

FIG. 3. (a) and (b) Magneto-conductance (MC) and magneto-EL intensity (MEL) defined by $\{J(H) - J(0)\}/J(0)$ and $\{I_{EL}(H) - I_{EL}(0)\}/I_{EL}(0)$ (I_{EL} the EL intensity) for the magnetic field strength H, respectively, measured for the SY-OLED under the bias of 3.5 and 4 V. The broken lines are results of fit for each curve using a non-Lorentzian function $\{H/(H + H_0)\}^2$ (H_0 is the quarter-saturation field). (c) and (d) Bias-dependence of MC and MEL, respectively.

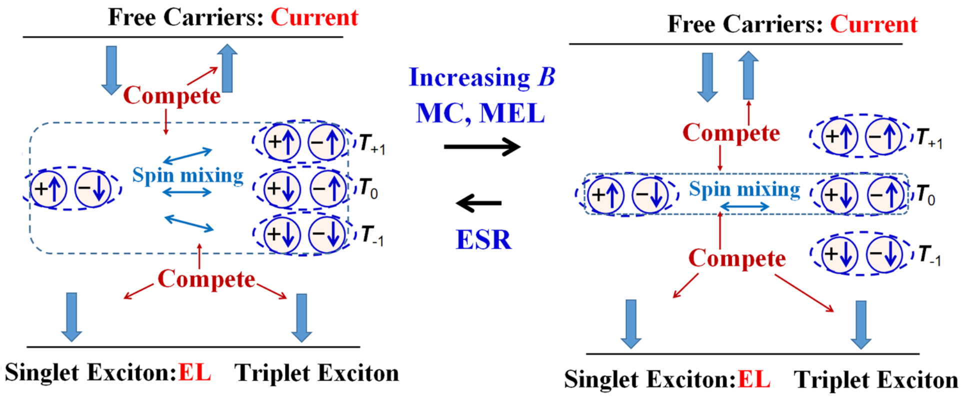

FIG. 4. Schematic of the model for MC and MEL under active OLED operation.

MC. The observed similarity demonstrates a strong correlation between MFEs and the ESR responses. Since the ESR responses were obtained via the e-h pairs, this similarity suggests that the MFEs occur through e-h pairs.

In the model based on e-h pairs as the origin of MFE, it is understood that spin mixing occurs between degenerated sub-levels of SP and TP at zero magnetic fields and it becomes limited to only mixing between S and T_0 under a finite magnetic field due to Zeeman splitting (Fig. 4), and the induced mixing-limitation leads to changing the densities of carriers and excitons depending on the magnetic field. The EDMR and ELDMR transitions occur so as to reduce the population difference between the sublevels made by the Zeeman splitting. Therefore, as pointed out recently,[15] the ESR responses and MFEs are expected to be given with opposite signs, corresponding well to the observed relation with opposite signs between the ESR responses and MFEs. This correspondence confirms that the MFEs mainly arise from e-h pairs and that ESR techniques provide information directly related to MFEs. Particularly, in the present result, the existence of a trion and a bipolaron was not necessary to assume for explaining the bias-dependent MC and MEL curves. Such a trion could be present at low temperature,[28,29] but we emphasize that the greatest contribution to the MFE-magnitude around room temperature is the conversion efficiency between SP and TP.

According to the e-h pair model described above, the mixing occurs only while e-h pairs are present in the OLED. Hence, under OLED operation, the mixing is expected to compete with the dissociation and recombination processes of the pairs. Namely, when the dissociation and recombination of e-h pairs become faster by increasing the bias, there would be no sufficient time for the spin mixing to occur, causing reductions of MFE. This is an explanation for the bias-dependent reductions of MC and MEL signals shown in Figs. 3(c) and 3(d). It has been recently proposed that the spin-mixing between S and T_0 is associated with Δg.[42] Thus, Δg could affect the competition relation between the spin-mixing and the dissociation /recombination processes.

The present model is partly similar to those proposed previously for the MFEs of OLEDs.[3,44] However, we emphasize that the present model was derived based on the observations of the similarity between the bias-dependent MFEs and ESR responses. There have been multiple models for explaining the MFEs of OLEDs,[39] and the main reason for it is that probing techniques to examine the origin of the MFEs have not been sufficiently developed. This research showed that the EDMR and ODMR properties are directly correlated with the MFEs when they are induced via the e-h pairs. Moreover, the EDMR process observed from the hole-only diode could also appear with increasing the bias depending on materials used for the OLED. The bias-dependent ESR experiments are thus effective and can be applied hereafter to discriminate coexistent MFE processes.

IV. CONCLUSIONS

The bias dependent behaviors of e-h pair under OLED operation were investigated from ESR and MFE measurements. The EDMR signal was not detected from the electron-only device and the hole-only device only gave a much smaller EDMR signal than the OLED device. Both the observed EDMR and ELDMR signals are primarily given by the spin-dependent reaction of e-h pairs. Non-linear bias-dependent relations were found between J and ΔJ and between I_{EL} and ΔI_{EL}, because the field-induced dissociation into carriers and recombination into emissive excitons of e-h pairs outweigh the increase of the pair induced by carrier injection from the electrodes. The bias-dependence curves of MC and MEL were demonstrated to be similar to those of EDMR and ELDMR, respectively. This similarity indicates that bias-dependent behaviors of e-h pairs determine MFEs of OLEDs at room temperature. The efficiency of MFEs largely depends on whether the spin-mixing between SP and TP can occur efficiently against competing recombination and dissociation processes of e-h pairs. This model was derived based on the observations of the similarity between the bias-dependent MFEs and ESR responses. The bias-dependent ESR experiments are effective to examine coexistent MFE processes.

SUPPLEMENTARY MATERIAL

See the supplementary material for a comparison of the line-shape of the EDMR spectrum in the hole-only devices at different biases and the EDMR features of MEH-PPV LED fabricated in air.

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