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3 Review Alignment and patterning of organic single crystals for field-effect 3 transistors

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ABSTRACT

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Organic field-effect transistors are of great importance to electronic devices. With the emergence of various preparation techniques for organic semiconductor materials, the device performance has been improved remarkably. Among all of the organic materials, single crystals are potentially promising for high performances due to high purity and well-ordered molecular arrangement. Based on organic single crystals, alignment and patterning techniques are essential for practical industrial application of electronic devices. In this review, recently developed methods for crystal alignment and patterning are described.

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1. Introduction

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Organic semiconductors have aroused increasing attentions in recent years due to their outstanding performances and potential applications in electronic devices, such as organic field-effect transistors (OFETs) [1-6], organic solar cells [7], organic lightemitting diodes (OLEDs) [8]. These devices exhibit numerous advantages compared with inorganic semiconductor counterparts, including flexibility, low-cost and low-temperature processability [4,9–11]. Of all those fabricated organic electronic devices, OFETs are critically fundamental components for integrated circuits, primarily act as switches and signal-processing elements for practical applications, like radio-frequency identification tags and active matrix displays [2,3,12]. Normally, device performances have extremely close relationships with crystalline structures and imperfections of the crystalline structures always account for poor performances [13,14]. Imperfections such as grain boundaries and molecular disorders not only influence the quality of obtained crystalline morphologies, but also hinder the charge transport by scattering charge carriers. Therefore, single-crystal organic semiconductors, which perform excellent properties with the highest order as well as purity, lead to superior charge carrier mobility

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among organic electronic materials [15–18]. So far, many methods 30 have been proposed to fabricate organic devices based on organic 31 single crystals [19-21]. 32

Despite of all the advantages above, the scale-up of organic 33 single crystals for practical applications is still challenging since 34 the growth orientation and location as well as alignment of single 35 crystals are usually difficult to control. While large-scale industrial 36 applications with high integration demand high uniformity and 37 minimal cross-talk between neighboring devices [22], it is of 38 extreme importance to pattern single crystals at well-designed 39 locations. The development of patterning and alignment techni-40 ques is indispensable for the realization of integrated devices, and 41 will undoubtedly facilitate the progress of single-crystal organic 42 semiconductors for practical applications [23]. Over the last 43 decade, many efforts have been devoted to promote various 44 strategies for growing organic semiconductor molecules into well-45 aligned patterns as well as ordered arrays with the positions and 46 locations under control, precisely. In this article, we review the 47 recent progress of this research topic [24-36]. 48

2. Crystal alignment

Charge carrier transport in organic single crystals has been 50 demonstrated to be anisotropic [37–39]. Alignment of the crystals 51 52 in a unidirectional fashion is, hence, necessary to achieve the 53 potentially uniform OFET performance. Concentration and/or

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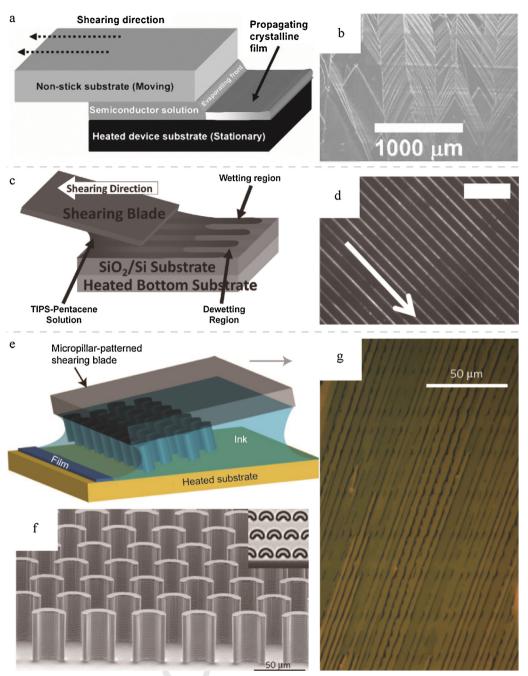


Fig. 1. (a) Schematic illustration of solution-shearing. (b) Bright-field optical microscopy (OM) image of prepared TMS-4T film, showing elongated crystalline structures. (c) Schematic illustration of substrate patterning and crystalline thin film growth by solution-shearing method. (d) Cross polarized optical microscopy (CPOM) image of solution-sheared TIPS-pentacene thin films with 0.5 µm patterned line width. The scale bar is 25 µm and the white arrow indicates the shearing direction. (e) Schematic illustration of solution-shearing using a crescent micropillar-modified blade. The arrow shows the shearing direction. (f) Scanning electron micrograph (SEM) of the micropillar-patterned blade. (g) CPOM image of TIPS-pentacene thin film with micropillars.

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temperature gradients as well as tilted substrates have been used to control the crystallization direction [27,29,40–46]. Solutionshearing and droplet-pinned crystallization (DPC) method are two typical and facile approaches to align organic crystals.

As depicted in Fig. 1a, a solution of organic material was sandwiched between the heated device substrate, which can be modified to improve wetting, and the shearing substrate, which can be modified to cause dewetting. As the upper substrate moved steadily, the front of solution evaporated and created nuclei followed by additional organic molecules continuously flowing towards nuclei and self-organizing to form aligned structures. After solution-shearing, uniform crystalline film of quarterthio-65 phene (TMS-4T) were deposited parallel to the direction of the 66 shearing movement and extended over the entire heated substrate 67 (Fig. 1b) [47]. The shearing rate is a crucial parameter since fast 68 shearing rates lead to very thin films and moderate crystalline 69 structures. By optimizing the shearing speed, the conjugated 70 backbones of 6,13-bis(triisopropylsilylethynyl)pentacene (TIPS-71 pentacene) can be packed more tightly, resulting in reduced π - π 72 stacking distance from 3.33 Å to 3.08 Å [48]. Since the introduced 73 74 lattice strain within the crystal lattice effectively enlarged orbital overlap between component molecules, the charge carrier 75

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