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Numerical Study of Optimization of Layer Thickness in Bilayer Organic Light-Emitting Diodes*

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Abstract: A numerical model for bilayer organic light-emitting diodes (OLEDs) is developed under the basis of trapped charge limited conduction. The dependences of the current density on the layer thickness, trap properties and carrier mobility of the hole transport layer (HTL) and emission layer (EML) in bilayer OLEDs of the structure anode/HTL/EML/cathode are numerically investigated. It is found that, for given values of the total thickness of organic layers, reduced depth of trap, total density of trap, and carrier mobility of HTL as well as EML, there exists an optimal thickness ratio of HTL to EML, by which a maximal quantum efficiency can be achieved. Through optimization of the thickness ratio, an enhancement of current density and quantum efficiency of as much as two orders of magnitude can be obtained. The dependences of the optimal thickness ratio to the characteristic trap energy, total density of trap and carrier mobility are numerically analyzed.

Key words: organic light-emitting diodes; bilayer; optimization
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1 Introduction

Organic light-emitting diodes (OLEDs) have received considerable attention since the first demonstrations of practical electroluminescent device based on molecular and polymer materials[1~3]. The easy fabrication, the high efficiency, the compatibility with flexible and curved substrates and prospect for low cost make them appealing candidates for display applications. Much progress have been made in the manufacture of color- and white-emitting OLEDs[4~6], as well as in the researches on device lifetime and stability[7~9]. Bilayer OLEDs composed of two organic layers sandwiched between two electrodes, have generally better quantum efficiency than single layer OLEDs[10]. In bilayer OLEDs, one of the organic layers is responsible for light emission, called emission layer (EML), and the other is responsible for hole transport and electron blockade (for n-type luminescent materials as EML), called hole transport layer (HTL), or electron transport and hole blockade (for p-type luminescent materials as EML), called electron transport layer (ETL). Under application of proper bias on bilayer OLEDs, holes will be injected from the anode into the HTL and electrons will be injected from cathode into the EML. The injected holes and electrons migrate towards the HTL/EML interface, and light will be emitted through the recombination of holes and electrons in the EML near the interface. The thick-

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ness ratio of HTL to EML plays an essential role for the quantum efficiency of device. If HTL is too thin, bilayer OLEDs will behave like a single layer OLEDs, which has generally poor efficiency. But if HTL is very thick, the number of holes arriving at the HTL/EML interface might be much less than that of the electrons, which leads to also a poor device efficiency. Thus, for given transport and trap properties of HTL and EML, there must exist an optimal thickness ratio, by which the device reaches a maximal quantum efficiency. In this work, we report the results of numerical simulation for the optimization of the thickness ratio of HTL to EML in bilayer OLEDs on the basis of trapped charge limited (TCL) conduction theory[11~13].

2 Numerical model

Organic luminescent materials have generally poor electric conductivity with carrier mobility ranges from 10^{0} to $10^{-10}\,\mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{s})$, and most of them are single carrier conducting. That is, they have either much better conductivity for holes than for electrons, generally several orders of magnitude larger, or vise versa. So the contributions of electrons in HTL and holes in EML for current are negligible in many cases. The following discussions are based on bilayer OLEDs with the anode/HTL/EML/cathode structure as shown in Fig. 1, but the results are also valid for bilayer OLEDs with the structure of anode/EML/ETL/cathode. To simplify the problem, we assume: (1) In HTL there only exist free and trapped holes as well as hole current and in EML only free and trapped electrons as well as electron current, which means electron density and electron current in HTL as well as hole density and hole current in EML are neglected, and all the electrons and holes reaching the HTL/EML interface are recombined. Thus, the quantum efficiency of device is proportional to the device current density. (2) The mobility of holes and electrons are independent of electric field. (3) The energy barrier at HTL/EML interface is so small that its influence on carrier conduction can be neglected. The electric potential (Φ), field (F), total hole density (p), total electron density (n), and current density (j) at position x in HTL ($0\leqslant x\leqslant d_p$) or EML ($d_p < x\leqslant d$) are associated through following equations.

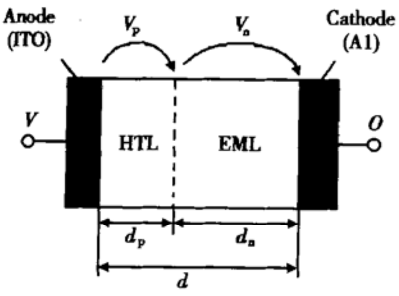

Fig. 1 Structure of a bilayer OLED

$$\frac{\mathrm{d}F}{\mathrm{d}x} = \begin{cases} \dfrac{qp}{\epsilon_0\epsilon_{rp}} & 0\leqslant x\leqslant d_p \\ -\dfrac{qn}{\epsilon_0\epsilon_{rn}} & d_p < x\leqslant d \end{cases} \tag{1}$$

$$p = \begin{cases} p_f + p_t & 0\leqslant x\leqslant d_p \\ 0 & d_p < x\leqslant d \end{cases} \tag{2}$$

$$n = \begin{cases} 0 & 0\leqslant x\leqslant d_p \\ n_f + n_t & d_p < x\leqslant d \end{cases} \tag{3}$$

$$j_p = q\mu_p p_f F - qD_p\frac{\mathrm{d}p_f}{\mathrm{d}x} \quad 0\leqslant x\leqslant d_p \tag{4}$$

$$j_n = -q\mu_n n_f F + qD_n\frac{\mathrm{d}n_f}{\mathrm{d}x} \quad d_p\leqslant x\leqslant d \tag{5}$$

$$j_p = j_n = j \tag{6}$$

$$F = -\frac{\mathrm{d}\Phi}{\mathrm{d}x} \tag{7}$$

where q is the elementary charge, ϵ_0, ϵ_{rp} and ϵ_{rn} are the permittivity of vacuum, the relative dielectric constant of HTL and EML respectively, p_f and p_t are the concentrations of free holes and trapped holes in HTL respectively, and n_f, n_t are that of electrons in EML. μ_p and D_p are the mobility and diffusion coefficient of holes in HTL, μ_n, D_n are that of the electrons in EML, d_p, d_n and d are the thickness of HTL, EML and the sum of them respectively. The zero point of coordinate x is located at the anode/HTL interface. Assuming that Einstein relation is still valid, then we have:

$$D_p = \frac{\mu_p k_B T}{q} \tag{8}$$

$$D_n = \frac{\mu_n k_B T}{q} \tag{9}$$

where k_B is Boltzmann constant and T is ambient temperature.

Experimental results show that the transport of charge carriers in organic materials can be described by trapped charge limited (TCL) conduction with an exponential trap distribution. According to TCL theory, the trap density in organic layer is distributed in energy as follows[14]:

$$h_p(E_{tp}) = \frac{H_{tp}}{k_B T l_p} e^{-\frac{E_{tp}}{k_B T l_p}} \tag{10}$$

$$h_n(E_{tn}) = \frac{H_{tn}}{k_B T l_n} e^{\frac{E_{tn}}{k_B T l_n}} \tag{11}$$

where E_{tp} is the energy of hole trap-states respective to the energy of the highest occupied molecular orbit (HOMO), and E_{tn} is the energy of electron trap states respective to the lowest unoccupied molecular orbital (LUMO); $h_p(E_{tp})$ is the density of hole trap states per unit energy in the vicinity of trap energy E_{tp}, and $h_n(E_{tn})$ is that of electron traps. l_p and l_n are the reduced trap depths, which are defined as the ratio of the characteristic trap energy of trap distribution to thermal energy $k_B T$, that is $l_p = E_{tcp}/k_B T$ and $l_n = E_{tcn}/k_B T$. H_{tp} and H_{tn} are the total density of hole trap states in HTL and electron trap states in EML, respectively. In organic semiconductors, the concentration of free charges ρ_f is generally much smaller than that of trapped charges ρ_t, and they are associated through following equations[15]:

$$\rho_f = \begin{cases} \theta \rho^{l_p} & 0 \leqslant x \leqslant d_p \\ \theta \rho^{l_n} & d_p < x \leqslant d \end{cases} \tag{12}$$

$$\rho = \begin{cases} qp & 0 \leqslant x \leqslant d_p \\ -qn & d_p < x \leqslant d \end{cases} \tag{13}$$

$$\rho_f = \begin{cases} qp_f & 0 \leqslant x \leqslant d_p \\ -qn_f & d_p < x \leqslant d \end{cases} \tag{14}$$

$$\theta = \begin{cases} qN_{HOMO} \left(\dfrac{\sin\left(\dfrac{\pi}{l_p}\right)}{qH_{tp}\left(\dfrac{\pi}{l_p}\right)} \right)^{l_p} & 0 \leqslant x \leqslant d_p \\ -qN_{LUMO} \left(\dfrac{\sin\left(\dfrac{\pi}{l_n}\right)}{qH_{tn}\left(\dfrac{\pi}{l_n}\right)} \right)^{l_n} & d_p < x \leqslant d \end{cases} \tag{15}$$

where N_{HOMO} is the density of states of HOMO of the HTL material, and N_{LUMO} is the density of states of LUMO of the EML material.

Eqs. (1) ~ (9) and (12) ~ (15) are predigested to a single variable differential equation, which is solved by using standard numerical methods.

3 Results and discussion

Figure 2 shows the dependences of current density on the thickness ratio d_p/d_n for different combinations of E_{tcp} and E_{tcn} with $E_{tcp}/E_{tcn} = 0.75$, $\mu_p = 10^{-4} \mathrm{cm}^2/(\mathrm{V \cdot s})$, $\mu_n = 10^{-5} \mathrm{cm}^2/(\mathrm{V \cdot s})$ and $H_{tp} = H_{tn} = 10^{19} \mathrm{cm}^{-3}$. There exist clearly optimal thickness ratios at $d_p/d_n = 6.1$ (for the curve with $E_{tcp} = 0.05$eV and $E_{tcn} = 0.067$eV) and $d_p/d_n = 5.8$ (for the curve with $E_{tcp} = 0.06$eV and $E_{tcn} = 0.08$eV), by which the current density and quantum efficiency reaches a maximum. The optimal thickness ratio is dependent on the carrier mobility, characteristic trap energy, and total trap density of HTL and EML. It can also be seen that, the current density for $d_p/d_n < 1$ is very small, this is because the current through an organic layer increases linearly with its mobility and decreases exponentially with its characteristic trap energy and thick-

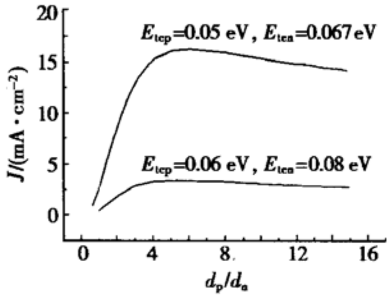

Fig. 2 Dependences of current density on the thickness ratio d_p/d_n for diferent combinations of E_{tcp} and E_{tcn} with $E_{tcp}/E_{tcn} = 0.75$, $\mu_p = 10^{-4} \mathrm{cm}^2/(\mathrm{V \cdot s})$, $\mu_n = 10^{-5} \mathrm{cm}^2/(\mathrm{V \cdot s})$ The values in parentheses are characteristic trap energy E_{tcp} and E_{tcn}, respectively. The rest parameters: $d = 200$nm, $H_{tp} = H_{tn} = 10^{19} \mathrm{cm}^{-3}$, $N_{HOMO} = N_{LUMO} = 10^{20} \mathrm{cm}^{-3}$, $\epsilon_{rp} = \epsilon_{rn}$, $T = 300$K, $V = 10$V.

ness. As $\mu_p \gg \mu_n$, $E_{tcp} < E_{tcn}$, and $d_p < d_n$, the main part of the operation voltage will drop off the electric less conductive EML in order to satisfy the current continuity equation, which results in the low current density of the device. Figure 3 shows the dependences of the optimal thickness ratio on the characteristic trap energy under constant E_{tcp}/E_{tcn} ratios with $\mu_p = 10^{-4}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $\mu_n = 10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$ and $H_{tp} = H_{tn} = 10^{19}\mathrm{cm}^{-3}$. The optimal thickness ratio $(d_p/d_n)_{opt}$ decreases as the characteristic trap energy E_{tcp} increases. This is the result of stronger influence of the characteristic trap energy on the current density than the mobility. When E_{tcp} and E_{tcn} are both very small, the optimal thickness ratio will be determined mainly by the mobility, and it tends to be near the value of μ_p/μ_n. When E_{tcp} and E_{tcn} are both great, the optimal thickness ratio will be determined mainly by the characteristic trap energies. While $E_{tcp}/E_{tcn} \ll \mu_p/\mu_n$ in Fig. 3, the optimal thickness ratios decrease as E_{tcp} increases, and tend to be constant in relevant to the E_{tcp}/E_{tcn} ratios.

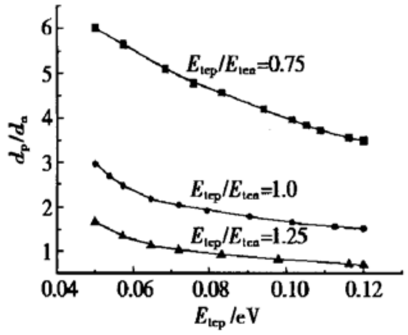

Fig. 3 Dependences of the optimal thickness ratio on the characteristic trap energy under constant E_{tcp}/E_{tcn} ratios with $\mu_p = 10^{-4}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $\mu_n = 10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $H_{tp} = H_{tn} = 10^{19}\mathrm{cm}^{-3}$ The rest parameters: $d = 200\mathrm{nm}$, $N_{HOMO} = N_{LUMO} = 10^{20}\mathrm{cm}^{-3}$, $\epsilon_{rp} = \epsilon_{rn} = 2$, $T = 300\mathrm{K}$, $V = 10\mathrm{V}$.

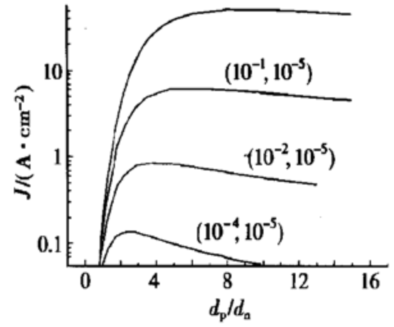

Fig. 4 Dependences of current density on the thickness ratio d_p/d_n for different mobility ratios with $E_{tcp} = 0.09\mathrm{eV}$, $E_{tcn} = 0.12\mathrm{eV}$ The rest parameters: $d = 200\mathrm{nm}$, $H_{tp} = H_{tn} = 10^{18}\mathrm{cm}^{-3}$, $N_{HOMO} = N_{LUMO} = 10^{20}\mathrm{cm}^{-3}$, $\epsilon_{rp} = \epsilon_{rn} = 2$, $T = 300\mathrm{K}$, $V = 10\mathrm{V}$.

The dependences of current density on the thickness ratio d_p/d_n for different mobility ratios with $E_{tcp} = 0.09\mathrm{eV}$, $E_{tcn} = 0.12\mathrm{eV}$, $H_{tp} = H_{tn} = 10^{18}\mathrm{cm}^{-3}$ and $d = 200\mathrm{nm}$ are shown in Fig. 4. The values in parentheses are μ_p and μ_n in $\mathrm{cm}^2/(\mathrm{V\cdot s})$, respectively. It is clearly to see that the maximum of current density shifts towards high d_p/d_n values, as the mobility ratio μ_p/μ_n increases for a constant μ_n value ($10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$). The cause is that when μ_p increases while μ_n and the total thickness d kept constant, the current density in HTL will be elevated to be greater than that in EML, thus the thickness of HTL must be increased, and that of EML must meanwhile be decreased in order to reduce the current density in HTL and increase the current density in EML to satisfy Eq. (6), which leads to the increase of $(d_p/d_n)_{opt}$. Moreover, it can be seen from the curve with $\mu_p = 10^{-1}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $\mu_n = 10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$ that the current density of the device with $d_p/d_n = (d_p/d_n)_{opt} = 9.2$ is about a hundred times greater than that of the device with $d_p/d_n = 1.0$, that is, through optimization of the thickness ratio, the quantum efficiency of bilayer OLEDs can be improved as much as two orders of magnitude. Figure 5 shows the dependences of the optimal thickness ratio $(d_p/d_n)_{opt}$ on the hole mobility under constant μ_p/μ_n ratio for different characteristic trap energies and trap densities with $d = 200\mathrm{nm}$. The optimal thickness ratio $(d_p/d_n)_{opt}$ kept constant as the hole mobility μ_p increases for given values of characteristic trap energy, total trap density, and mobility ratio μ_p/μ_n. We are not surprised, because the current density in organic layers is proportional to the carrier mobility, so the current density in HTL and ETL will increase with the equal rate for given μ_p/μ_n ratios when μ_p increases, and thus the optimal thickness ratio will be kept unchanged.

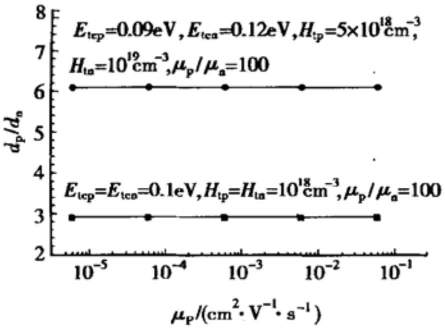

Fig. 5 Dependences of the optimal thickness ratio d_p/d_n on the hole mobility under constant for different characteristic trap energies, total trap densities, and μ_p/μ_n ratios The rest parameters: $d = 200$nm, $N_{HOMO} = N_{LUMO} = 10^{20}\mathrm{cm}^{-3}$. $\epsilon_{rp} = \epsilon_{rn} = 2$. $T = 300$K. $V = 10$V.

The dependences of current density on the thickness ratio d_p/d_n for different total trap densities, with $\mu_p = 10^{-4}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $\mu_n = 10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $E_{tcp} = 0.1$eV, $E_{tcn} = 0.1$eV, and $d = 200$nm are shown in Fig. 6. The optimal value of d_p/d_n increases from about 2.5 to about 15 when H_{tp} decreases from $5\times10^{18}\mathrm{cm}^{-3}$ to $5\times10^{17}\mathrm{cm}^{-3}$, if H_{tn} keeps constant ($10^{19}\mathrm{cm}^{-3}$). This is because that the current density decreases with the thickness and the total trap density. When the trap density H_{tp} increases, the thickness of HTL must be decreased and that of EML be increased in order to reach a maximal quantum efficiency. Figure 7 shows the dependences of the optimal thickness ratio $(d_p/d_n)_{opt}$ on the total hole trap density for different H_{tp}/H_{tn} ratios. The optimal thickness ratio increases with the total hole trap density H_{tp} for given H_{tp}/H_{tn} ratios, and the tendency of the increases is similar to each other. The reason is that, the current density decreases with both the trap density and the characteristic trap energy or the reduced trap depth, and the quantitative relation can be approximately expressed as $j_p \propto H_{tp}^{-l_p}$ and $j_n \propto H_{tn}^{-l_n}$, and the ratio of current density is then,

$$j_p/j_n \approx \alpha^{-l_n} H_{tp}^{l_n - l_p} \qquad (16)$$

where α is the ratio of trap density H_{tp}/H_{tn}. From Eq. (16) is to see that, the current density ratio j_p/j_n increases with H_{tp} for $l_p < l_n$ or $E_{tcp} < E_{tcn}$, and the thickness of HTL must be increased and that of EML be decreased in order to reduce j_p to satisfy Eq. (6), which leads to the increase of $(d_p/d_n)_{opt}$. It can be predicted that for given H_{tp}/H_{tn} ratios, if $E_{tcp} > E_{tcn}$, the optimal thickness ratio will decrease with H_{tp}.

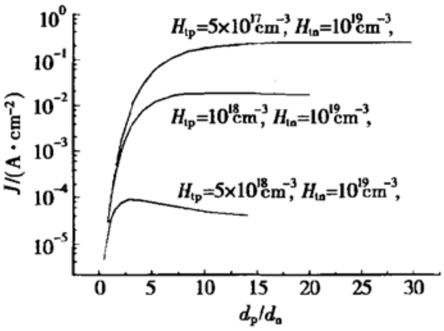

Fig. 6 Dependences of current density on the thickness ratio d_p/d_n for different total trap densities, with $\mu_p = 10^{-4}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $\mu_n = 10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $E_{tcp} = 0.1$eV, $E_{tcn} = 0.1$eV The rest parameters: $d = 200$nm, $N_{HOMO} = N_{LUMO} = 10^{20}\mathrm{cm}^{-3}$, $\epsilon_{rp} = \epsilon_{rn} = 2$, $T = 300$K, $V = 10$V.

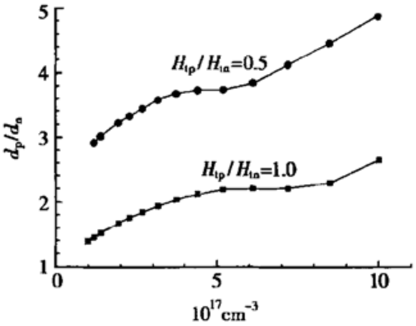

Fig. 7 Dependences of the optimal thickness ratio $(d_p/d_n)_{opt}$ on the total hole trap density for different H_{tp}/H_{tn} ratios, with $\mu_p = 10^{-4}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $\mu_n = 10^{-5}\mathrm{cm}^2/(\mathrm{V\cdot s})$, $E_{tcp} = 0.09$eV, $E_{tcn} = 0.12$eV The rest parameters: $d = 200$nm, $N_{HOMO} = N_{LUMO} = 10^{20}\mathrm{cm}^{-3}$, $\epsilon_{rp} = \epsilon_{rn} = 2$, $T = 300$K, $V = 10$V.

4 Conclusion

Numerical investigations show that for given values of characteristic trap energy, total density of trap, carrier mobility, and total thickness of organic layers, there exists an optimum value for the thickness ratio d_p/d_n, by which a maximal current density, and hence quantum efficiency and luminance can be achieved. Following results were obtained: (1) The quantum efficiency by an optimal thickness ratio can be two orders of magnitude greater than that by an un-optimized one. (2) For $H_{tp} \approx H_{tn}$ and $\mu_p > \mu_n$, which is popular for bilayer OLEDs, the optimal thickness ratio is greater than 1 by $E_{tcp} < E_{tcn}$, and in the vicinity of 1 by $E_{tcp} > E_{tcn}$. If both the characteristic trap energies E_{tcp} and E_{tcn} are very small, then the optimal thickness ratio will be mainly determined by the mobility ratio μ_p/μ_n. If both E_{tcp} and E_{tcn} are great, then the optimal thickness ratio will be mainly determined by E_{tcp}/E_{tcn} ratio. (3) For the given values of E_{tcp}, E_{tcn}, H_{tp}, and H_{tn}, the optimal thickness ratio depends only on μ_p/μ_n ratio; (4) For the given value of H_{tp}/H_{tn} ratio, the optimal thickness ratio will increase by $E_{tcp} < E_{tcn}$ and decrease by $E_{tcp} > E_{tcn}$ with the increase of H_{tp}.

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双层有机电致发光器件有机层厚度优化的数值研究*

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摘要: 在陷阱电荷限制电流传导理论的基础上, 提出了双层有机电致发光器件的数值模型, 研究了结构为"阳极/空穴输运层(HTL)/发光层(EML)/阴极"的器件中电流密度和量子效率随有机层的特征陷阱能量、陷阱密度和载流子迁移率的依赖关系. 研究发现, 对于给定的HTL和EML的特征陷阱能量、陷阱密度和载流子迁移率, 存在一个最优的HTL和EML之间的厚度比率, 在此最优厚度比下, 器件的电流密度和量子效率达到最大. 通过有机层厚度的优化, 器件的电流密度和量子效率可提高多达两个数量级. 另外, 还研究了最优厚度比随有机层特征陷阱能量、总陷阱密度和载流子迁移率之间的定量关系.

关键词: 有机发光器件; 双层; 优化
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