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Emission Mechanism in PLED under DC Magnetic Field

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Introduction

Double infection of charge carrier into a conjugated polymer film is known to result in the light emission that originate from singlet excitons formed at the recombination of charges, electroluminescence (EL) [1,2]. Polymer light-emitting diodes (PLEDs) based on that phenomenon find many useful applications [3]. For choosing a proper polymer for the PLEDs it is important to understand, which part of the energy released at the recombination may be converted into the light. In a simpleminded scheme this part is 1/4 assuming that meeting of two opposite charged carriers in the singlet state is enough for producing a singlet exciton. However in organic materials with low mobility of charge carriers a new intermediate state is known to be formed at the recombination. This state is called usually as a polaron pair, and it represent two charges in a potential well formed due to their Coulomb attraction. The pairs can be in a singlet and triplet states, and what is important, their lifetime may be long enough for mutual intersystem crossing caused by a hyperfine interaction [4,5].

A scheme of energy levels of polaron pairs in magnetic field and main processes within the pairs are shown in Fig.1. One can see that because of intersystem crossing between singlet and triplet polaron pairs, initial ratio of charge meeting events in a singlet and triplet state (1 to 3) can result in a quite different ratio of the rates of formation of singlet and triplet excitons. Critical parameters here are rate constants of recombination of charges inside the pairs of different multiplicity, k_S and k_T. Depending on relative values of these rate constants, one can imagine the quantum yield of the EL, determined as a ratio of a number of recombination events to the amount of singlet exciton produced. $QY=k_S/(k_S+3k_T)$, to change from 1 to 0, being equal to 0.25 at $k_S=k_T$.

Thus, it becomes important and interesting to estimate relative values of these rate constants and lifetime of the pairs in order to get knowledge about perspectives of using any particular polymer in PLED devices.

Pairs are produced from their precursors in the same spin state as precursors had. During lifetime of the pair a spin evolution takes place, and different substates of the pair become mixed with each other due to intersystem crossing induced by a hyperfine interaction of electron spin with magnetic moment of nucleus (proton). For mixing, energy levels of polaron pairs must have the same position, so in zero magnetic field all four sublevels are mixed, and in an external magnetic field, which is more strong than hyperfine field, mixing of the singlet substate with one triplet substate m=0 is only possible. This case is shown in the Fig.1.

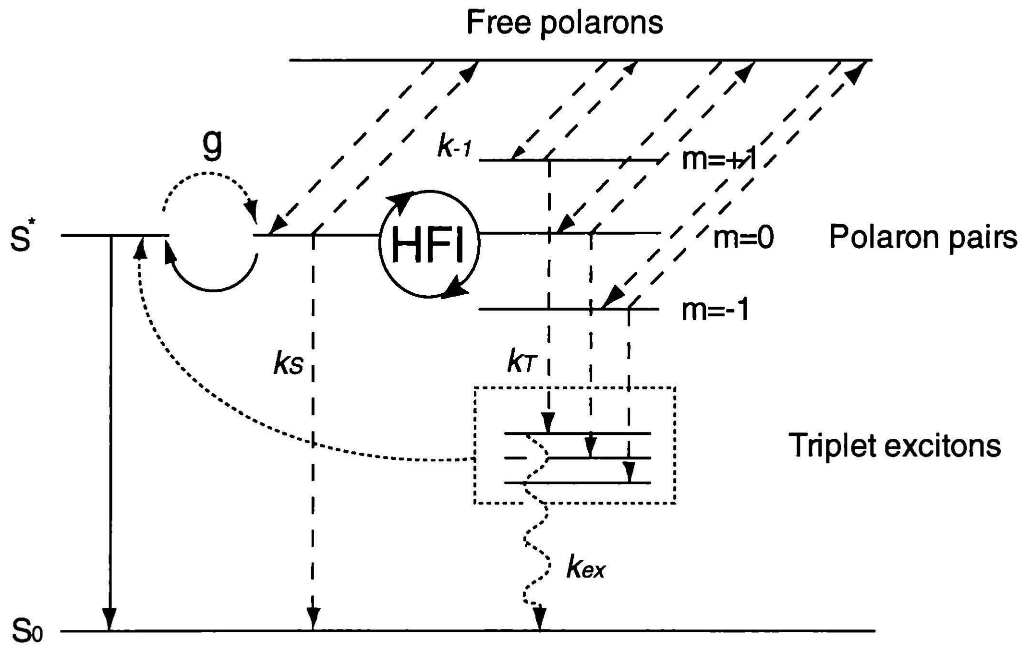

Fig.1 A scheme of main processes responsible for the EL at free carrier injection into a semiconducting polymer. The picture corresponds to the case of a DC magnetic field superimposed on the sample. Polaron pair's formation is shown at recombination of free charge carriers. HFI shows the mixing of magnetic substates of polaron pairs by a hyperfine interaction [6].

We are going to see changes in the intensity of EL appearing as a result of recombination of free charge carriers [7]. As it follows from the Fig.1, in zero magnetic field the quantum efficiency of the fluorescence is

$$QE_0 = L_0 = \frac{k_S}{k_S + 3k_T + 4k_{-1}}$$

In a high magnetic field

$$QE_B = L_B = \frac{k_S}{2(k_S + k_T + 2k_{-1})}$$

Relative change of the fluorescence intensity at the switching on the magnetic field

$$\frac{\Delta L}{L} = \frac{QE_B - QE_0}{QE_0} = \frac{k_T - k_S}{2(k_S + k_T + 2k_{-1})} \qquad (1)$$

Thus, the sign of relative changes of the recombination fluorescence intensity permits estimating the ratio k_S/k_T.

In the present work, we investigate changes of the EL intensity using the Magnetic Field Spin Effect techniques in order to estimate these parameters. As observables we are using here changes of the EL intensity, when working with double injection of carriers into a polymer film.

Experimental

Figure.2 shows the molecular structure of poly [2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylene-vinylene](MEH-PPV).

The structure of PLED used in this study is ITO/PEDOT: PSS/MEH-PPV/MgAg. A buffer layer of poly (3,4-ethylene dioxythiophene) (PEDOT) doped with poly (styrene sulfonate) (PSS) was overcoated onto the ITO electrode as the hole transport layer. The active luminescent layer, MEH-PPV was then spun on top of the PEDOT layer, and subsequently Mg-Ag alloy deposition onto the top of the film by means of evaporation. The effective area of this device was approximately 4mm^2. The emission light was collimated and collected with round lenses, and the integrated EL intensity, L, was measured by the photomultiplier. Measurements were done using DC magnetic field in the range from zero to 4500G under high vacuum (10^{-4}[Pa]). Temperature of the sample was 298K.

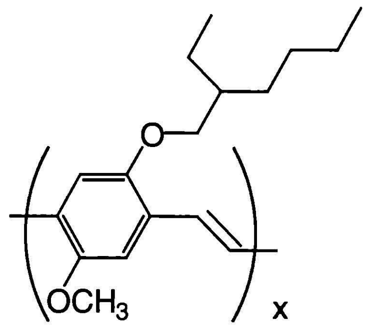

Fig.2 Molecular structure of MEH-PPV

Results and Discussion

Figure.3 shows the applied external DC magnetic field B and the corresponding responses of the EL intensity. These results show that the luminescence changes are positive and becomes bigger at higher magnetic field strength applied. The positive change may serve as evidence that $k_S > k_T$ according to formula (1). Because the triplet states of polaron pairs split by applying magnetic field, the rate of change of EL intensity increase with an increase in magnetic field strength.

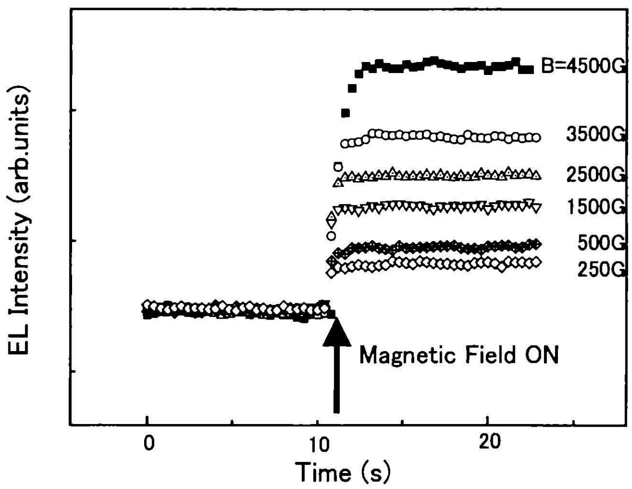

Fig.3 Changes of EL intensity of MEH-PPV applied DC magnetic field.

Conclusion

We demonstrated the magnetic field effect on the EL intensity of MEH-PPV of PLED. This can be explained that it is formed intermediate state called polaron pair at recombination in emission mechanism of PLED. By applying DC magnetic field because of the change of mixing of magnetic substates of polaron pairs, the ratio of the rates of formation of singlet and triplet excitons change.

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