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## SUPPLEMENTAL: A Simple Model of Burst Nucleation

Alexandr Baronov, Kevin Bufkin, Dan W. Shaw, Brad L. Johnson, David L. Patrick Advanced Materials Science and Engineering Center, Western Washington University, Bellingham, WA 98225

## SUPPORTING INFORMATION: EXPERIMENTAL PROCEDURES

ITO-coated glass microscope slides were cleaned by rinsing with isopropyl alcohol and coated with bis(2ethylhexyl)sebecate (BES) (Sigma-Aldrich) by spin coating. Solvent layer thickness was measured by interferometry. Slides were mounted solvent side down against a transparent window in the lid of the home-made deposition chamber. The chamber was sealed and purged with Ar until the oxygen concentration was below 100 ppm. Tetracene (sublimed grade, Sigma-Aldrich) was sublimed from a heated crucible through a pinhole opening at temperatures between 175 - 225 C, depending on the desired deposition rate. The resulting vapor was swept into a flow of Ar gas directed onto the sample through a tapered nozzle. All chemicals were used as received. Further details of the experimental system, including an analysis of the hydrodynamics of gas flow, temperature profiles, and vapor deposition are given in Ref. [1].

Crystal formation was observed in situ by polarized optical microscopy, with the sample viewed through a window in the deposition chamber. Crystals were usually immobile, but if the solvent layer was disturbed small crystals were observed to move with the flow. This suggests crystals were not initially attached to the substrate, i.e. growth did not originate by heterogeneous nucleation at the ITO/solvent interface. Crystals were also fully wet by BES and therefore completely submerged in the solvent. Thus crystals appeared to form as free-floating nuclei.

Atomic force microscopy measurements of crystal thickness were performed using a Digital Instruments Multimode AFM in intermittent contact mode on six representative crystals from the sample shown in Figure 3. The thickness of each crystal was determined by measuring its height relative to the surrounding substrate at 15 randomly selected positions over the area of the crystal (Figure 1). Error bars represent the standard deviation in the thickness measurements.

X-ray diffraction measurements were performed using a Panalytical XPert powder diffractometer on samples that had been briefly rinsed in hexanes to remove excess BES (Figure 2). Reflections occur exclusively from the (001) planes, indicating uniform out-of-plane crystallographic alignment, with the ab-plane of the unit cell oriented parallel to the plane of the substrate. The peak in Figure 2 denoted by an asterisk is attributed to 5,12-napthacenequinone, an oxidation impurity of tetracene.

Optical micrographs of rinsed samples obtained by polarizing optical microscopy showed that the overwhelming majority of aggregates were single crystals, as evidenced by their uniform extinction when rotated between crossed polarizers (Figure 3).



FIG. 1. Average crystal thickness of six representative crystals measured by AFM. Error bars are the standard deviation. Crystal thickness is approximately independent of lateral size.

 D. W. Shaw, K. Bufkin, A. A. Baronov, B. L. Johnson, and D. L. Patrick, J. Appl. Phys. **111**, 074907 (2012).



FIG. 2. X-ray diffractogram of tetracene deposited onto BEScoated ITO/glass after rinsing briefly in hexane to remove excess BES.



FIG. 3. Polarized optical micrographs acquired after deposition showing a representative crystal viewed with different orientations of the sample stage with respect to the polarizers in 15 deg. increments. The uniform change in brightness indicates the crystal is a single monodomain. Most crystals behaved in this way, though a small minority was twinned. Scale bar = 500  $\mu m$ .