Organic Light Emitting Diodes: materials, device structures and light extraction

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Abstract: Organic Light Emitting Devices (OLEDs) are presented with particular emphasis on materials, device structures and strategies to improve light extraction. For the fabrication of efficient OLEDs, two types of electroluminescent materials are used (small molecules and polymers) with efficiencies and lifetimes which are now acceptable for small size or low content information displays. Two main approaches are described to enhance external efficiency, one is based on direct modification of the species emission and the other on modification of emitted guided modes propagation within the device. The features of the different approaches are evidenced through examples of devices reported in the literature.

Keywords: OLED; organic light-emitting diode; fluorescence and phosphorescence multilayer structure; light extraction; light out-coupling; microcavity; nano-patterned materials.

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1 Introduction

π-conjugated organic semiconductor materials are considered as one of the most promising system for the future development of less expensive and flexible electronic devices such as Organic Light Emitting Diodes (OLEDs), Organic Field Effect Transistors (OFETs) and Organic Photovoltaic solar cells (OPVs). The growing interest is mainly due to the quasi-infinite possibilities offered by the organic chemistry to tailor the organic materials via molecular structural modifications. Predictive structure-property relationships can be established and the optimisation of characteristic properties to suit a desired function is possible via chemical engineering at the molecular scale. OLEDs are one of the most promising next generation flat-panel displays. The key advantages of OLEDs for display applications are their self-emitting property, high contrast ratio, very fast response time, high luminous efficiency, full colour capability, wide viewing angle, low power consumption, low weight, potential large area colour displays and flexibility. Since the mid-1980s, organic materials have been seen as a key component of a promising display technology able to challenge Liquid-Crystal Displays (LCDs), the actual predominant display technology (Geffroy et al., 2006).

Electroluminescence is the emission of light from materials with injected current and it has been observed from single crystals of anthracene in the 1960s (Helfrich and Schneider, 1965). Besides the high quantum efficiency already obtained with such organic crystals, no application has emerged at that time due to the very high working voltage needed and the technical difficulties required making reliable electrical contacts. Nevertheless, these studies have led to a good understanding of the basic physical processes involved in organic electroluminescence i.e., charges injection, charges transportation, excitons formation and light emission. A first breakthrough was achieved in 1987 by Tang and van Slyke (1987) from Kodak when they reported efficient and low voltage OLEDs from p-n heterostructure device using thin films of vapour deposited organic materials. They used a hole-transporting molecular film of aromatic diamine and an emitting layer of electron-transporting metal chelate tris-hydroxyquinoline aluminium (Alq_3) between the transparent Indium Tin Oxide (ITO) anode and a low work-function alloy of aluminium and magnesium as the cathode. This two-layer structure with highly purified materials and properly chosen electrodes permitted OLED to operate at voltages below 10 V for the first time. A second breakthrough appeared in 1990 by the discovery at the University of Cambridge of electroluminescence from polymers (Burroughes et al., 1990). The Cambridge group used a poly P-Phenylene Vinylene (PPV) conjugated polymer in a single-layer device structure and obtained for the fist time efficient green-yellow light from a polymer. These two important demonstrations pave the way for intense research and development both in academia as well as in industry and currently various OLED-based commercial products like mobile phones and mp3 players are on the market. The OLEDs technology is actually almost limited to small displays but it has a good chance to expand in the near future even for larger and higher resolution displays. Moreover, the tremendous progress in device luminous efficiency (Ikai et al., 2001) and the emerging viability as a commercial display open new markets for White OLEDs (WOLEDs), like backlight for liquid crystal displays or lighting sources. WOLEDs can be considered as potential next generation of large area solid state lighting sources to replace traditional incandescent white light sources, thanks to the potential of energy saving, the high efficiencies and the possibility to fabricate thin and flexible devices (D'Andrade and Forrest, 2004).

The basic structure of an OLED, depicted in Figure 1, consists of a thin film of the light emitting material sandwiched between two electrodes. The anode is transparent and is usually made of Indium Tin Oxide (ITO) while the cathode is reflective and is made of metal. The thickness of the organic layer is very thin (typically between 100 nm and 150 nm thick) to avoid the need of high working voltage because the organic material is almost an insulator. The phenomenon of electroluminescence (Kalinowski, 1997) involves

- charge injection
- charge transport
- carriers recombination
- formation of excited states and finally
- radiative decay as illustrated in Figure 2.

Figure 1 Basic OLED structure with bottom emission

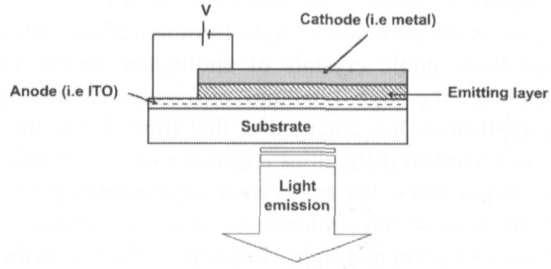
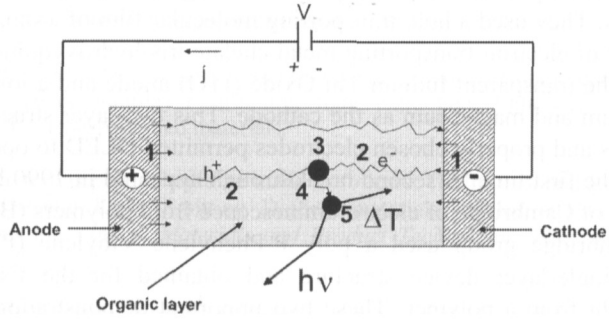


Figure 2 Basic phenomena of electroluminescence in OLEDs

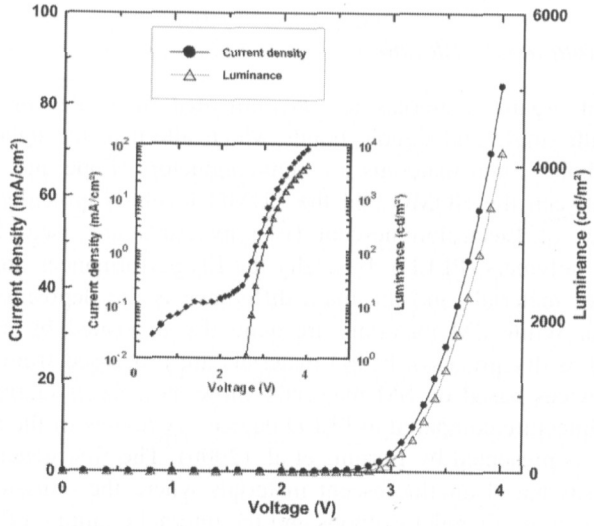


1: Charge carrier injection; 2: Charge carrier transport (holes and electrons);
3: Charge recombination (exciton formation); 4: Exciton diffusion;
5: Exciton radiative decay and photon emission; ΔT is non radiative decay.

When a voltage is applied between the electrodes, charges are injected in the organic material, holes from the anode and electrons from the cathode. The charges are transported inside the material, generally by hopping processes and then recombine to form excitons. The location of the recombination zone in the diode is function of carrier mobility within the organic layer. After diffusion, the exciton undergoes relaxation and a photon is emitted. The colour of the photon is a function of the energy difference between the Highest Occupied Molecular Orbital (HOMO) and the Lowest Unoccupied Molecular

Orbital (LUMO) levels of the electroluminescent molecule. The wavelength of the light emission can then be tuned by the extent of the conjugation in the molecule or the polymer. A typical I-V-L characteristic of OLEDs is shown in Figure 3. Above the threshold voltage, the current increases exponentially and light is emitted. The intensity of the emitted light is generally a function of the current density.

Figure 3 Typical current-voltage-luminance characteristic curves of OLEDs. The insert (in semi-logarithmic scales) emphasises the current and luminance voltage thresholds



The internal quantum efficiency (η_{int}) of an OLED device is defined as:

$$\eta_{int} = \frac{N_{ph}}{N_e} = \frac{P_{int}/hv}{I/e} \tag{1}$$

where N_{ph} is the number of photons generated in the active area per second, N_e is the number of electrons injected into the device per second, P_{int} is the internal optical power and I is the current injected into the device. Ideally, all the photons emitted by the active region should escape from the OLED device, but in a real device, some of the light is trapped or lost inside the layered structure or the substrate. The useful quantum efficiency or external quantum efficiency (η_{ext}) is then given by:

$$\eta_{ext} = \eta_{int}.\eta_{out} \tag{2}$$

where η_{out} is the fraction of photons that can escape the device. It has been shown (Greenham et al., 1994) that in planar structure like OLEDs, $\eta_{out} \sim 1/2n^2$ with n the refractive index of the organic material. Assuming that for most organics used in OLEDs, $n = 1.7$, η_{out} is typically around 17% only. However, Kim et al. (2000) have shown that if optical interferences with the cathode reflector are taken into account, $\eta_{out} \sim A/n^2$, where A is around 0.75 and 1.2 for isotropic and in-plane dipoles, respectively.

In view of the dimensions involved, the ability to control emission or propagation in the undesired modes by means of a modification of the photonic environment at the optical wavelength scale has thus attracted much attention. In this respect OLEDs has benefited of previous investigations on their inorganic counterpart, which suffer

from losses of the same origin but in a larger proportion due to the higher refractive indices involved (in the case of GaAs about 3% only of the light is extracted). After some general considerations on OLED structures and materials performances, some of the main strategies employed to enhance the emission of light are reviewed in the third part of this paper.

2 Materials and device structures

2.1 Materials and device lifetime

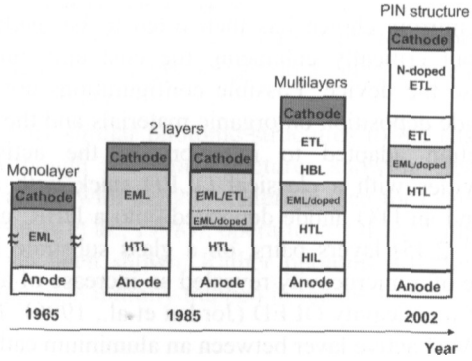
Most luminescent organic materials are π-conjugated molecules or macromolecules, i.e., materials with single and double bonds which alternate throughout the molecule or polymer backbone. Such materials are semiconductors (Pope and Swenberg, 1999) since the gap between the HOMO and the LUMO levels is typically in the 1.5–3 eV range. Two types of Electroluminescent (EL) materials are used: Small Molecules (SM-OLED) and polymers (PLED). Basically the EL performances are very similar for the two classes of materials and the main difference is the deposition process of the organic thin film. While SM materials are generally deposited by evaporation under vacuum (referred as dry process), PLED materials are processed from solutions (called wet process). Devices based on SM materials allow more layer engineering and more sophisticated architecture compared to PLED devices. A review of the main luminescent polymer families is presented by Bernius et al. (2000). The first generation of efficient OLED devices was based on fluorescent materials where the emission of light is the result of recombination of singlet excitons and the internal quantum efficiency is limited to 25% according to spin statistical considerations. For SM-OLED, the efficiency is generally improved by doping the emitting layer (doping rate about 1–2 wt%) with highly luminescent organic dyes. This solution has been widely used to tune the colour and to improve the device lifetime (Tang et al., 1989). Typical efficiency values collected in the literature for SM-OLED are reported by Geffroy et al. (2006). The second generation uses phosphorescent materials where all exactions can be converted into emissive triplet states through efficient intersystem crossing. Baldo et al. (1998) reported for the first time the use of green phosphorescent dye to increase the device efficiency. High efficient green and red electro-phosphorescent emitters have been demonstrated with internal quantum efficiencies approaching 100% (O'Brien et al., 1999; Adachi et al., 2001). The presence of heavy atoms like iridium or platinum increases spin-orbit coupling, favours intersystem crossing and allows radiative triplet transitions (Valeur, 2002). Device stability is an important issue for emissive technology like OLEDs and particularly differential aging of the primary three colours. It is generally assumed that for display applications, except probably for portable electronics, a lifetime of over 20,000 h for a reasonable brightness level of at least 100 cd/m^2 is necessary. Today, lifetimes reported for green and red fluorescent materials (SM materials and polymers) have a half-life of more than several tens of thousands hours at an initial luminance of 100 cd/m^2. Nevertheless, the blue lifetime remains weaker and is around 20,000 h (100 cd/m^2). For phosphorescent materials (PHOLED) lifetimes are also in excess of 20,000 h for high level of luminance as shown by Chwang et al. (2005) for red and green emitting materials. The development of efficient blue phosphorescent materials has been

more difficult, but now sky blue PHOLED materials are available with good efficiency as well as lifetime exceeding 10,000 h.

2.2 Device structures

The evolution of OLEDs architectures as a function of time is shown in Figure 4. For SM-OLED an increase of the complexity of the device structures has been reported. The first EL device made by Helfrich and Schneider (1965) used a simple monolayer structure and since the breakthrough of Kodak group, more layers are added such as hole injecting layer, hole transporting layer, hole blocking layer, emitting layer and electron transporting layer. It has been shown that the electroluminescence efficiency of OLEDs can be increased by carrier or exciton confinement within a multilayer device (So and Forrest, 1991; Ohmori et al., 1993). The confinement of charge carriers can increase capture of carriers and the confinement of excitons can improve energy transfer from the host to the guest. In comparison, the PLED technology uses a more simple structure consisting of two or three polymer layers only. In the earlier 2000s a new approach for low power display technology has been demonstrated by Leo group at University of Dresden by combining electrically doped transport layers and phosphorescent doped emitting layer in a diode structure called p-i-n junction (He et al., 2004). Low operating voltages close to the thermodynamic limit (no interface barrier and virtually no ohmic losses over the transport layers) have been achieved with such devices. Like for inorganic semiconductors, organic Multiple-Quantum-Well (MQW) structures have also been fabricated consisting of alternating layers of doped and undoped organic material (Huang et al., 2000; Qiu et al., 2002; Yang et al., 2002). This technique has been used to finely tune the colour properties of white OLEDs (Choukri et al., 2006). Very recently, new structure called Tandem OLEDs using multiple EL units connected in series by using a doped p-n junction have been fabricated (Liao et al., 2004). The luminous efficiency and the driving voltage are found to scale almost linearly with the number of connected units and values as high as 32 cd/A or 132 cd/A are obtained for devices using a fluorescent or a phosphorescent emitter, respectively. By combining two luminance-enhancement techniques like microcavity and tandem OLEDs, a record efficiency of 200 cd/A has been obtained by Cho et al. (2006) using phosphorescent cavity two-unit device.

Figure 4 Evolution of OLED device structures



HTL: Hole Injection Layer; HTL: Hole Transporting Layer; EML: Emitting Layer; HBL: Hole Blocking Layer; ETL: Electron Transporting Layer.

In most reported device structures, the light is seen from the substrate side (bottom emission) but it is also possible to built devices where the light escapes from the last deposited layer (top emission) as shown by Bulovic et al., (1997) and Gu et al. (1999). The top emitting OLEDs structure allows for an easier integration with black-plane electronics such as silicon active matrix as well as a higher aperture ratio for light to escape from the device (Pribat and Plais, 2001).

3 Nanostructured OLED devices

The strategies considered for the improvement of light extraction can be classified within two main approaches: the enhancement of external efficiency

- through direct modification of the species emission or
- through modification of guided modes propagation.

3.1 Driving the emitter

3.1.1 Planar microcavities

Due to the planar structure of OLEDs and the thickness involved, microcavity structures can be easily implemented in such devices. The optical mode density control through interferences between the direct and the reflected emitted fields of an emitter placed within the cavity can be used to redirect the 4π sr. radiated spontaneous emission towards the cavity modes to favour light extraction. The energy extracted into radiating modes can be controlled by the cavity thickness and losses, i.e., mirrors reflectivity and layers absorption (Benisty et al., 1998).

Maximum extraction efficiencies of 38 and 47% for metal and hybrid planar microcavity OLEDs respectively have been calculated in the case of monochromatic emission from an emitter oriented in the plane of the device (Wasey and Barnes, 2000). The difference accounts for the mirrors nature. While presenting a wide wavelength range of reflectivity, metal mirrors experience losses due to light absorption, exciton quenching at the interface and coupling to the so-called Surface Plasmon (SP) modes. Such effects are avoided with the use of Distributed Bragg Reflectors (DBR) which are constituted of a stack of dielectric layers pairs with a high refractive index contrast. In return, DBR mirrors have a limited wavelength/angle range of high reflectivity.

A trade-off in the mirrors choice has then often to be made in order to optimise the reflectivities without critically enhancing the cost and number of technological steps required to realise the device. Possible configurations are also restricted by the difficulty to control oxide deposition on organic materials and the need of materials with extraction work function adapted to injection on the active organic materials. Thus, hybrid microcavities with a classical OLED stack with a top metallic (silver, Aluminium) cathode and an ITO anode deposited onto a DBR, generally made of SiO_2 ($n = 1.5$) and TiO_2 ($n = 2.45$) layers pairs, on a glass substrate have often been used. Experimentally, the use of a microcavity revealed an increase of almost twice the energy extracted in the case of a noncavity OLED (Jordan et al., 1996). The device studied was based on a dye-doped Alq3 active layer between an aluminium cathode and an ITO anode deposited onto a DBR on glass structure. Extraction efficiencies improved by a factor of

around 1.6 without eye detectable angular colour modification have also been demonstrated with adapted cavity thickness. Another strategy employed consists of optimising the emission efficiency at a given wavelength. This can be done using narrow emitting species (Tsutsui et al., 1994) or with micro-cavities showing a normal resonance in the lower wavelength side of the emission peak close to the steep decrease of the intensity spectrum (Tokito et al., 1997). For the low order cavities, oblique resonances corresponding to lower emission wavelength are suppressed. Although the overall extraction (both spectrally and angularly integrated) is not improved, a sharply directed narrowed emission with increased intensity at the resonant wavelength can be obtained. The induced spectral narrowing has shown to be useful for the realisation of pure colours emitters (Tokito et al., 1999). Directionality, while being not suited for display applications, can be employed for example in the case projection devices.

3.1.2 Photonic band gap crystals (PBG crystals)

A modification of the optical environment of an emitter by means of a wavelength scale periodical modulation of the refractive index can be used to affect its emission properties (Yablonovitch, 1987). Three-dimensional periodic structures, so called photonic crystals, can open a frequency gap in the photon density of states leading to the inhibition of the emission at the undesired frequencies and directions in the patterned material. The emission will then be redistributed in the structure allowed modes. A description of possible photonic structures geometries and applications is given by Joannopoulos et al. (1997).

The PBG crystal concept was more recently applied to the specific case of LEDs. The effect of a strong two dimensional periodic variation of the refraction index (higher than 2) of a thin slab, provided by an hexagonal array of cylindrical holes through the slab, is expected to increase the extraction efficiency following the suppression of the waveguided modes of frequencies falling in the band gap (Fan et al., 1997). The extraction efficiency for an emitting dipole in the middle of a few periods structure has been calculated to jump sharply from 15% to 70% for the band gap frequencies. In addition to a frequency band gap leading to a direct change of the emission properties, the photonic structure provided a cut-off frequency above which light is extracted through diffraction processes. In this respect, a second strategy was to take benefits from this later effect with the realisation of a photonic band gap structure around the emitting zone (Boroditsky et al., 1999). In this case, the spontaneous emission is not directly affected, rather the periodic structure produces a coherent scattering of the guided modes generated by the source. Photoluminescence measurements following optical excitation of the unpatterned area demonstrated a six-fold increase of the light extracted.

The possibility to apply similar photonic structures to the case of OLEDs has been widely investigated. Although photonic structures showing frequency gaps in given directions have been realised with organic materials (see for example studies performed on luminescent opals (Petrov et al., 1998)), operational OLEDs implementing useful band gaps effects have not yet been shown. Rather photonic structures have been used to outcouple the propagating light initially trapped in the device.

3.2 Modification of the light propagation properties

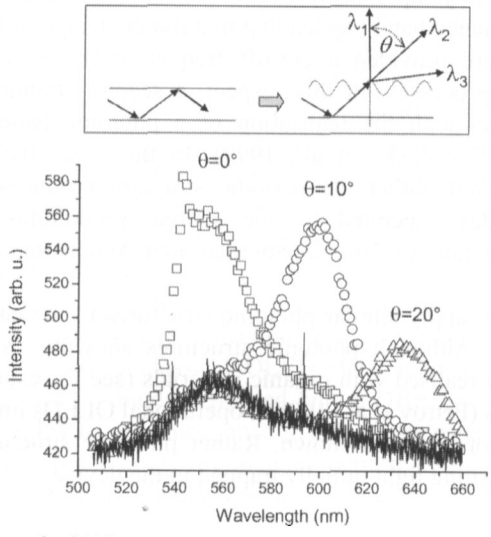
Most of the devices studied were integrating the periodic structure directly in the emissive area. First electrically driven structured OLEDs were incorporating a sub-micrometric periodic grating realised by traditional lithography means on a substrate used as a mould for OLED. Such procedure has revealed the possibility to double the extraction efficiency due to Bragg diffraction of the modes initially trapped in the layers (Lupton et al., 2000).

The action of the grating results more precisely in a modification of the trapped wave vector following diffraction effects. Diffracted waves with an in-plane wavevector lower than the free space wave vector k_0 will radiate in a direction θ given by:

$$k_0 \sin\theta = \pm\beta_g \pm p\frac{2\pi}{\Lambda} \tag{3}$$

where β_g is the propagation constant of a trapped mode in the layers, Λ is the grating period, and p is an integer corresponding to the diffraction order. The result is an increased extraction with an angular dependent emission spectrum related to the dispersive nature of the grating. Such effect is clearly illustrated in Figure 5, where the emission of the modulated layer is characterised, in comparison with the emission of the planar one, by an additional intensity band which spectral position depends on the collection direction θ. This additional band corresponds to a part of the energy initially trapped inside the layer, which is extracted by the grating leading to an increase of the external efficiency (see the above scheme in Figure 5). The angular dependence of the extracted light spectral structure, shown for three observation angles $\theta = 0°$, 10° and 20°, is related to the dispersive nature (equation (3)) of the grating.

Figure 5 Angle dependent photoluminescence of a Rubrene doped Alq3 layer deposited onto a planar (full line) and surface modulated (symbols) aluminium electrode. The modulation period was Λ = 430 nm for a Δh = 65 nm amplitude (see online version for colours)

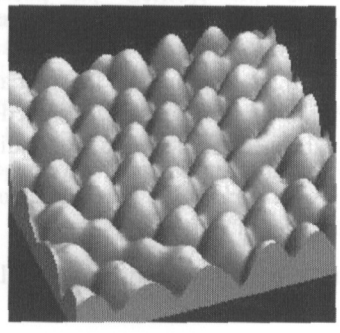


Source: Rocha et al. (2003)

The exploration of sub-micrometric periodical structures to control the organic devices emission properties has been supported by the development of a new set of patterning methods, the so-called soft lithography. Such methods do not employ solvents, a main drawback of traditional lithography for organic devices, and allow patterning through single step low cost process. Among these techniques, embossing (Lawrence et al., 2002), stamping (Granlund et al., 2000) and optically induced mass-transport patterning (Cocoyer et al., 2006) methods have successfully been implemented in electrically driven organic devices. The ease to realise structures in the optical wavelength scale with tailored characteristic dimensions through simple techniques adapted to organic materials has favoured the investigation of optimised geometries for the improvement of OLEDs performances. The ability to structure any of the OLED layer stack interface, from the ITO anode to the metallic cathode, is of particular interest. Indeed, the possibility to pattern the interface close to the maximum of the electric field transverse distribution of the guided mode can enhance the coupling with the radiative modes (Yariv, 1973). The optimal case will correspond to structures presenting the higher optical confinement factor (the ratio of the optical intensity comprised in the patterned area to the total optical intensity of the mode). In this respect, in the case of small molecules based OLEDs, the layer deposition by vacuum evaporation techniques should be helpful in allowing a transfer to each interfaces of a periodic surface modulation initially present on the substrate.

Soft-lithography methods have also shown the ability to realise bi-dimensional gratings in view of an increased control on the trapped modes propagating over all the device plane directions. Recently, a three-fold increase of the photoluminescence intensity extracted from an Alq3 doped rubrene layer has been achieved using in-plane omni-directional structures (Figure 6) generated by means of an original single step all-optical patterning method (Hubert et al., 2005). Operating OLEDs implementing bi-dimensional stamped surface gratings have evidenced an almost doubling of the extraction efficiency in comparison with one dimensionally patterned device (Ziebarth et al., 2004). An overall increase of 25% was shown by the authors, which is equivalent to the enhancement recently observed for an OLED patterned through conventional lithographic means (Fujita et al., 2005). Although different device geometry prevents a direct comparison between the above publications, the lower symmetry of the squared gratings compared to the omni-directional patterning may be partly responsible for the observed lower extraction efficiencies.

Figure 6 Atomic force microscopy image (4 × 4 μm) of bi-dimensional structures realised following a single step all optical patterning process on azo-dye copolymer film (Hubert et al., 2005). Modulation amplitudes reach 70 nm (see online version for colours)



Alternatively, randomly textured surfaces have also been successfully applied in the case of LEDs to redirect trapped photons in the free space (Windisch et al., 1999; Schnitzer et al., 1993).

Although significant enhancements in the extraction efficiency have been demonstrated, there is still room for improvement in view of the energy lost in the devices.

In this respect, another example where such periodic structures can be useful concerns coupling of the emission into the SP modes associated with the metallic cathode (Barnes et al., 2003). SP modes are the result of the interaction between an electromagnetic wave and the free electrons of the metal leading to a hybrid propagating wave trapped at the interface between the dielectric and the metal. As SP modes are characterised by a defined momentum, their propagation properties can be manipulated in a manner a similar to the guided modes discussed above (Ritchie et al., 1968). The ability of periodic structures to couple the energy carried by the SP mode into radiation modes with an efficiency of up to 80% has been demonstrated (Moreland et al., 1982). The use of photonic structures with frequency gaps (see for example Salt et al., 2000) in order to block or enhance the coupling to the SP modes has also been suggested.

Concurrently to the patterning of the active layers to outcouple light guided in the high-index organic/anode layers, studies performed to expand the substrate escape cone have led to comparable extraction efficiencies. The introduction of a small refractive index silica aerogel layer between the OLED stack and the substrate has demonstated to be a simple and efficient method to suppress the substrate wave-guiding effect (Tsutsui et al., 2001). Other approaches consisted of introducing scattering elements in the substrate or modifying its geometry. Substrates of appropriate refractive index and presenting a spherical surface in the backside of the OLED have shown to enhance the total energy extraction by a factor of 3 with a normal emission improved by a factor of 10 (Madigan et al., 2000). More recently, Möller and Forrest (2002) have demonstrated that the use of a substrate patterned with an array of 10 μm diameter microlenses could give equivalent extraction efficiency while correcting the angular modification of the radiated pattern.

4 Conclusions

In summary, we have described materials and OLED device performances showing that the technology is now mature for industrial applications like advanced displays or solid-state lighting. Highly efficient devices can be obtained by using phosphorescent materials with nearly 100% internal quantum efficiency. Lifetimes of commercial products (music players, displays for mobile phone) are typically rated at 10,000–20,000 h. However, larger display panels for TV sets seem to be still many years away. The key issue of light trapping in the planar OLED structure has been discussed through the description of strategies investigated to improve light extraction. The employed strategies have been classified into two main approaches: the extraction improvement by means of

- a direct modification of the material emission
- a modification of the emitted light propagation in the OLED structure.

Experimentally comparable efficiency improvements have been evidenced with both approaches. A doubling of the extracted light intensity has been obtained with cavity and nanopatterned OLEDs, showing that there is still room for optimisation. The angular and spectral modifications of the device emission may also be useful for the applications involving projection devices, in the case of displays based conversion filters or to directly generate pure colours.

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