



# Demonstration of determination of electron and hole drift-mobilities in organic thin films by means of impedance spectroscopy measurements



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## ABSTRACT

The electron and hole drift-mobilities of tris(8-hydroxy-quinolinato) aluminium (AlQ<sub>3</sub>) thin films have been determined using impedance spectroscopy (IS) measurements. The theoretical basis of drift-mobility measurement with IS rests on a theory for single-injection space-charge limited current. The electron drift-mobilities in AlQ<sub>3</sub> which measured from the frequency dependence of both capacitance and conductance had the same electric-field dependence and were identical to those measured by a time-of-flight (TOF) transient photocurrent technique. To estimate the hole drift-mobility in AlQ<sub>3</sub>, a 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]-biphenyl thin film was inserted as an injection layer to reduce the hole injection barrier between an injection electrode and the AlQ<sub>3</sub> thin film. The hole drift-mobilities of AlQ<sub>3</sub> thin films measured with IS were identical to those measured by the TOF method. These results are essential for clarifying the mechanism of the carrier transport of organic light emitting diodes (OLEDs) with the aim of improving OLED performance.

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## 1. Introduction

Organic light-emitting diodes (OLEDs) have attracted a widespread interest for their potential use as flat-panel displays and surface-emitting lights. However, further improvements in the efficiency and lifetime of OLEDs are necessary before they can be widely used in practical applications. Charge-carrier transport is one of the primary physical processes in OLEDs and understanding it is of fundamental importance for the improvement of OLED performance. Impedance spectroscopy (IS) is a powerful tool for measuring the key physical quantities governing the charge-carrier transport, including charge-carrier drift-mobility [1–12], localized-state distributions [13,14], and carrier recombination [15]. IS is particularly suitable for determining the charge-carrier drift-mobility of the organic thin films in OLEDs because it can be used with organic semiconducting thin films with a thickness of 50–100 nm, which is comparable to the organic layers in OLEDs. The  $-\Delta B$  method [1–12] and  $\omega\Delta G$  method [7–9] have previously been proposed for determining charge-carrier drift-mobility, where  $-\Delta B$  is negative differential susceptance,  $\omega$  is angular frequency, and  $\Delta G$  is differential conductance. When determining the charge-carrier drift-

mobility of organic thin films by IS measurements, the injection barrier between the injection electrode and the organic thin film needs to be less than certain threshold values [7,8], which for the  $-\Delta B$  and the  $\omega\Delta G$  methods were calculated to be 0.2 eV and 0.4 eV, respectively [7,8].

Tris(8-hydroxy-quinolinato) aluminium (AlQ<sub>3</sub>) is one of the most promising materials for the electron transport layer and/or light emissive layer (EML). Both electrons and holes are injected and transported in the AlQ<sub>3</sub> EML. The determination of both the electron and the hole drift-mobilities in AlQ<sub>3</sub> is therefore of fundamental importance. The first study on electron-transport in AlQ<sub>3</sub> thin film by IS measurements was reported by Berleb et al. [2]. However, in that study electron drift-mobility was not estimated. The determination of the hole drift-mobility in AlQ<sub>3</sub> was difficult because of the high hole injection barrier between an injection electrode and the AlQ<sub>3</sub> thin film. Inserting an injection layer between the injection electrode and the AlQ<sub>3</sub> thin film made it possible to determine the hole drift-mobility in AlQ<sub>3</sub> and only the hole drift-mobility using the  $-\Delta B$  method was reported [10].

In this paper, we report on the determination of both electron and hole drift-mobilities in AlQ<sub>3</sub>. The electron drift-mobilities were obtained from IS measurements of AlQ<sub>3</sub> electron-only devices (AlQ<sub>3</sub> EODs) using both the  $-\Delta B$  and the  $\omega\Delta G$  methods. Results were identical to those measured by a time-of-flight (TOF) transient photocurrent technique. The hole drift-mobility was obtained from IS measurements of AlQ<sub>3</sub> hole-only devices with a thin film of 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]-biphenyl (NPB) as the hole injection layer (AlQ<sub>3</sub> HODs) using the  $\omega\Delta G$  method and was identical to those estimated by the  $-\Delta B$  method and measured by the TOF method. The observation provides a key information understanding on the charge-carrier transport mechanism in organic thin films.

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## 2. Experimental details

We used vacuum deposition to fabricate organic and metal thin films for the devices (pressure  $\sim 5 \times 10^{-5}$  Pa). The glass substrates were cleaned ultrasonically in deionized water and by UV-ozone treatment before the vacuum deposition. The Alq<sub>3</sub> EODs, sample 1, were fabricated as follows. A 110-nm-thick Al electrode, 210-nm-thick Alq<sub>3</sub> thin film, 0.5-nm-thick LiF thin film, and 150-nm-thick Al electrode were deposited on the glass substrate in series while maintaining the vacuum condition. Negative bias was applied to the LiF/Al electrode. With the Alq<sub>3</sub> HODs, sample 2, a 5-nm-thick NPB thin film, 180-nm-thick Alq<sub>3</sub> thin film, and 150-nm-thick Au electrode were deposited on the glass substrates with patterned indium-tin oxide (ITO) electrodes in series, similar to the Alq<sub>3</sub> EODs. Positive bias was applied to the ITO electrode. Sample 3, which consists of ITO/5-nm-thick NPB/110-nm-thick Alq<sub>3</sub>/Au is also one of the Alq<sub>3</sub> HODs. Table 1 shows the structures of the Alq<sub>3</sub> EODs and HODs in detail. The samples were carried in a glove box (with less than 1 ppm of water) without exposure to the laboratory atmosphere and sealed by a cap-glass with epoxy resin adhesives. The measurement area was  $1 \times 1$  mm<sup>2</sup>.

Fig. 1(a) shows the energy diagram of the Alq<sub>3</sub> EOD (Al/Alq<sub>3</sub>/LiF/Al). We used the LiF/Al thin film as an electron injection electrode. An LiF ultrathin film is typically used as the injection layer between Al and Alq<sub>3</sub> layers to improve the electron injection efficiency in small molecule OLEDs. The efficiency of the electron injection directly from the Al electrode to the Alq<sub>3</sub> thin film is low because the work function of Al and the energy level of the lowest unoccupied molecular orbital (LUMO) of the Alq<sub>3</sub> thin film are 4.1 eV and 3.0 eV, respectively, and the energy barrier is high. The electron injection can be improved by the band bending caused by charge distribution driven by the working function difference between the LiF and Alq<sub>3</sub> layers [16] and the increase of electron concentration by chemical reaction of the LiF, Alq<sub>3</sub>, and Al layers [17]. The LiF/Al electrode is therefore suitable for use as the electron injection electrode in the IS measurements. Fig. 1(b) shows the energy diagram of the Alq<sub>3</sub> HOD (ITO/NPB/Alq<sub>3</sub>/Au). The ITO thin film is a hole injection electrode. The hole injection barrier between the ITO electrode and the energy level of the highest occupied molecular orbital (HOMO) of Alq<sub>3</sub> is high because the work function of ITO  $W_f$  is 5.1 eV and the energy level of the HOMO of Alq<sub>3</sub> is 5.7 eV. We therefore inserted the 5-nm-thick NPB thin film to reduce the hole injection barrier. The hole injection barrier  $\Phi_B'$  between the  $W_f$  of ITO and the HOMO energy level of the NPB thin film and the hole injection barrier  $\Phi_B''$  between the HOMO energy levels of the NPB thin film and of the Alq<sub>3</sub> thin film are estimated to be 0.3 eV and 0.3 eV, respectively as the HOMO energy level of NPB is 5.4 eV. Injection barriers,  $\Phi_B'$ , and  $\Phi_B''$ , are comparable to the threshold value of the injection barrier of 0.4 eV for the  $\omega\Delta G$  method [7–9].

The IS was measured with a Solartron 1260 impedance analyzer connected to a 1296 dielectric interface. The frequency range was from 10 mHz to 1 MHz, and the measurements were made at room temperature.

## 3. Results and discussion

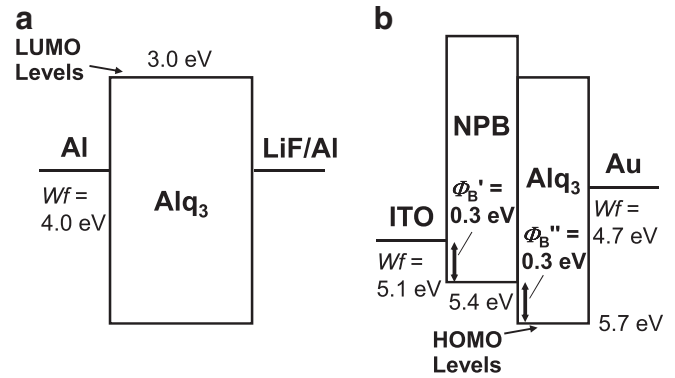
### 3.1. Determination of electron drift-mobility in Alq<sub>3</sub>

Fig. 2(a) shows the frequency ( $f$ ) dependence of capacitance ( $C$ ) in sample 1. The  $C$ - $f$  curves at  $V = 4$ –8 V exhibited a minimum value

**Table 1**

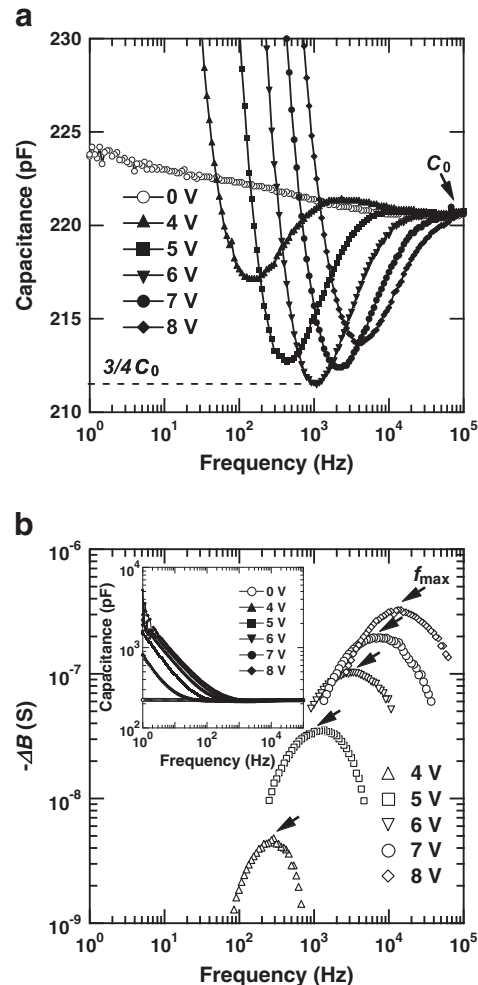
Device structures of Alq<sub>3</sub> electron-only and Alq<sub>3</sub> hole-only devices.

	Anode	Charge injection layer	Organic layer	Cathode
Sample 1	Al	–	210-nm Alq <sub>3</sub>	LiF/Al
Sample 2	ITO	5-nm NPB	180-nm Alq <sub>3</sub>	Au
Sample 3	ITO	5-nm NPB	110-nm Alq <sub>3</sub>	Au



**Fig. 1.** Energy diagrams of (a) the Alq<sub>3</sub> electron-only devices (EODs), which consist of Al/Alq<sub>3</sub>/LiF/Al and (b) the Alq<sub>3</sub> hole-only devices (HODs), which consist of ITO/NPB/Alq<sub>3</sub>/Au.  $W_f$  is a work function,  $\Phi_B'$  is the injection barrier between the  $W_f$  of ITO and the highest molecular orbital (HOMO) energy level of NPB, and  $\Phi_B''$  is the injection barrier between the HOMO energy level of NPB and the HOMO energy level of Alq<sub>3</sub>.

caused by the competition between the transit-time effect and the carrier trapping in the distributed localized state [9,10]. A transit-frequency proportional to the reciprocal to the transit-time is in the vicinity of the



**Fig. 2.** (a) Frequency dependence of capacitance ( $C$ ) in sample 1 at DC bias voltages of 0, 4, 5, 6, 7, and 8 V; sample 1 consists of Al/Alq<sub>3</sub>(210 nm)/LiF(0.5 nm)/Al. (b) Frequency dependence of  $-\Delta B = -\omega \cdot (C - C_0)$  in sample 1 at DC bias voltages of 4, 5, 6, 7, and 8 V. Inset shows the  $C$ -frequency curves for all measured frequency range at DC bias voltages of 0, 4, 5, 6, 7, and 8 V.  $-\Delta B$  is negative differential susceptance,  $\omega$  is angular frequency, and  $C_0$  is a geometrical capacitance, 220.4 pF.  $f_{max}$  is the frequency of the maximum value at the  $-\Delta B$ -frequency curves.

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