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Spin-injection into organic light-emitting diodes with a ferromagnetic cathode and observation of the luminescence properties

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Abstract

Electroluminescence (EL) properties of organic light-emitting diodes (LEDs) under an external magnetic field were investigated for observation of spin-polarized luminescence. When an external magnetic field was applied to organic LEDs with an Fe cathode, circularly polarized light was observed, while devices with an Al cathode did not show such behavior. In order to confirm whether the circularly polarized light was certainly due to spin-injection or not, photoluminescence (PL) property of Alq_3 (*tris*-(8-hydroxyquinolinato)-aluminum) film on Fe and that of Alq_3 film on Al under an external magnetic field were investigated. In the case of Alq_3 on Fe film, degree of circular polarization from EL was much higher than that from PL. This suggested that circular polarization on EL was mainly due to spin-injection.

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Keywords: Spin-injection; Photoluminescence; Electroluminescence; Magneto-optical effects; Organic light-emitting diode

Recently, a research field of spintronics has attracted much attention [1]. One of the hot topics is spin-injection into semiconductors and circularly polarized luminescence due to spin-injection has been observed from inorganic GaAs-based light-emitting diodes (LEDs)[2, 3]. On the other hand, study of spin-injection into organic LEDs has also been attracted [4-6]. However, no clear evidence for spin-injection has been observed in organic LEDs so far. In this study, luminescence properties of organic LEDs with a ferromagnetic cathode under an external magnetic field were investigated and the effect of spin-injection was discussed in terms of the spin-polarized luminescence effect.

Organic LEDs with a ferromagnetic cathode Fe as a spin injector were prepared by using conventional magnetron sputtering and vacuum vapor deposition. Optimized stacking structure on our experiments was glass-substrate / ITO (In_2O_3 + SnO_2 10 wt%, a transparent anode; thickness, 40 nm) / TPD (*N*,*N*'-Bis(3-methylphenyl)-*N*,*N*'-diphenylbenzidine, a hole transport layer; 45 nm) / Alq_3 (*tris*-(8-hydroxyquinolinato)-aluminum, an emissive layer; *d*) / Al-O(aluminum-oxide; 1.0 nm) / M (a cathode; 20 nm) / Al (for capping cathode; 120 nm). Emissive layer thickness *d* was changed from 30 nm to 65 nm. As a cathode M, Fe and a non-magnetic metal Al for comparison were used. A thin Al-O layer as a tunnel barrier for effective spin-injection was inserted at the interface between the Alq_3 layer and cathode [6, 7]. When an external magnetic field was applied perpendicular to the surface of organic LEDs with an Fe cathode, circularly polarized light was observed, while devices with an Al cathode did not show such behavior. Degree of circular polarization *P* on devices with an Fe cathode linearly increased with increasing the applied magnetic field and was 0.45 % at room temperature when the external magnetic field was 0.35 T. The detail of the Electroluminescence (EL) measurement was described in ref. [8].

In order to confirm whether the observed circularly polarized light was certainly due to spin-injection or not, photoluminescence (PL) properties for Alq_3 films on a metal under an external magnetic field were investigated. The sample structure for PL measurements was Si-substrate / M' (M' = Fe or Al, 20 nm) / Alq_3 (50 nm). M' corresponds to the above cathode material M in organic LEDs. Under a base pressure of $< 4.0 \times 10^{-4}$ Pa, Fe or Al was deposited by using an electron beam deposition at the respective deposition rates of

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0.03 nm/s or 0.10 nm/s, respectively. Subsequently without breaking vacuum, Alq_3 was evaporated from a crucible at 0.20 nm/s. During these depositions, the substrate was kept at -2 ºC.

Figure 1 shows the measurement system of PL properties under an external magnetic field [9]. Samples were excited at λ = 395 nm by a mode-locked Ti-sapphire laser equipped with a BaB_2O_4 crystal. P was defined as $(I_{\sigma+} - I_{\sigma-})/(I_{\sigma+} + I_{\sigma-}) \times 100$ (%); σ^+ and σ^- indicate right and left circularly polarized light, respectively; $I_{\sigma+}$ and $I_{\sigma-}$ indicate the intensity of σ^+ and that of σ^-, respectively. All measurements were carried out at room temperature.

Figure 2 shows the result of P measurement at 0.35 T for EL and that for PL; (a) M = M' = Al and (b) M = M' = Fe. Open circles indicate P on EL measured at around 520 nm corresponding to the wavelength of the peak position in Alq_3 luminescence [8]. P on PL measurements was calculated using PL spectra of both σ^+ and σ^-. In Fig. 2(a), P from PL is close to zero in all wavelength range measured and is almost the same as that from EL. These mean that influence of an external magnetic field is negligible up to 0.35 T and the Zeeman effect on Alq_3 molecules has not been observed, in both cases of PL and EL measurements. On the other hand, in Fig. 2(b), P from EL is much higher than that from PL. Thickness of the Alq_3 layer (50 nm) in samples for PL measurements was less than spin diffusion length (about 60 nm [8]). The fact that PL measurements showed no clear magneto-optical effects in this thickness indicated that the magneto-optical effects on a ferromagnetic cathode did not affect EL up to 0.35 T. Therefore, these strongly suggested that the origin of circular polarization observed at EL measurements was mainly due to spin-injection from the Fe cathode into the Alq_3 layer [10]. For the future prospect, the Zeeman effect on Alq_3 molecules in the circular polarization on EL could be evaluated by PL experiments under a higher magnetic field because the Zeeman effect would be observed sensitively when a higher magnetic field was applied.

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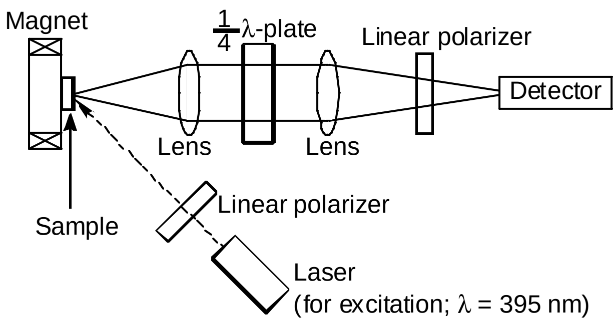

Fig. 1. Measurement system of PL properties under an external magnetic field.

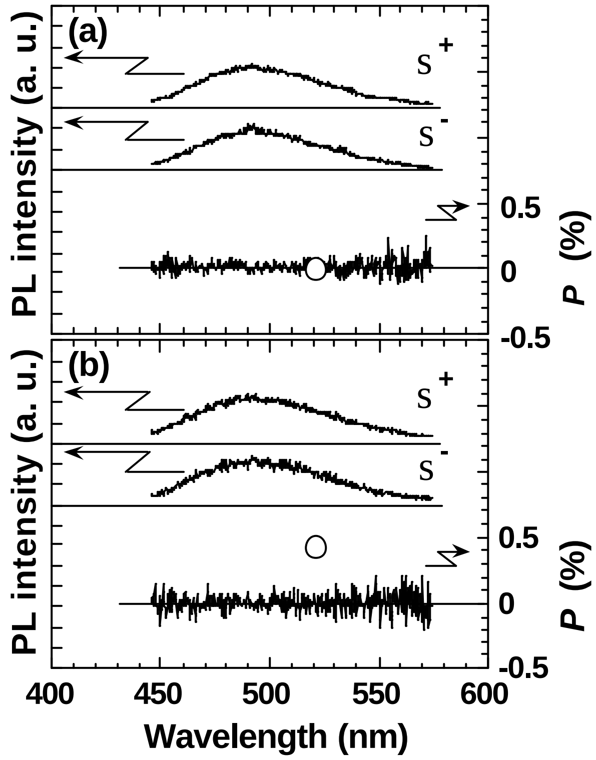

Fig. 2. The result of degree of circular polarization P measurement at 0.35 T for EL and that for PL; (a) M = M' = Al and (b) M = M' = Fe. P on EL measurements is indicated by an open circle in each figure.

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