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Supplementary Information for: Graphene induced electrical percolation enables more efficient charge transport at a hybrid organic semiconductor/graphene interface

Nicolas Boulanger,[a] Victor Yu,[b] Michael Hilke,[b] Michael F. Toney,[c] and David R. Barbero*[a]

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Grazing incidence X-ray diffraction (GIXD) data was measured in-situ using an area detector. The samples were placed on a heating stage while the X-ray were incoming at an incident angle $\phi = 0.13°$ (see Fig. S1a). The diffracted beams were collected on an area detector, and formed the diffractograms shown Fig. S1b and c for the films on both graphene and silicon. The polar angle χ is defined as shown on the diffraction patterns at 40 °C, with 0° being the out-of-plane direction and 90° the in-plane direction.

Heating to 240 °C removed all long-range order formed during initial spin-coating, and rendered the films disordered. Upon cooling, the film on silicon started to first form edge-on lamellae, visible by a weak 100_z diffraction spot at q≈0.324 Å^{-1} along the z axis. The first face-on lamellae appeared at ≈160 °C, and their amount remained very low (< 1%) compared to edge-on lamellae. On the graphene surface, face-on lamellae first formed at a temperature of ≈205 °C, as shown by the small diffraction peak at approximately the same q in xy.

From the grazing incidence data, it was possible to extract polar χ plots showing the peaks localization along the χ angle, as defined Fig. S1b,c. An example of such plots for both substrates is shown Fig. S2 for the 100 peak at 180 °C and 40 °C. This allows for visualizing the orientation of the peaks for both the edge-on and face-on orientations.

By integrating the polar χ plots for the (100) peak over the whole χ range, it is possible to obtain the total I_100 counts and therefore compare the overall amount of crystallites between the film on silicon and the film on graphene, as shown Fig. S3 where it can be seen that the total amount of crystallites is smaller for the film on graphene compared to the film on silicon.

Cross-sections along q_z have also been extracted to visualize the evolution of the 010 peak in the out-of-plane direction, which is indicative of face-on lamellae formation. Example of such cross-sections are shown Fig. S4a,b during the cooling process at different temperatures for both films on silicon and graphene. It is shown that a peak corresponding to face-on lamellae formation appears on graphene as the film is cooled down, whereas no such peak was detected on silicon. Note that due to the grazing incidence configuration, the diffraction patterns were taken at an angle of 8 degrees away from the out-of-plane direction in Fig. S4a,b. We moreover also measured the (010) peak on both surfaces using a point detector in the Bragg configuration at room temperature after cooling down on the 2-1 beamline at a 12 keV energy at SSRL. This data provides actual out-of-plane diffraction and is shown Fig. 3b in the article. This confirms that the film on graphene has face-on lamellae, whereas on silicon no diffraction peak could be observed due to the weak amount of face-on.

The I-V characteristics of the samples were measured from the bottom to the top of the film, as explained in the article. I-V curves measured at 40 °C are shown in Fig. S5, where the characteristic quadratic evolution of space-charge-limited current (SCLC) current with applied bias can be observed.

[a] *Nano-Engineered Materials & Organic Electronics Laboratory, Umeå University, Umeå, Sweden, E-mail: david.barbero@umu.se*
[b] *Department of Physics, McGill University, Montréal, Québec, H3A 2T8, Canada*
[c] *Stanford Synchrotron Radiation Lightsource, Menlo Park, CA, USA*

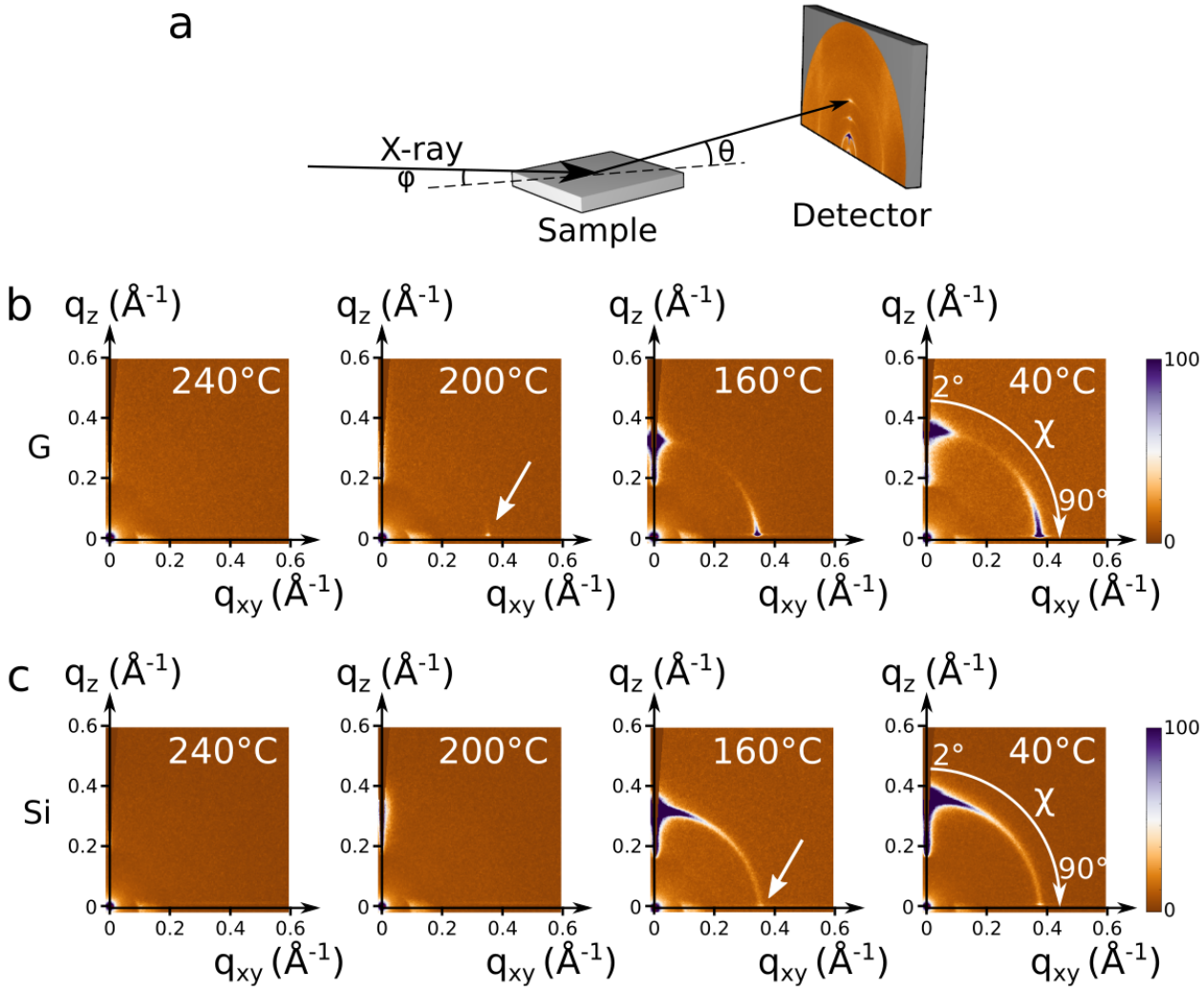

Fig. S1 2D grazing incidence diffraction measured in-situ during cooling of the samples. a) set-up of the GIXD measurement using a heating stage and an area detector. Diffraction patterns of P3HT films on both graphene (b) and silicon (c) at various temperatures during cooling. The scales in (b) and (c) indicate the intensity of the diffraction in arbitrary units from 0 (brown) to 100 (blue). Films were first annealed at 240 °C, and probed with a synchrotron X-ray beam at an incident angle α=0.13° during slow cooling. The angle χ is defined in the diffraction patterns at 40 °C in (b) and (c). The arrows at 200 °C in (b) and at 160 °C in (c) point to the appearance of a diffraction spot in the (xy) plane, indicating the formation of in-plane (100) lamellae.

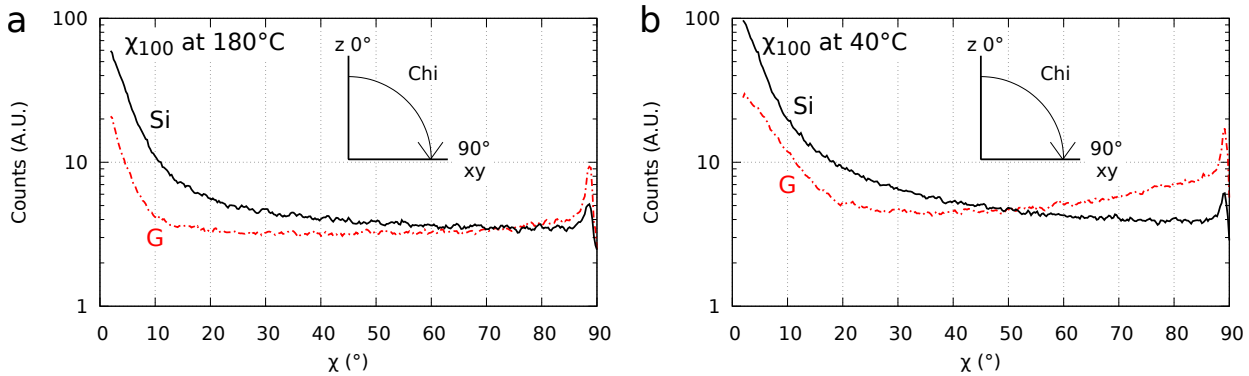

Fig. S2 Chi plots for the 100 peak on silicon and graphene taken at (A) 180 °C and (B) 40 °C.

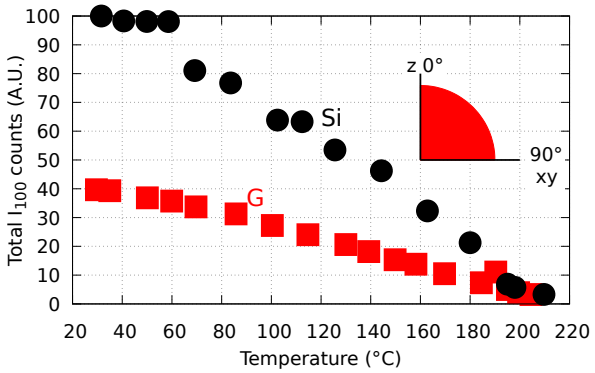

Fig. S3 Overall amount of crystallites from the (100) diffraction ring integrated from χ= 0 to 90° on silicon (Si) and on graphene (G), as a function of temperature.

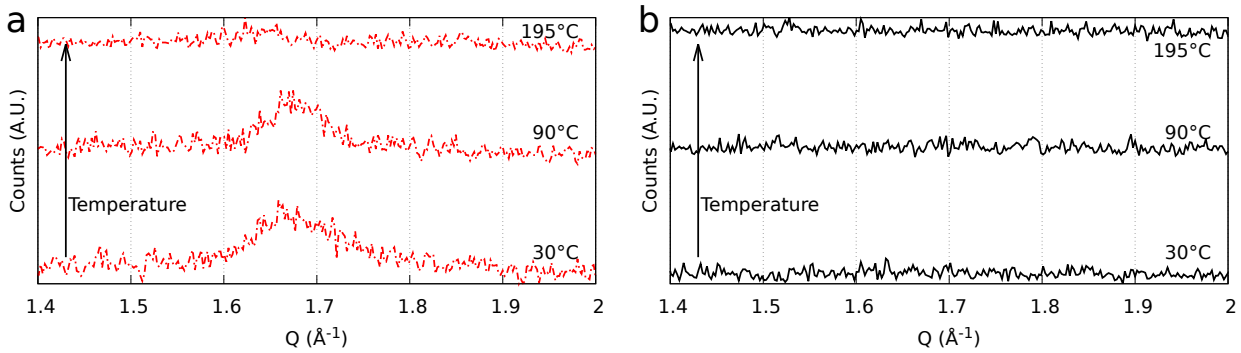

Fig. S4 010 peak for the films on graphene and silicon. The evolution of the 010 peak during cooling is shown for the film on (a) graphene and (b) silicon.

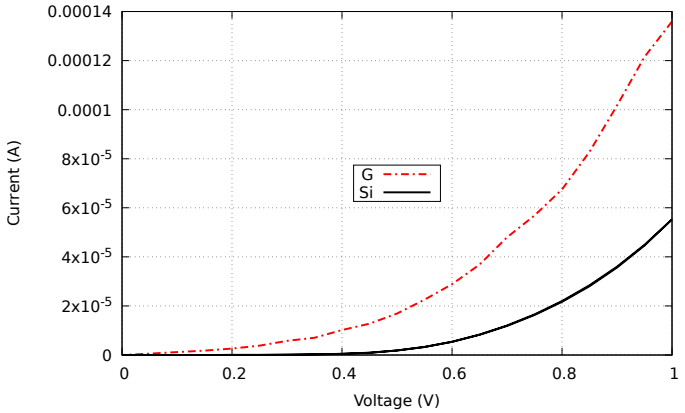

Fig. S5 I-V curves for the films on silicon and graphene measured at 40 °C.