



Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl20>

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Version of record first published: 12 Sep 2012.

To cite this article: Yoshitaka Makino, Akinori Okada, Shu Hotta & Takeshi Yamao (2012): Spectrally-Narrowed Emissions from Organic Transistors Composed of Layered Crystals Laminated on a Two-Dimensional Diffraction Grating (Spectral Narrowing from Organic Crystal Transistors), *Molecular Crystals and Liquid Crystals*, 566:1, 8-12

To link to this article: <http://dx.doi.org/10.1080/15421406.2012.701108>

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Spectrally-Narrowed Emissions from Organic Transistors Composed of Layered Crystals Laminated on a Two-Dimensional Diffraction Grating (Spectral Narrowing from Organic Crystal Transistors)

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We observed current-injected spectrally narrowed emissions (SNEs) from a light-emitting field-effect transistor (LEFET) characterized by both layered organic semiconductor crystals and a two-dimensional (2D) diffraction grating. The grating fabricated using nanoimprint lithography had circular air holes forming a triangular lattice. We laminated the p- and n-type crystals in this order on the grating and deposited heterogeneous metal drain and source electrodes. The several SNE peaks were definitively observed when we applied square-wave alternating-current voltages to the gate contact.

Keywords Current-injected spectrally-narrowed emission; organic light-emitting field-effect transistor; two-dimensional diffraction grating; organic crystal; organic layered structure; photonic crystal

Introduction

Organic light-emitting field-effect transistors (OLEFETs) are attracting attention as a candidate to achieve the current-injected spectrally-narrowed emission (SNE) and laser oscillation [1,2]. In the previous studies we reported the current-injected SNE from the OLEFETs where an organic crystal was laminated on top of a diffraction grating that was a part of the gate insulator [3,4]. Meanwhile, we have confirmed that the strong light emission arises from the OLEFETs using a layered structure of p- and n-type organic crystals [5].

In the present studies we combined the layered p- and n-type organic crystals and the two-dimensional (2D) diffraction grating in the OLEFET with heterogeneous metal drain and source electrodes. As a result, we have definitively observed the well-resolved strong SNEs under current injection.

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Experiments

We used 1,4-bis(5-phenylthiophen-2-yl)benzene (abbreviated as AC5, see Fig. 1(a)) [6] as a p-type material, and 1,4-bis{5-[4-(trifluoromethyl)phenyl]thiophen-2-yl}benzene (AC5-CF₃, Fig. 1(b)) [7] as an n-type one. These crystals were grown in vapor phase [8]. For AC5 (AC5-CF₃), we used the source and growth temperatures of 290 and 250°C (265 and 200°C), respectively.

Figures 1c and d show a schematic diagram and a top-view micrograph of the OLEFET, respectively. The 2D diffraction grating (SCIVAX RSLH230/200-4) was fabricated using the nanoimprint lithography. Circular air holes formed a triangular lattice (Fig. 1(e)) onto a cyclic polyolefin resist layer that covered an SiO₂ layer (300 nm in thickness) of an Si wafer. The hole pitch a was 480 nm. The hole diameter and thickness were 238 nm and 225 nm, respectively. The inset of Fig. 1(e) indicates the Brillouin zone and high-symmetry points (Γ , M, K) in the reciprocal lattice [9].

The AC5 and AC5-CF₃ crystals (250–600 nm in thickness) were laminated in this order on the grating (Fig. 1). We further deposited an alloy layer (10 nm) of Mg and Ag, Ag (40 nm) and Au (40 nm) layers in this order on the crystals from over a tungsten wire in close contact with the crystal. The geometry of the deposition metal sources relative to the crystals was described in the literature [10]. The channel length and width were 21 μm and 530 μm , respectively. The grating formed a gate insulator together with the SiO₂ layer. The direction along the channel width was rotated 14° clockwise from the direction connecting Γ and M.

The current-injected emissions were measured in vacuum ($\sim 10^{-3}$ Pa) using the same setup [3, 5] and driving circuit [11] described previously. For the measurements, we designated the source (MgAg) and drain (Au) electrodes as the electron- and hole-injection contacts, respectively. We set the drain (source) voltage from 60 (–60) to 90 V (–90 V). The amplitude of a square-wave alternating-current (AC) gate voltage was varied from 70 to 100 V with a fixed frequency of 50 kHz.

We also measured the optically-excited emission spectra using the same setup as before [12]. For this purpose we took spectra occurring from the device configuration and from the layered crystals laminated on the 2D grating (without the electrodes). With the device we observed the emission parallel to the direction along the channel width. Regarding the layered crystals, the substrate was manually rotated with steps of 5° around the normal to the crystal plane. We used an ultraviolet light (330–380 nm, 0.03 or 0.34 W cm^{–2}) for the excitation source.

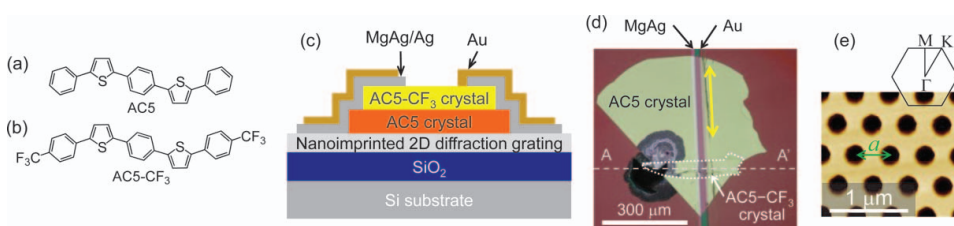


Figure 1. Structural formulae of (a) AC5 and (b) AC5-CF₃. (c) Schematic structure of the OLEFET with a 2D diffraction grating. (d) Top view micrograph of the device. The yellow arrow indicates the direction along the channel width. The diagram (c) represents a cross-sectional view cut along the line A-A'. (e) Atomic force micrograph of the grating. The hole pitch is indicated by a . The inset shows the Brillouin zone and high-symmetry points (Γ , M, K) in the reciprocal lattice.

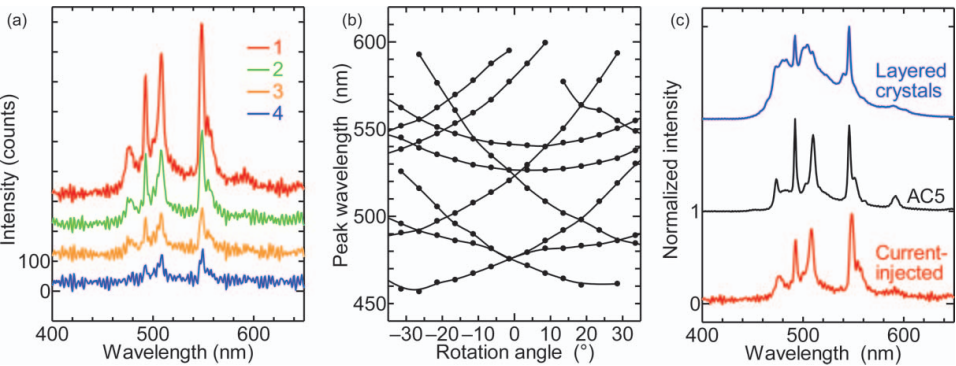


Figure 2. (a) Current-injected spectrally-narrowed emissions from the OLEFET with various source (V_S), drain (V_D), and gate (V_G) voltages at a fixed gate frequency of 50 kHz. 1. V_S : -90 V, V_D : 90 V, V_G : 100 V; 2. V_S : -80 V, V_D : 80 V, V_G : 90 V; 3. V_S : -70 V, V_D : 70 V, V_G : 80 V; 4. V_S : -60 V, V_D : 60 V, V_G : 70 V. In the spectra the spectral data were smoothed using a 3-point adjacent average. (b) Rotation angle dependence of the peak wavelengths for the layered crystals laminated on the 2D diffraction grating (without the electrodes). The data were taken by the optical excitation. (c) Comparison of the current-injected SNE (Spectrum 1) with the optically-excited spectra obtained from the OLEFET device configuration. The portions of the single-layer AC5 crystal and the layered crystals were selectively excited on the channel.

Results and Discussion

Figure 2(a) shows the current-injected emission spectra from the OLEFET. Their peak intensities increased with increasing the magnitude of source (drain) voltage and AC gate voltage amplitude. The SNEs became prominent with increasing peak intensities. Different from our previous studies [3, 4], several lines were concurrently peaked. By peak profile fitting using the pseudo-Voigt functions [13], five distinct lines were sharply resolved with Spectrum 1 at 477.0, 492.6, 508.0 548.2 and 555.3 nm. The line positions and their full-width at half maxima (FWHM) for Spectra 1–4 were summarized in Table 1.

We also observed the several SNE peaks in the fluorescence spectra of the layered crystals laminated on the 2D grating (without the electrodes). Picking up notable peaks, we depict in Fig. 2(b) the rotation angle dependence of their wavelengths. In the diagram, solid curves connect the relevant peaks. The angles 0 to $\pm 30^\circ$ along the abscissa axis represent the Brillouin zone boundaries of M to K; see Fig. 1(e). In other words the direction from

Table 1. Fitted peak positions and their full-width at half maxima (FWHM) of the current-injected spectrally-narrowed emission lines (Spectra 1–4) from the OLEFET device of Fig. 2(a).

Spectrum	Peak position (FWHM) (nm)				
1	477.0 (9.5)	492.6 (3.8)	508.0 (6.9)	548.2 (4.3)	555.3 (9.8)
2	477.5 (9.8)	492.8 (3.2)	507.8 (9.3)	548.4 (4.1)	555.1 (10.5)
3	477.8 (9.2)	492.5 (2.8)	507.6 (9.6)	548.3 (4.1)	555.0 (12.7)
4	$-^a$	$-^a$	507.8 (8.8)	548.7 (4.3)	555.2 (10.1)

^a Fitted data are not shown due to low reliability.

Γ to M is taken as the origin (0°) and that from Γ to K corresponds to the angles of $\pm 30^\circ$. Note that the diagram is nearly symmetrical with respect to 0° due to the triangular lattice symmetry.

The present configuration including the crystals and grating can be regarded as a triangular lattice photonic crystal (PC). The wavelength range where the SNEs were observed (~ 455 – 600 nm) was equivalent to the reduced frequency (a/λ , λ is the wavelength) of ~ 0.80 – 1.05 . These numbers were larger than those corresponding to the photonic band gap calculated for the PC with the similar parameters [14]. Thus we conclude that the SNEs are propagated as the guiding modes of the PC.

Figure 2(c) compares current-injected Spectrum 1 (Fig. 2(a)) with optically-excited spectra obtained from the OLEFET device configuration. To obtain the latter spectra, we selectively excited on the channel portions of the single-layer AC5 crystal and the layered crystals. The peak positions for the single-layer AC5 crystal coincide with those of Spectrum 1 within 4.2 nm. This indicates that the current-injected SNE peaks dominantly arise from the AC5 crystal. Some of the peaks for the layered crystals, however, were invisible behind the broad band emission.

Under the same operating voltages, the maximum peak intensity of the device made of the layered crystals was twice as large as that for a single-layer AC5 crystal device. (Both the devices were fabricated on the grating. The single-layer device had the channel length $25\ \mu\text{m}$ and the width $718\ \mu\text{m}$.) The emission of the single-layer device peaked at 479.69 nm with FWHM of ~ 20 nm, the spectral narrowing being less represented.

Conclusions

We have fabricated the OLEFET characterized by the layered crystals of p-type AC5 and n-type AC5-CF₃ as well as the 2D diffraction grating fabricated by the nanoimprint lithography. The grating was used for the optical resonator and a part of the gate insulator. We have been successful in observing the several well-resolved SNE peaks by applying square-wave AC voltages to the gate contact. Our results indicate that the 2D grating can be used in producing the current-injected SNEs.

Acknowledgements

This work was supported by Grants-in-Aid for Science Research (B) and (C) from the Ministry of Education, Culture, Sports, Science and Technology, Japan. This work was also supported in part by the Mazda Foundation. The authors thank SCIVAX Corporation for preparing the 2D gratings on the substrate. Thanks are also due to Dr. Kazunobu Kojima for discussions on the light propagation within the PC.

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