Supporting Information
Low-Energy Electron Irradiation Damage in Few-Monolayer Pentacene Films
A. Tebyani\textsuperscript{1}, F. B. Baalbergen\textsuperscript{1}, R. M. Tromp\textsuperscript{1,2}, S. J. van der Molen\textsuperscript{1}

1: Huygens-Kamerlingh Onnes Laboratorium, Leiden Institute of Physics, Leiden University, Niels Bohrweg 2, P.O. Box 9504, NL-2300 RA Leiden, Netherlands
2: IBM T. J. Watson Research Center, 1101 Kitchawan Road, P.O. Box 218, Yorktown Heights, New York, New York 10598, USA
Part A: Filtering procedure to remove the background from diffraction images

Fig. S1 A median filter is applied to the Fourier transform of each diffraction image to remove the low-frequency noise and background. (a) a measured diffraction peak, (b) Fourier transform of the diffraction peak, (c) median filter applied to (b), (d) inverse Fourier transform of (c). The original and filtered images are normalized to the same value, for both the top and the bottom pairs.
Part B: Damage cross-sections of more samples

Damage cross-sections of (0,0) diffraction peak measured with different beam currents on samples with pentacene film thicknesses of 2-4 monolayers.

Fig. S2 Damage cross-sections of (0,0) diffraction peak versus incident electron energy, measured with two different electron beam currents on the same sample with film thickness of 4 monolayers.
Fig. S3 Damage cross-sections of (0,0) diffraction peak versus incident electron energy measured with different electron beam currents. Datasets with beam current densities 45.6 pA/µm² and 5.06 pA/µm² belong to the same sample. The dataset with beam current density 7.1 pA/µm² belongs to a different sample. Both samples have a film thickness of 3 monolayers.
Fig. S4 Damage cross-sections of (0,0) diffraction peak versus incident electron energy measured on a sample with pentacene film thickness of 2 monolayers.
Part C: Evolution of widths of Lorentzian fits during irradiation

Fig. S5 Evolution of widths of Lorentzian fits to line profile of a (1,1) diffraction peak during irradiation with 10 eV electrons. The thickness of pentacene film is 4 monolayers and the beam current density is 2.96 pA/µm². This figure is obtained from the same recording as Fig.2.
Fig. S6 Bright-field LEEM image of a sample with film thickness of 4 monolayers, obtained with electron energy 3.8 eV. The regions selected by an illumination aperture for damage recordings appear dark after irradiation with 16 eV, 14 eV and 12 eV electrons, respectively, from top to bottom.
Part E: Increase of background intensity in diffraction images due to irradiation

The figures below show the percentage of intensity change for all pixels across the entire diffraction image for the last few frames at the end of a period of exposure compared to the beginning. The images show an increase in the background intensity and a decrease in the intensity around the diffraction spots, indicating that more electrons are scattered incoherently.

Fig. S7 (a) Percentage of change in intensity for all pixels across the diffraction image. The intensity of the last few frames at the end of a period of irradiation is compared with the first few frames. Intensity changes in regions with white color exceed 100%. Thickness of the pentacene film is 4 monolayers. Energy of incident electrons is 8 eV. The beam current density is 2.96 pA/\mu m^2.
Fig. S7 (b) Percentage of change in intensity for all pixels across the diffraction image. The intensity of the last few frames at the end of a period of irradiation is compared with the first few frames. Intensity changes in regions with white color exceed 100%. Thickness of the pentacene film is 4 monolayers. Energy of incident electrons is 16 eV. The beam current density is 2.96 pA/µm²
Fig. S8 Changes in the Electron Energy Loss Spectra of a sample with film thickness of 3 monolayers after a period of irradiation with electron energy 16.4 eV. The peak at 0 eV is the original beam, i.e. the (0,0) diffraction peak. The peak around 14 eV visible in the initial spectrum is due to excitation within the layer caused by the electrons. The peak at the end of the loss spectrum is associated with secondary electrons. This peak increases in intensity during irradiation.
**Part G: Irradiation experiment on a HOPG flake**

Similar irradiation experiments on a HOPG flake yield a negligible decay of intensity of diffraction peaks for the same electron doses. Fig. S9 shows a representative result for 7-eV electrons. Intensity fluctuations in Fig. S9 (especially for the HOPG sample) are attributed to electron beam fluctuations. This result rules out carbon deposition from the background pressure as being responsible for the decay of intensity of the diffraction spots of pentacene.

The HOPG flakes were exfoliated in ambient conditions on a silicon substrate, and subsequently transferred inside the microscope and heated (at UHV pressure of 1.0E-9 or better) at 500°C for many hours to be cleaned.

Exposure measurements were carried out at room temperature, similar to measurements on pentacene samples.

Note again that both the growth of pentacene layers and the measurements on them are carried out in a UHV pressure of ~1.0E-9 mbar or lower to ensure a clean environment for our experiments.

![Graph showing evolution of amplitudes of Lorentzian fits to 0th-order diffraction spot for an HOPG flake compared to a pentacene film upon irradiation with 7 eV electrons.](image)

Fig. S9 Evolution of amplitudes of Lorentzian fits to 0th-order diffraction spot for an HOPG flake compared to a pentacene film upon irradiation with 7 eV electrons. The pentacene film is four-monolayers in thickness. The beam current density is 2.96 pA/µm² for the pentacene sample and 16.1 pA/µm² for the HOPG sample.