

POLARIZED ELECTROLUMINESCENCE FROM OLED BASED ON ALQ₃ AS ACTIVE LAYER

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Abstract. The polarized photoluminescence spectra of TPD organic thin films and electroluminescence spectra of OLEDs with ITO/TPD/Alq₃/Al configuration were studied. It has been shown that the linear polarization degrees of photoluminescence of TPD thin films and electroluminescence from the fabricated OLED can be increased by about 3 and 69 times, respectively, using a method of oblique-angle deposition. Such an enhancement is reached due to a more ordered molecular alignment in the organic thin films. The obtained results can be fruitful for designing highly efficient organic light-emitting diodes with optimized parameters.

Keywords: tris-(8-hydroxyquinoline) aluminum, N, N'-bis(3-methylphenyl)-N, N'-diphenylbenzidine, polarized photoluminescence, polarized electroluminescence, organic light-emitting diode

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1. Introduction

The global organic light-emitting diode (OLED) market size is valued at USD 56.37 billion in 2024 and is expected to reach approximately USD 344.58 billion by 2034, according to Precedence Research. One of the most pressing problems in OLED technology is the production of diode structures that emit linearly polarized light, with a dichroic ratio of approximately 40 [1] sufficient for practical use. Such diode structures can serve as the light sources for various purposes – from the backlight of liquid crystal displays and high-contrast OLED screens to the stereoscopic projection systems, optical communications, encryption, and biomedical applications (in particular, for treating wounds, post-traumatic stress disorders, and pain relief) [1–4]. It has been established that linearly polarized light emission can be obtained by incorporating uniaxially oriented materials (e.g., oligomers or liquid crystals) into emitting layers using uniaxial alignment methods, in particular, tensile/shear/polishing, alignment on specific substrates, or Langmuir–Blodgett deposition or liquid crystal self-organization [1, 2]. Although these methods provided some progress, the insufficient degree of polarization remains a significant obstacle in practice.

The OLED devices with the structure of ITO/TPD/Alq₃/Al are among the most widely studied [5–12]. N, N'-bis(3-methylphenyl)-N, N'-diphenylbenzidine (TPD) as a hole transporting and tris-(8-hydroxyquinoline) aluminium (Alq₃) as an electroluminescent layer are used. Koide et al. [5], using ITO/TPD/Alq₃/Al OLED devices as an example, demonstrated that H_μCP soft

lithography can efficiently and rapidly transfer nanoscale hole-blocking chemisorptive patterns to OLED anodes, thereby generating luminous feature sizes as small as 1.0 μm . Ogawa et al. [6] proposed the conduction mechanism of OLED with the structure of ITO/TPD/Alq₃/Al using a one-dimensional discontinuous model. Kim et al. [7] studied the temperature dependence of current–voltage–luminance characteristics of OLED in a device structure of ITO/TPD/Alq₃/Al to understand the electrical conduction mechanism. Lee et al. [8] studied the electrical and optical characteristics as a function of thickness variation of the electron injection material LiF on the OLEDs in the ITO/TPD/Alq₃/LiF/Al device. Devabhaktuni and Prasad [9] transformed ITO/TPD/Alq₃/Al sandwich OLEDs into a sensor by converting the cathode (Al) surface into an active sensing area. The book [10] contains a collection of research developments of different OLEDs. Wang and Ye [11] discussed the influence of Alq₃ deposition thickness and ZnO buffer layer on the photoelectric performance of the OLED device with the structure of ITO/TPD/Alq₃/ZnO/Al. Havare [12] focused on characterizing the performance of the enhanced interface of OLED devices using the self-assembled monolayer (SAM) technique. The optical characterization of OLED devices with configurations ITO/TPD/Alq₃/Al and ITO/SAM/TPD/Alq₃/Al was performed to investigate the effect of aromatic SAM molecules on the luminance and quantum efficiency.

It is known that the oblique deposition method for thin organic films can be used to produce OLED devices with approximately 30% higher quantum efficiency [13]. When the evaporated substance is deposited on a substrate at an inclined angle relative to the vapor flow, islands form that shade parts of the substrate and prevent vapor from entering the shaded areas, leading to ordered, porous, and columnar film growth [13, 14].

Based on our previous studies, we expect low-molecular-weight compounds to be more amenable to orientation-alignment than high-molecular-weight compounds [15–17]. Thus, in [15] we reported the creation of a luminescent material with specific properties, engineered by depositing Alq₃ thin film on ZnO microdisks covered with a silicon substrate. The analysis of the polarized photoluminescence (PL) indicated an almost twofold increase in the polarization degree of the visible PL emission for the obtained samples. The polarization-dependent emission arises from the anisotropic nature of molecular alignment. One possible explanation for the observed increase in the degree of polarization is the interaction between the Alq₃ thin layer and the ZnO surface, which enhances the alignment of molecules in the Alq₃ layer. We also found that the deposition of thin films of dicyanomethylene pyran (DCM) and its derivatives at an oblique angle has a negligible effect on the change in the linear polarization degree of PL [16]. In [17], we show that this parameter of the PL of an Alq₃ thin film can be increased by about 10 times using oblique-angle deposition. This is due to the larger ordering of molecular alignment in this thin film.

This work is a logical continuation of our research presented in [17], which is devoted to the creation of diode structures that emit linearly polarized light with characteristics sufficient for practical use. In the present study, we report the fabrication of an OLED device with polarized emission based on the "small" organic compound Alq₃.

2. Experiment

Organic tris-(8-hydroxyquinoline) aluminium (Alq₃) layers with the thickness less than 50 nm and N, N'-bis(3-methylphenyl)-N, N'-diphenylbenzidine (TPD) layers with the thickness of about 30 nm were thermally deposited in the vacuum 10^{-4} Pa onto the optical glass substrates, transparent for the light with the wavelength exceeding 300 nm or onto

transparent conductive indium-tin-oxide (ITO) glass at the two deposition angles (i.e., the angles between the incoming vapour direction and the normal of substrate) equal to 0° and 80° . The ITO glass with surface resistivity of 70–100 ohm/sq and Alq₃ powder (99.995% purity) were purchased from Sigma-Aldrich Corporation. The powder was purified by recrystallization. We found that the powder's X-ray diffraction pattern is very similar to that of α -phase Alq₃ [15]. TPD powder (99 % purity) was also purchased from Sigma-Aldrich Corporation. Thickness control during the processing was provided by a quartz-crystal deposition-rate controller. Structures of the TPD and Alq₃ molecules are shown in Fig. 1.

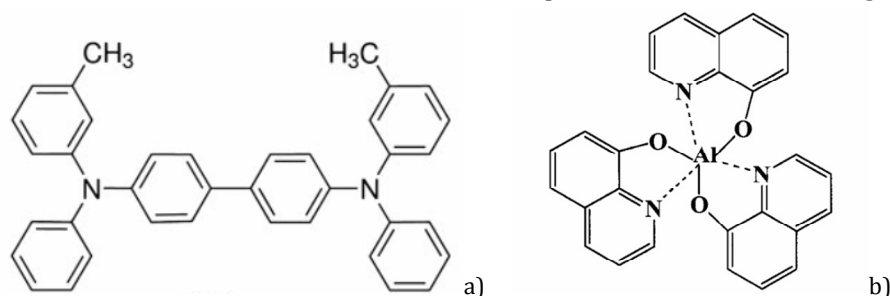


Fig. 1. Chemical structure of the TPD (a) and Alq₃ (b) molecules.

To produce OLEDs, the contact as the top electrode (cathode) to Alq₃ was formed using thermally evaporated Al [18]. The Al was evaporated through the shadow masks onto a circular area with a diameter of ~ 5 mm. A liquid photo-positive resist based on o-naphthochinon-diazide and novolack "Positive 20" (KONTAKT CHEMIE; a type of phenol-formaldehyde resin) was used as the photoresist-insulator [18]. The schematic image of the fabricated OLED is shown in Fig. 2.

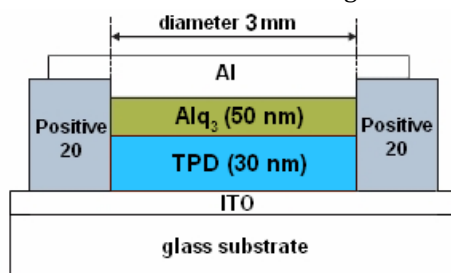


Fig. 2. Schematic structure of the organic light-emitting diodes based on TPD and Alq₃ thin films.

The surface morphology of our samples was examined using a SOLVER P47-PRO atomic force microscope (AFM).

The photo- and electroluminescence (EL) spectra of the samples were measured at room temperature using a portable fiber optic spectrometer AvaSpec-ULS2048L-USB2-UA-RS (Avantes BV, Apeldoorn, Netherlands) with an input slit of 25 μ m, a diffraction grating of 300 lines/mm, and a resolution of 1.2 nm. In the PL and EL measurements, the linear polarizer LPVISE050-A for the wavelength range of 400–700 nm (Thorlabs Inc., Newton, USA) was placed in front of the light-collecting slit in the direction parallel or perpendicular to the oriented organic layers to couple the polarized emission from the sample.

3. Results and discussion

AFM micrographs of the TPD and Alq₃ thin films are displayed in Fig. 3 and Fig. 4, respectively. As expected, the surface topography of the organic thin films deposited on substrates at deposition angles of 0° and 80° was significantly different.

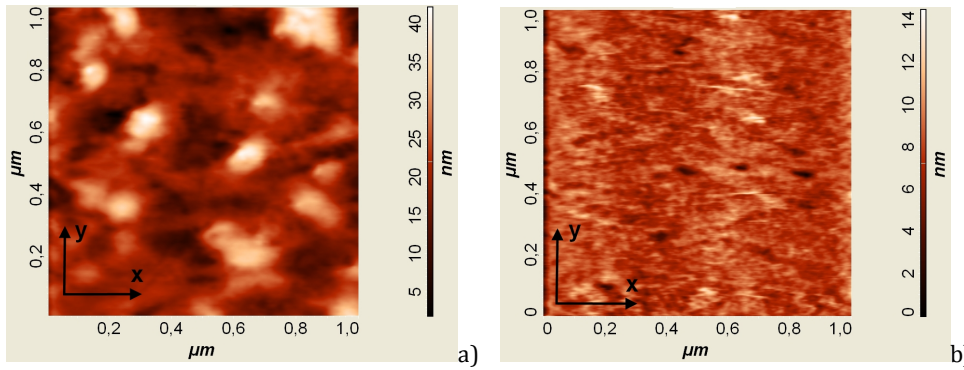


Fig. 3. AFM images of TPD thin films placed on the glass substrates, which were obtained at the deposition angles of 0° (a) and 80° (b).

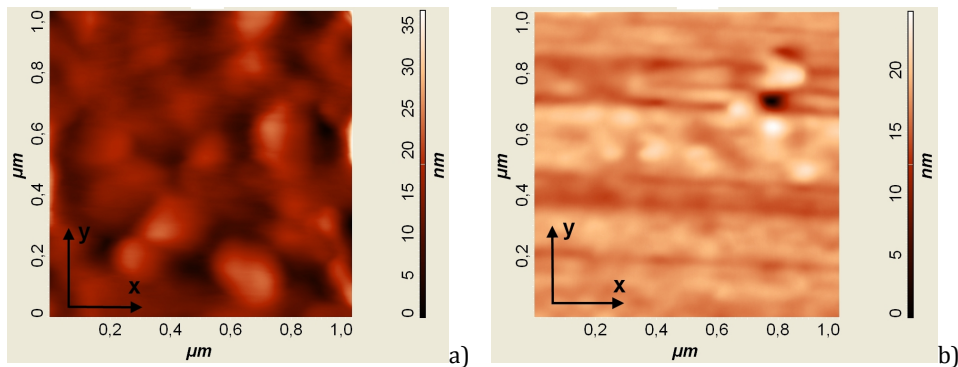


Fig. 4. AFM images of Alq₃ thin films placed on the glass substrates, which were obtained at the deposition angles of 0° (a) and 80° (b).

The polarized PL spectra of TPD thin films are shown in Fig. 5. For all experimental samples, luminescence was observed within the wavelength range of 390–550 nm. The spectra contain a number of overlapping bands with maxima at approximately 409, 423, 451, and 484 nm. The identification of the individual PL bands was performed using Gaussian fits. The most intense band was observed at 423 nm. In general, the PL spectra are similar to those presented in [19–21].

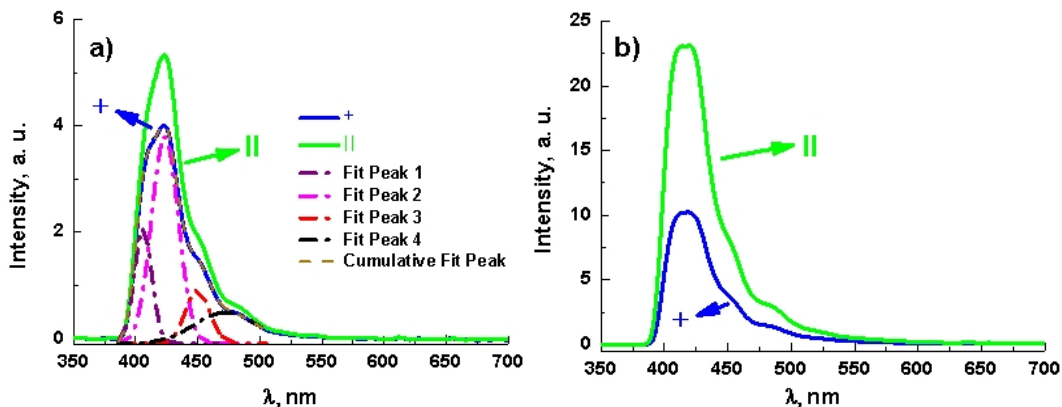


Fig. 5. The PL spectra of TPD thin film polarized parallel to X axis (curve ||) and Y axis (curve +): a) obtained at the deposition angle of 0° , b) obtained at the deposition angle of 80° (see also Fig. 3). The dotted line presents an example of approximation of the PL complex band with Gaussians for TPD thin film (polarization parallel to Y axis). «The approximation was performed in the energy scale and afterwards the Gaussian contours were transformed to the wavelength scale.

Using PL spectral data, the degree of linear polarization ρ was calculated using the equation:

$$\rho = (I_{\perp} - I_{\parallel}) / (I_{\perp} + I_{\parallel}), \quad (1)$$

where I_{\perp} and I_{\parallel} – the luminescence intensities of the perpendicular and parallel components, respectively.

The degrees of linear polarization of PL for TPD thin films deposited on substrates at deposition angles of 0° and 80° did not exceed 0.12 and 0.38, respectively. Therefore, the degree of linear polarization of the PL of the TPD thin film increased by about 3.17 times due to the application of the oblique-angle deposition method.

Meanwhile, according to [17], the degree of PL linear polarization of the Alq₃ film was increased by about 10 times using the oblique-angle deposition method. Such a difference is explained by the fact that TPD is a high-molecular organic compound with a small permanent dipole moment equal to 0.41 D [22], whereas Alq₃ is a low-molecular organic compound with the “small molecules” possessing a fairly high permanent dipole moment equal to 4.1 D [23]. The large molecules of the TPD compound are less easily ordered.

Fig. 6 presents the EL spectra of the heterostructure with the configuration ITO/TPD/Alq₃/Al (0° deposition angle, polarizer was not used). The electroluminescent emission was clearly visible to the naked eye in darkness (see inset in Fig. 6). Its intensity increased with increasing injected current. The broad asymmetric band from approximately 450 nm to approximately 675 nm manifests a peak at about 530 nm. According to the literature data, under normal conditions, the Alq₃ compound emits green light with a maximum at the same wavelength [17].

The polarized room-temperature EL spectra of OLEDs with the ITO/TPD/Alq₃/Al configuration, fabricated by the conventional method and by the method of oblique-angle deposition, are shown in Fig. 7. The degrees of linear polarization of EL for these OLEDs were found to be equal to 0.01 and 0.69, respectively.

The polarizer performance can also be expressed by a single parameter, the dichroic ratio (N):

$$N = I_{\perp} / I_{\parallel}, \quad (2)$$

where I_{\perp} and I_{\parallel} – the luminescence intensities of the perpendicular and parallel components, respectively.

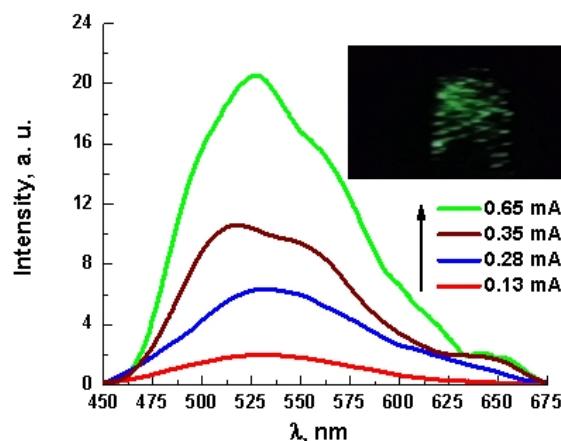


Fig. 6. The room-temperature EL spectra of ITO/TPD/Alq₃/Al OLED (0° deposition angle, polarizer was not used) under different injected currents at forward bias. Inset – the photograph of light emission.

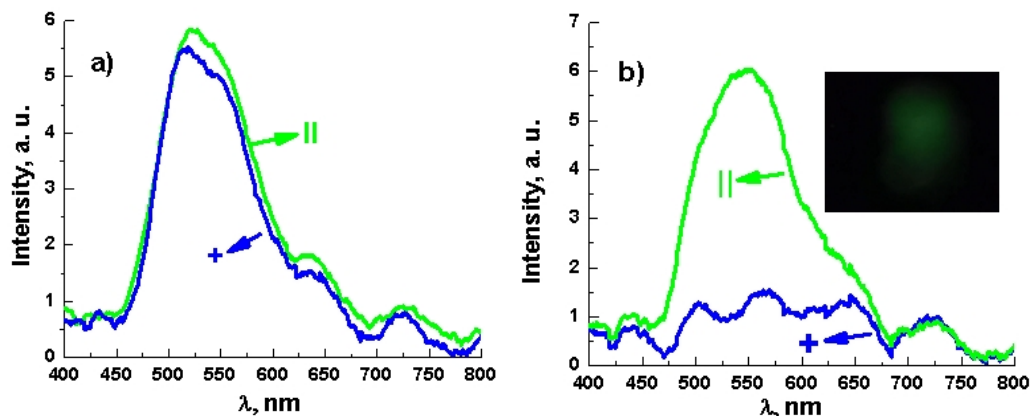


Fig. 7. The room-temperature EL spectra of OLEDs with TPD/Alq₃ thin films polarized parallel to X axis (curve ||) and Y axis (curve +) at the forward current of 0.29 mA and applied voltage of 20.1 V, for the organic layers deposited on: a) substrate at the deposition angle 0°; b) substrate at the deposition angle 80° (see also Fig. 3, Fig. 4). Inset – photograph of light emission.

Among the first known achievements, one can outline OLED with polarized emission and a dichroic ratio of 2.4 [24]. The proposed OLED in this work, with the ITO/TPD/Alq₃/Al configuration and organic layers deposited obliquely, emits polarized light with a dichroic ratio of 5.6.

4. Conclusions

In summary, the polarized photoluminescence spectra of thin organic films of TPD were studied. It has been found that the degree of linear polarization of TPD thin-film photoluminescence can be increased by approximately 3 times using oblique-angle deposition.

The polarized electroluminescence spectra of OLEDs with an ITO/TPD/Alq₃/Al configuration, fabricated by conventional and oblique-angle deposition methods, were investigated. It has been found that the degree of linear polarization of the electroluminescence from an OLED with the ITO/TPD/Alq₃/Al configuration can be increased by about 70 times, using oblique-angle deposition of the organic layers.

It is possible to choose other organic low-molecular-weight compounds for OLED production in further investigations. The obtained results can be helpful in the optimization and design of highly efficient OLEDs.

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Conflict of interest. The authors declare no conflict of interest.

Authors contribution. Monitoring and control of research implementation, [Kapustianyk V.]; co-author of the research idea, [Turko B., Karbovnyk I., Kushnir O.]; investigation, [Turko B., Eliyashevskyy Y., Vasil'yev V., Kolomiets V., Pokora R.]; responsible for coordination of the research, [Bovgyra O.]; consulting on dissemination of research results, [Adamiv V.].

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Анотація. Досліджено спектри поляризованої фотолюмінесценції тонких органічних плівок TPD та спектри електролюмінесценції органічних світловипромінювальних діодів з конфігурацією ITO/TPD/Alq₃/Al. Показано, що ступені лінійної поляризації

фотолюмінесценції тонких плівок TPD та електролюмінесценції виготовлених діодів можна збільшити приблизно в 3 та 69 разів відповідно, використовуючи метод осадження під гострим кутом до нормалі. Таке покращення досягається завдяки більш впорядкованому молекулярному вирівнюванню в тонких органічних плівках. Отримані результати можуть бути корисними для розроблення високоефективних органічних світлодіодів з оптимізованими параметрами.

Ключові слова: трис-(8-гідроксихінолін) алюміній, N, N'-біс(3-метилфеніл)-N, N'-дифенілбензидин, поляризована фотолюмінесценція, поляризована електролюмінесценція, органічний світловипромінювальний діод