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Modeling of transient electroluminescence overshoot in bilayer organic light-emitting diodes using rate equations

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ABSTRACT

When a voltage pulse is applied under forward biased condition to a spin-coated bilayer organic light-emitting diode (OLED), then initially the electroluminescence (EL) intensity appearing after a delay time, increases with time and later on it attains a saturation value. At the end of the voltage pulse, the EL intensity decreases with time, attains a minimum intensity and then it again increases with time, attains a peak value and later on it decreases with time. For the OLEDs, in which the lifetime of trapped carriers is less than the decay time of the EL occurring prior to the onset of overshoot, the EL overshoot begins just after the end of voltage pulse. The overshoot in spin-coated bilayer OLEDs is caused by the presence of an interfacial layer of finite thickness between hole and electron transporting layers in which both transport molecules coexist, whereby the interfacial energy barrier impedes both hole and electron passage. When a voltage pulse is applied to a bilayer OLED, positive and negative space charges are established at the opposite faces of the interfacial layer. Subsequently, the charge recombination occurs with the incoming flux of injected carriers of opposite polarity. When the voltage is turned off, the interfacial charges recombine under the action of their mutual electric field. Thus, after switching off the external voltage the electrons stored in the interface next to the anode cell compartment experience an electric field directed from cathode to anode, and therefore, the electrons move towards the cathode, that is, towards the positive space charge, whereby electron–hole recombination gives rise to luminescence. The EL prior to onset of overshoot is caused by the movement of electrons in the electron transporting states, however, the EL in the overshoot region is caused by the movement of detrapped electrons. On the basis of the rate equations for the detrapping and recombination of charge carriers accumulated at the interface expressions are derived for the transient EL intensity I, time t_m and intensity I_m corresponding to the peak of EL overshoot, total EL intensity I_t and decay of the intensity of EL overshoot. In fact, the decay prior to the onset of EL overshoot is the decay of number of electrons moving in the electron transporting states. The ratio I_m/I_s decreases with increasing value of the applied pulse voltage because I_m increases linearly with the amplitude of applied voltage pulse and I_s increases nonlinearly and rapidly with the increasing amplitude of applied voltage pulse. The lifetime τ_t of electrons at the interface decreases with increasing temperature whereby the dependence of τ_t on temperature follows Arrhenius plot. This fact indicates that the detrapping involves thermally-assisted tunneling of electrons. Using the EL overshoot in bilayer OLEDs, the lifetime of the charge carriers at the interface, recombination time of charge carriers, decay time of the EL prior to onset of overshoot, and the time delay between the voltage pulse and onset time of the EL overshoot can be determined. The intense EL overshoot of nanosecond or shorter time duration may be useful in digital communication, and moreover, the EL overshoot gives important information about the processes involving injection, transport and recombination of charge carriers. The criteria for appearance of EL overshoot in bilayer OLEDs are explored. A good agreement is found between the theoretical and experimental results.

1. Introduction

The phenomenon of electroluminescence (EL) in organic solids is based on several processes, and therefore, the conversion of electrical energy into light energy requires several steps such as

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injection, transport, capture, and radiative recombination of positive and negative charge carriers inside an organic layer with suitable energy gap, to yield visible light output. Thus, by optimizing these individual steps the efficiency of electroluminescent devices can be optimized. In fact, a very successful approach to optimize these individual steps separately is the concept of organic multilayer light-emitting diodes (OLEDs), in which there are heterostructures between different organic materials. The simplest OLED based on this concept consists of a heterostructure between a hole-conducting material, usually a triphenylamine derivative and an electron conducting aluminum chelate complex Alq_3, where the light emission takes place in the Alq_3 layer close to the organic–organic interface. As the relevant mechanism of EL in organic solids is injection-type luminescence, the optimization of the efficiency of OLEDs requires high and equal densities of positive and negative charge carriers at the internal interface. In the last two decades, the OLEDs have attracted worldwide attention as a candidate for next generation of flat-panel displays and environmentally friendly solid state lighting devices [1–10]. Furthermore, the discovery of bright organic electroluminescent diodes has stimulated intense research in order to understand the physics of injection, transport and recombination processes in organic semiconductors.

In fact, the time response of OLEDs is determined by the superposition of the elementary processes such as injection, migration, recombination of charge carriers and radiative and nonradiative decay of excitons produced during the recombination of charge carriers. Thus, the understanding of the time response of OLEDs involving superposition of elementary processes is complex. One of the ways to understand this fact involves the study of the kinetics of transient behavior of pulsed EL of OLEDs [10–36]. When a rectangular voltage pulse is applied to a spin-coated bilayer organic light emitting diodes, then the electroluminescence appears after a delay time and its intensity initially increases with time and then it attains a saturation value. At the end of the voltage pulse, initially the EL intensity decreases to some extent, however, after a particular time the EL intensity rises again, attains a peak value and then decays slowly. The appearance of delayed EL spike is known as EL overshoot effect. The peak intensity of the EL overshoot increases with increasing duration of the preceding voltage pulse. It varies approximately linearly with the applied voltage while the ratio of the peak intensity of overshoot to the saturated EL intensity decreases with increasing voltage. The decay of the spike signal is slightly super-exponential, the 1/e decay time exceeds the RC-time of the circuit and depends only marginally on the cell current/cell voltage during the on phase [15].

The study of overshoot effect in OLEDs is important because the intense overshoots of nanosecond or very short duration may be useful in digital communication. Furthermore, the overshoot gives information related to the processes involved in injection, transport and recombination of charge carriers in OLEDs, and it is useful in determining several parameters of the OLEDs. Nikitenko et al. [15] have reported that the charge carriers accumulated at the interface between hole transporting layer and electron transporting layer is responsible for the overshoot effect produced during the turning off the applied voltage pulse in spin-coated bilayer OLEDs. In the present paper, considering the model proposed by Nikitenko et al. [15] and using the rate equations for the generation and recombination of electrons in electron transporting states (i.e. LUMO energies) expressions are derived for the temporal characteristics of the overshoot in spin-coated bilayer OLEDs, which are able to explain satisfactorily the rise, decay, time t_m and intensity I_m corresponding to the peak of EL overshoot, and time delay between the end of voltage pulse and onset of overshoot. The EL decay prior to the onset of EL overshoot is shown to be the decay of the number of electrons moving in the electron transporting states. It is to be noted that such analytical expressions for different parameters of EL overshoot have not been derived till now. Furthermore, the criteria for the occurrence of overshoot in bilayer OLEDs are also explored. The theoretical results are found to be in good agreement with the experimental results. The physical concepts related to the EL overshoot obtained from the expressions based on rate equations are much simple and clear. It is shown that the lifetime of trapped charge carriers at the interface, recombination time of charge carriers, decay time of the EL prior to onset of overshoot, and delay time between end of the voltage pulse and onset time of EL overshoot can be determined from the temporal characteristics of the overshoot in bilayer OLEDs. It is to be noted that, in single layer OLEDs the interface between the cathode and organic material or the interface between the anode and organic material is responsible for the overshoot effect; however, in the bilayer OLEDs, the interfacial layer between hole transporting layer and electron transporting layer is responsible for the overshoot effect. Hence, the kinetics of EL overshoot in bilayer OLEDs is quite different from that of the single layer OLEDs.

2. Basic model for the EL overshoot

In bilayer OLEDs, the overshoot occurs in spin-coated layers, but it does not occur in vapor deposited samples [15]. This fact shows the existence of an interfacial layer of finite thickness between hole and electron transporting layers, in which both transport molecules coexist, whereby the interfacial energy barrier impedes both hole and electron passage. It has been found that when an additional thin layer of an insulating material is introduced between the hole transporting layer and electron transporting layer, then also the formation of interfacial barrier gives rise to the appearance of EL overshoot [21]. When a voltage pulse is applied to an OLED, positive and negative space charges are established at the opposite faces of the interfacial layer. Subsequently, the charge recombination occurs with the incoming flux of injected carriers of opposite polarity. When the voltage is turned off, the interfacial charges recombine under the action of their mutual electric field. Thus, after switching off the external voltage electrons stored in the interface next to the anode cell compartment experience an electric field directed from anode to cathode, and therefore, the electrons move towards cathode, that is, towards positive space charge, which consequently, acts as an electron sink. As such, the electrons are not able to migrate towards the anode and get discharged non-radiatively, and consequently, the probability of electron–hole recombination increases. Thus, the appearance of overshoot effect in OLEDs can be understood qualitatively. It has been reported that the effect gradually vanishes when a positive bias is applied to the OLED while maintaining the amplitude of the voltage pulse fixed. Moreover, it saturates as the bias becomes negative because the probability for an electrons to recombine becomes unity irrespective of how large the total electric field inside the interfacial layer becomes after turning off the driving voltage.

Nikitenko et al. [15] have developed a model for the role of the internal contact region, which results from the interpenetration of the two organic–organic layers deposited by spin coating, Fig. 1 sketches schematically this model. The interfacial layer has been characterized by a thickness l_i, energy barriers H_e' for electrons and H_h' for holes with $H_h' > H_e'$. Hence, holes will be accumulated more than electrons in the interface. σ_h and σ_e denote the hole and electron surface charge densities, respectively and E_i denotes the electric field in the interface region. The electric fields E_h and E_e in the adjacent transporting carriers regions (thickness l_h for