Available online at www.sciencedirect.com
SCIENCE DIRECT
Thin Solid Films 489 (2005) 235 – 244
thin solid films
www.elsevier.com/locate/tsf

Dependence of the emission from tris (8-hydroxyquinoline) aluminum based microcavity on device thickness and the emission layer position

C.H. Cheung[a], A.B. Djurišić[a,*], C.Y. Kwong[b], J. Chan[c], A.D. Rakić[c], H.L. Tam[d], K.W. Cheah[d], Z.T. Liu[a], W.K. Chan[e], P.C. Chui[b]

[a]Department of Physics, University of Hong Kong, Pokfulam Road, Hong Kong
[b]Department of Electrical and Electronic Engineering, University of Hong Kong, Pokfulam Road, Hong Kong
[c]School of Information Technology and Electrical Engineering, The University of Queensland, Brisbane Qld4072, Australia
[d]Department of Physics, Hong Kong Baptist University, Kowloon Tong, Hong Kong
[e]Department of Chemistry, University of Hong Kong, Pokfulam Road, Hong Kong

Received 24 January 2005; received in revised form 22 April 2005; accepted 22 April 2005
Available online 3 June 2005

Abstract

In this work, we present a systematic study of the emission from bilayer organic microcavity light emitting diodes with two metal mirrors. The devices consisting of two organic layers, N,N′-di(naphthalene-1-yl)-N,N′-diphenylbenzidine as the hole transport layer and tris (8-hydroxyquinoline) aluminum as the emitting layer, and two metal mirrors were fabricated and characterized by transmittance, reflectance, photoluminescence, and electroluminescence measurements. The effects of layer thickness, interface position, and the choice of anode (bottom mirror) were investigated. The transmittance and reflectance spectra were modeled using a transfer matrix model, and the optical functions for all the materials used were determined by spectroscopic ellipsometry. The dependence of the photoluminescence and electroluminescence spectra on the device thickness and interface position is discussed.

PACS: 85.60.Jb; 85.60.Bt
Keywords: Organic light emitting diode; Electroluminescence; Evaporation

1. Introduction

Organic materials have been attracting lots of interest for application in organic light emitting diodes (OLEDs) due to their potential high brightness and possible inexpensive fabrication on large area and/or flexible substrates. A large number of different materials for OLED application have been developed in recent years. In addition to the research on novel materials, novel device structures have been investigated as well. Among those, microcavity OLEDs have been attracting considerable attention due to their potential to achieve spectral narrowing, brightness enhancement, or multipeak emission from the same emitting layer [1–5]. Microcavities can operate in two regimes: weak coupling [1–5] and strong coupling regime [6–15]. The strong coupling regime has been of more interest for fundamental physics studies, but the materials used in strongly coupled cavities are of limited interest for practical applications. This is mainly due to the perceived need for high Q factor cavities and the narrow linewidth of the emitting material [11], although large Rabi splitting was demonstrated recently for a low Q factor cavity with two metal mirrors [7]. Narrow exciton linewidth also does not appear to be a mandatory requirement for the existence of a non-zero Rabi splitting. The Rabi splitting is given by the following equation [8]:

$$E_{\mathrm{R}} = \left[\frac{\hbar\alpha_e c}{e}\gamma_{\mathrm{e}}\Gamma - \frac{1}{4}(\gamma_{\mathrm{c}} - \gamma_{\mathrm{e}})^2\right]^{1/2} \quad (1)$$

where γ_{c} and γ_{e} are the spectral widths of the cavity and exciton modes respectively, α_e is the peak absorption

* Corresponding author.
E-mail address: dalek@hkusua.hku.hk (A.B. Djurišić).

0040-6090/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2005.04.106

coefficient, and Γ is the cavity confinement factor that describes the overlap between the cavity mode and the absorption. It is obvious that there is a non-zero splitting when the spectral widths of the cavity and exciton modes are equal. However, the splitting can only be observed if the splitting energy is sufficiently large compared to the broadening. Therefore, the condition for the observation of the strong coupling can also be stated as [16]:

$$\Omega_0 > \gamma_c + \gamma_e, \tag{2}$$

where Ω_0 is the on-resonance splitting (which is not necessarily the same for reflectance, transmittance, absorptance, and photoluminescence [17,18]). Thus, the main condition for observation of the strong coupling is that the splitting is large enough. Very large Rabi splitting (0.43 eV) was reported for the polysilane based microcavities [6], in spite of the fact that the linewidth of polysilane is about 190 meV [6,8]. Strong coupling was also recently demonstrated in microcavities with a polycrystalline film of 3,4,7,8 naphtalenetetracarboxylic dianhydride [15]. Therefore, broad exciton resonance does not necessarily mean that the material will not exhibit polariton related features in the spectra of microcavity devices. However, there are several other phenomena which can cause spectral features with appearance similar to strong coupling. For example, the splitting between transverse electric (TE) and transverse magnetic (TM) modes can occasionally cause peak splitting which is rather similar to the Rabi splitting [19]. Although previous works on the angular dependence of the emission from the tris (8-hydroxyquinoline) aluminum (Alq) based microcavities showed blue shift without peak splitting [1–3], observation of TE and TM mode splitting would be expected for high mirror reflectivity.

In this work, we fabricated and characterized organic microcavity structures with two metal mirrors and N,N′-di(naphthalene-1-yl)-N,N′-diphenylbenzidine (NPB) as a hole transport layer and Alq as emitting layer. Silver (70 nm) was used as a cathode and top mirror, while either thin (25 nm) Cu or thick (80 nm) Ag films were considered as bottom mirrors. The fabricated devices were characterized by transmittance (T), reflectance (R), photoluminescence (PL), and electroluminescence (EL) measurements. The transmittance and reflectance were modeled using a transfer matrix model, and the index of refraction of all the materials used was determined by spectroscopic ellipsometry. For the devices with thin Cu mirror, unusual behavior involving multiple peaks in electroluminescence whose intensity ratio changes with the viewing angle is observed. Possible reasons for this behavior are discussed.

2. Experimental and calculation details

In this section, we describe sample preparation, determination of the index of refraction of materials using spectroscopic ellipsometry, device characterization using R, T, PL, and EL measurements, and finally calculation of the R, T, and the emission spectra.

2.1. Sample preparation

NPB and Alq (from H. W. Sands) were purified by sublimation before device fabrication. Both devices and thin films for spectroscopic ellipsometry measurements were fabricated by evaporation in high vacuum ($\sim 10^{-4}$ Pa). The layer thickness during deposition was detected by a quartz thickness monitor, and it was double checked after deposition using step-profiler and ellipsometry. Thin films for the spectroscopic ellipsometry measurements were fabricated either on Si or glass substrates. It was reported that the extinction coefficient for thin films with low refractive index on a transparent substrate (where two materials differ little in refractive index) cannot be accurately determined by ellipsometry and relates strongly to the noise in experimental spectra and the film thickness [20]. Also, the overestimation of the extinction coefficient of transparent glasses cannot be eliminated by surface roughness correction [21]. In our previous study of thin Alq films on a glass substrate, possible overestimation of the extinction coefficient in sub-band gap range was obtained [22]. To eliminate this problem silicon substrates were used in this study to characterize Alq and NPB films. Since it was shown that the Alq deposited on unheated substrate is amorphous [23], no significant influence of the substrate on the film structure and optical properties is expected. For copper and silver, films on glass substrates were fabricated. The back surface of the glass substrate was roughened in order to eliminate errors due to reflections from this surface. All the substrates were cleaned by acetone, ethanol, and deionized water.

Microcavity OLED devices had the following structure: quartz substrate/bottom mirror/NPB/Alq/Ag (70 nm). The devices with different bottom mirrors (Cu and Ag) were fabricated at the same time in order to eliminate possible effects of the thickness variation of the organic layers. In addition to microcavity OLEDs containing NPB and Alq layers, microcavity OLEDs comprising only a single Alq layer with the same total thickness were also fabricated in order to exclude possible effects of NPB on the observed phenomena. The microcavity OLEDs with NPB/Alq and Alq layers exhibited similar behavior, except that the blue emission from NPB upon 325 nm excitation was absent in microcavity OLEDs containing only Alq.

2.2. Material characterization

Thin films of Alq, NPB, silver, and copper were measured by an ellipsometer (J. A. Woollam V-VASE variable angle spectroscopic ellipsometer). Photoluminescence (excited at 325 nm by a HeCd laser) and absorption spectra (using Hewlett Packard 8453 UV-Vis spectrometer) measurements for the NPB and Alq films were also