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# Fast and cost effective fabrication of microlens arrays for enhancing light out-coupling of organic light-emitting diodes

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## Abstract

The efficiency of organic light-emitting diodes (OLEDs) deposited on flat substrates is strongly limited by the total internal reflection at the air-substrate interface. An effective strategy to reduce the amount of substrate modes and enhance the light out-coupling into the air is attaching a microlens array (MLA) on the external surface of OLEDs. In this study, polymeric MLA with periods between 1.2  $\mu\text{m}$  and 2.0  $\mu\text{m}$  are patterned by plate-to-plate nano-imprint lithography using metallic stamps structured by direct laser interference patterning. When MLA with a spatial period of 2.0  $\mu\text{m}$  and a structure depth of 200 nm are employed on red, green and blue OLEDs, the external quantum efficiency is increased to 11.4%, 6.6% and 12.7%, respectively, due to a reduction of internally reflected radiation at the air-MLA-glass interfaces.

**Keywords:** Organic light-emitting diode, light out-coupling, direct laser interference patterning, nano-imprint lithography, laser processing, polymers

## 1. Introduction

Recently, significant effort has been invested in research and development of organic light-emitting diodes (OLEDs) for lighting and display applications <sup>[1,2]</sup>. However, its overall efficiency is strongly limited by the low light out-coupling due to the large difference in the refractive indexes of

air, glass and organic layers [3,4]. In typical OLEDs, about 20-30% of the generated light is emitted directly into the air and roughly the same amount is coupled to the glass substrate as wave guided modes [5,6]. One simple and effective strategy to extract the substrate modes and therefore to enhance the device efficiency is attaching on the OLED front surface a microlens array (MLA) made with low-cost transparent polymers [7,8]. Commonly, such MLAs are fabricated with photolithography involving numerous time consuming processing steps, which can be a hindrance for mass production scale [8,9]. In part, this difficulty can be overcome by stamping the polymer with a structured master using hot-embossing methods based on conventional flat presses or continuous roll-to-roll (R2R) systems. A fast and cost effective method for producing the MLA master with high resolution and quality is Direct Laser Interference Patterning (DLIP). In this method, two or more laser beams are overlapped at a given intercepting angle to generate an interference pattern on the sample's surface. If the used energy density is above the material ablation threshold, then the material can be selectively ablated at the maxima positions of the interference pattern. The advantage of DLIP over traditionally micropatterning techniques is that only a single step is needed to obtain the final topography and simultaneously permitting high resolution (up to half of the laser wavelength) and throughput ( $\sim 1 \text{ m}^2/\text{min}$ ) [10–12]. Another important feature of the DLIP technology is its flexibility to pattern curved surfaces, such as cylindrical molds for R2R-NIL systems [13].

In this study, polyethylene terephthalate (PET) MLAs are attached on the top surface of state-of-the-art OLEDs based on three different emitting layers to extract light from the substrate modes into useful far field radiation. Using four-beam DLIP, hole-like structures are patterned on a metal stamp, which is then used as master to emboss the PET foils by nano-imprint lithography (NIL).

## 2. Experimental details

### 2.1 Picosecond-DLIP

Stainless steel (1.4301 AISI 304) molds were structured using a DLIP- $\mu$ Fab unit (developed by the TU-Dresden and FhG IWS), which is based on a solid-state ps-laser with 532 nm laser wavelength (frequency doubled) from neoLASE GmbH and a nominal output power of 2.7 W [12]. For all experiments, a pulse duration of 70 ps and a repetition rate of 10 kHz were set. Detailed descriptions of the used setup can be found elsewhere [14,15].

### 2.2 Plate-to-plate NIL

The patterns on the stamps were transferred to 200  $\mu\text{m}$  thick PET foils by plate-to-plate NIL using an electrohydraulic press from Paul-Otto Weber GmbH. A force of 200 kN, yielding a pressure of 2.2 MPa, was applied for 5 min, while the top and bottom plates were held at 85°C, which is the

PET glass transition temperature. The used parameters for the embossing process were taken from Ref. [16].

### 2.3 Topography characterization

The structured stamps and PET samples were characterized using a confocal microscope (Sensofar S neox) equipped with a LED white light source and using an objective with 150X magnification. This instrument has a vertical and lateral resolution of 1 nm and 140 nm, respectively.

### 2.4 OLED fabrication and characterization

Red, green and blue OLEDs were prepared on glass substrates using vacuum thermal evaporation (for further details, refer to the Supplementary Data). The MLAs were pasted onto the OLEDs glass surface with a very thin refractive index matching oil (Immersöl 518 F). The OLEDs were characterized in a home-made automated system constructed with a source-meter (Keithley 2400) together with a silicon photodiode and a calibrated spectrometer (Instrument Systems GmbH CAS140CT). The OLEDs external quantum efficiency (EQE) was measured in a calibrated integrating sphere (Labsphere LMS-100).

## 3. Results and discussion

### 3.1 Stamps and imprints topography

The first step of the MLA manufacturing is using DLIP to structure the stamps with the negative of the desired MLA pattern, which is namely a square arrangement of hole-like structures. The studied spatial periods are 1.2  $\mu\text{m}$ , 1.7  $\mu\text{m}$  and 2.0  $\mu\text{m}$ , which, according to previous theoretical works based on ray optics, should already enhance light out-coupling by roughly 30% [8,17]. In order to achieve homogeneous structured surfaces at high processing speeds, the laser parameters, i.e. laser power, pulse-to-pulse overlap and number of pulses, were optimized for each spatial period before patterning the full area. Analyzing the topography, it was found that power and overlap are the main parameters affecting the homogeneity (see Supplementary Data). The optimized DLIP parameters and the resulting laser fluence and spot size for each period are synthesized in Table 1.

	spatial period ( $\mu\text{m}$ )		
	1.2	1.7	2.0
pulse energy ( $\mu\text{J}$ )	2	16.8	16.8
overlap (%)	10	30	20
spot size ( $\mu\text{m}$ )	15	55	60
fluence ( $\text{J}/\text{cm}^2$ )	1.13	0.71	0.59

Table 1. Optimized laser processing parameters used to pattern the stamps.

Using the optimized parameters, hole-like structures were patterned on the  $2 \times 1 \text{ cm}^2$  stamps and then the patterns were transferred by plate-to-plate NIL to produce the MLA on PET. The images in Fig. 1 show the surface topography of the stamps structured with three laser pulses per spot (top row) and the imprinted PET foils (bottom row) with periods of  $1.2 \mu\text{m}$  (left column),  $1.7 \mu\text{m}$  (middle column) and  $2.0 \mu\text{m}$  (right column). A satisfactory homogeneity cannot be achieved in the texture with the lowest period of  $1.2 \mu\text{m}$ , since regions where the surface was not significantly modified are recognizable. This can be attributed to an insufficient pulse-to-pulse overlap due to the small laser spot diameter of only  $15 \mu\text{m}$ . In turn, the  $1.7 \mu\text{m}$  and  $2.0 \mu\text{m}$  period stamps show a better homogeneity, although irregular transition zones can be observed between the areas treated by adjacent laser spots.

The negative of the stamps is accurately reproduced on the PET foils as can be seen in the lower row of Fig. 1. The insets showing the reconstructed 3D surface at a larger magnification allow for a clear recognition of the lens-shaped pattern on the foils.

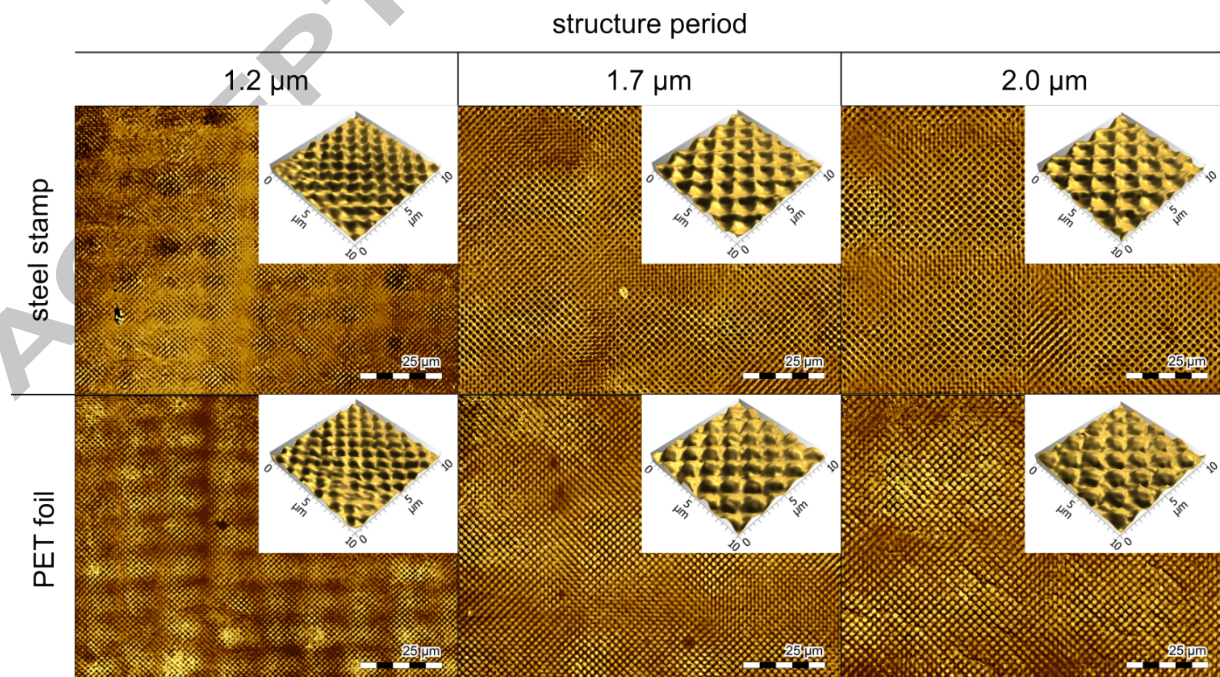


Fig. 1. Confocal microscopy images of the structured stamps (top row) with three laser pulses and the imprinted PET foils (bottom row).



Analyzing the topography of different regions of the stamps and imprinted foils, the average structure depth was determined and summarized as a function of number of pulses in Fig. 2. As expected, the structure depths in the stamps as well as in the PET foils increases with the number of laser pulses [16,18]. It can also be observed that the stamps produced with one and two laser pulses have a similar structure depth than the corresponding imprinted PET, which confirms the proper pattern transfer. Nevertheless, the structure depths on PET replicated from the stamps with three laser pulses are on average 80%-90% lower than the corresponding stamps, which hints that during the nano-imprint the cavities of the stamp were not completely filled, probably due to trapped air between the cavities and polymer.

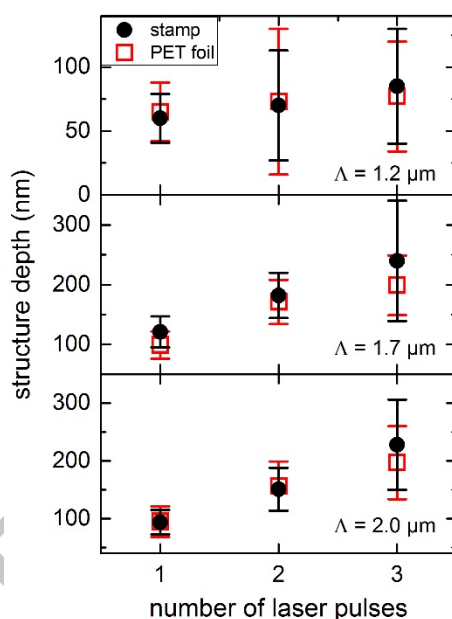


Fig. 2. Dependence of the structure depth of the patterned stamps and PET foils with the number of laser pulses for three pattern periods  $\Delta$ .

### 3.2 Light extraction enhancement of OLEDs

The textures with 1.2  $\mu\text{m}$  period have the shallowest ( $\sim 70$  nm) and less homogeneous structures, and therefore it is not expected that a significant OLED efficiency enhancement can be obtained from them. In turn, the textures with periods of 1.7  $\mu\text{m}$  and 2.0  $\mu\text{m}$  and treated with three pulses are similar regarding the mean structure depth and its standard deviation, suggesting that both samples have a comparable homogeneity. Therefore, MLA with a spatial period of 2.0  $\mu\text{m}$  and a structure depth of 200 nm, corresponding to the stamp processed with three pulses, were selected for testing the out-coupling enhancement.

The refractive index of the PET foils and glass substrates were obtained from ellipsometry measurements (see Supplementary Information). In the spectral from 430 nm to 700 nm, the refractive index of PET ranges from 1.595 to 1.572, whereas the refractive index of glass varies between 1.532

and 1.511. The index matching oil used to join PET foils and glass substrates has a refractive index of 1.518. Using Fresnel's equations to calculate the internal reflection between these materials, it was found that for incidence angles below  $70^\circ$  less than 2% of the generated light is reflected back.

Figure 3 shows the enhancement of the (a) EQE and (b) the spectrally resolved electroluminescence (EL) at a current density of  $15.4 \text{ mA/cm}^2$  for devices with MLA compared to the flat OLEDs. Despite the EQE depends on the current flowing through the device, the relative enhancement of these magnitudes when attaching the MLA is independent on the current. The observed EQE enhancement is attributed to the increased coupling of the substrate light modes into the PET foil and their effective emission to the air due to the interaction with the PET microtextures. Although the improvement in these OLEDs was about 6-13%, it is expected that optimizing the geometry and homogeneity of the structured stamp, the device efficiency could increase by up to 30% compared to the flat device.<sup>[8,17]</sup>

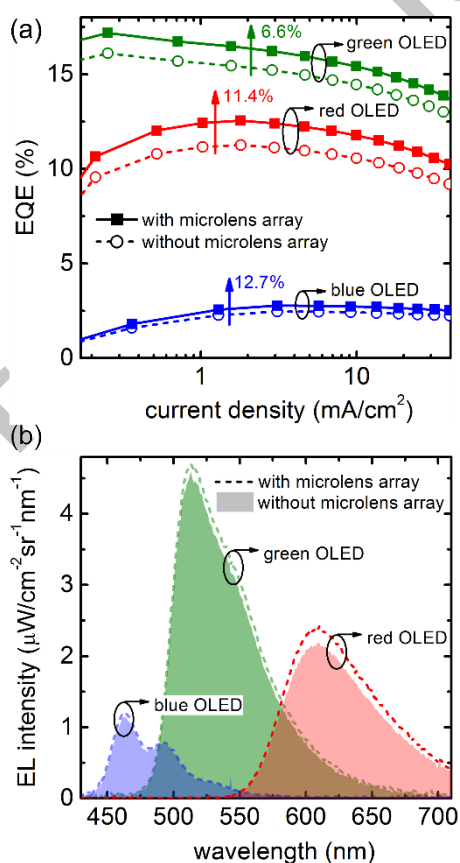


Fig. 3. (a) External quantum efficiency along with current density and (b) spectral electroluminescence intensity of OLEDs at  $15.4 \text{ mA/cm}^2$  with and without the polymeric microlens array.

#### 4. Conclusions

In this study, PET microlens arrays with lenses diameters between 1.2  $\mu\text{m}$  and 2.0  $\mu\text{m}$  were fabricated by nano-imprint lithography using stainless steel stamps structured by DLIP. The patterned PET foils were then successfully used to extract the substrate light modes from OLEDs with three different characteristic wavelengths into the air. The preliminary results are promising, since the optoelectronic characterization reveals that the device efficiency was enhanced by 6-13% with non-optimized MLA geometries. The results shown in this letter demonstrate as proof-of-concept that DLIP can be effectively used to structure in a single-step the stamps for imprinting MLA allowing its integration in large-scale OLEDs manufacturing facilities.

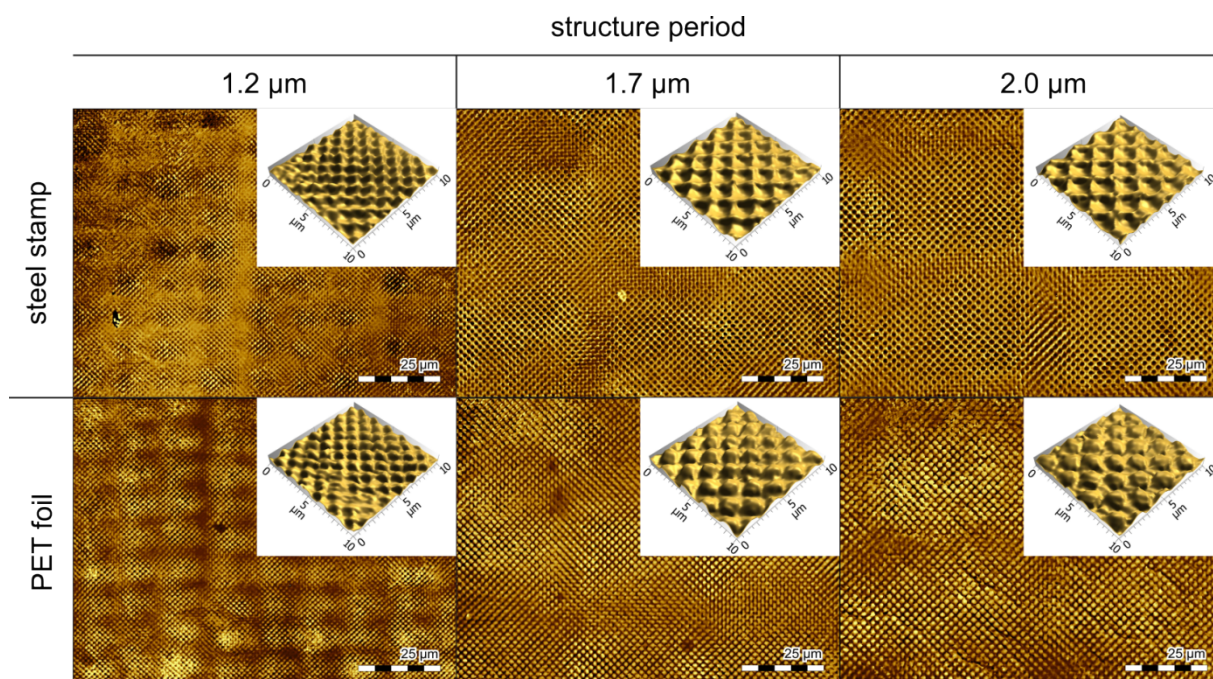
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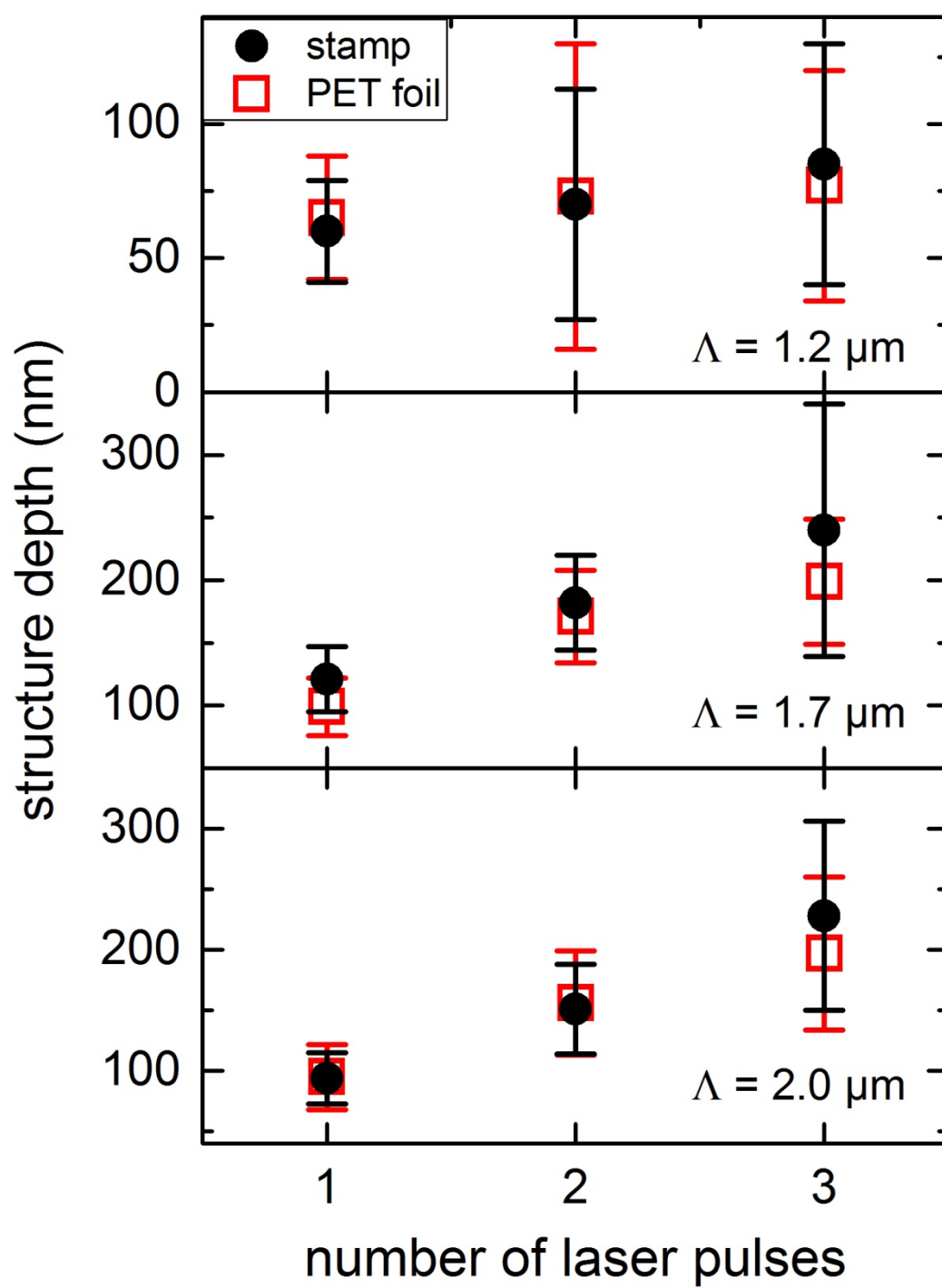
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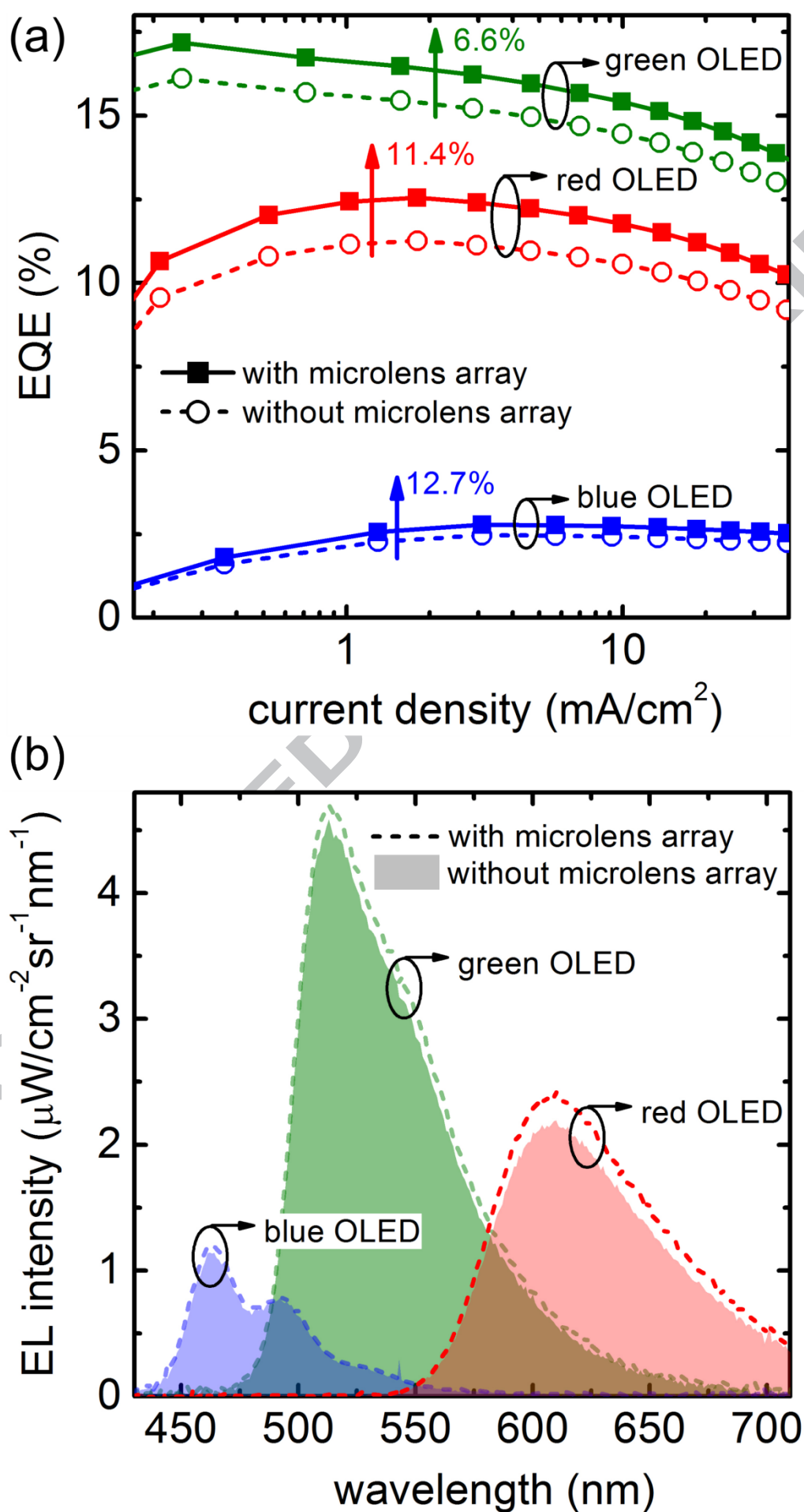
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**Declaration of interests**

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

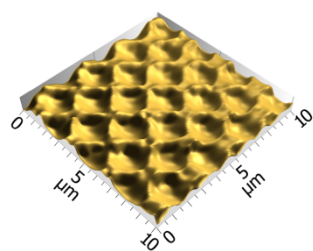
☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

None

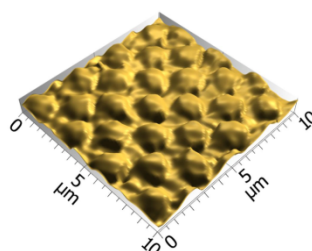
**Conflicts of Interest:** The authors declare no conflict of interest. The founding sponsors had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, and in the decision to publish the results.

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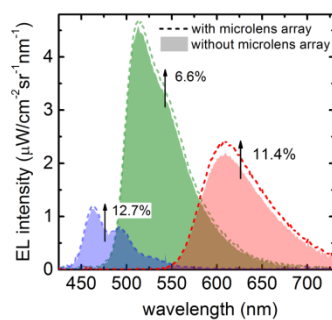




laser processed  
steel stamp



imprinted PET  
microlens array



OLEDs performance  
enhancement

#### Highlights

- Stainless steel stamps were structured by direct laser interference patterning.
- Polymeric microlens arrays were imprinted using the laser processed stamps.
- Microlens arrays were successfully used to enhance light out-coupling in OLEDs.