Supporting information

Electron dose during experiments

We were using low-dose conditions, keeping the illumination at an absolute minimum, taking a few selected images every 100 °C during the initial heating and blanking the beam in between. During reheating the dose was higher, with images acquired every 10 °C and blanking the beam in between. The dose for each single image varied between $2.3 \times 10^6$ e/nm$^2$ at 300 Kx and $4.8 \times 10^4$ e/nm$^2$ at 56 Kx. During the initial heating, we acquired on average 7 images at different magnifications for each temperature and with this the maximum final dose was less than $10^8$ e/nm$^2$ during initial heating. During re-heating, all images were acquired at 300 Kx and the final total dose during reheating was around $2.2 \times 10^8$ e/nm$^2$. High resolution images were acquired only after heating to 1200 °C and the dose was calculated to be $2.1 \times 10^7$ e/nm$^2$ for each image.

![Figure S1](image1.png)

Figure S1: Merging of smaller domains to form larger ones during heating from 1100 °C to 1200 °C.

![Figure S2](image2.png)

Figure S2: Raman Spectrum of graphitized sample at 1200°C

The spectrum consists of sharp D and G peak. The I_D/I_G ratio is 1.25 and the G peak position is at 1585 cm$^{-1}$. Also a well-defined 2D and D+G peak is visible in the spectrum indicating
the increased graphitic nature of the film. Comparing the position of the G peak and the $I_D/I_G$ ratio to graphite, nanocrystalline graphite, diamond like carbon (DLC) with 20 % sp³ content (a-C) and DLC with 85 % sp³ (ta-c) following the graphitization trajectory suggested by Ferrari et al.¹, the data fits well to nanocrystalline graphite and thus confirms the nanocrystalline nature with 100 % sp² content. The evolution of the sharp 2D and D+G peak also confirms the highly graphitic nanocrystalline nature of the freestanding layer at 1200 °C.

Figure S3: Transformation of a domain with disordered edges to defined faceted edges during prolonged heating. The size of the indicated domain is reducing during heating.

Figure S4: Deposition of amorphous carbon on the sample (a) sample before adsorption of amorphous carbon (b) sample with the adsorbed amorphous carbon and (c) after reheating to 1200 °C. After re heating the sample, the amorphous carbon completely graphitizes increasing the dimensions of the domains.
Figure S5: Migration of a small graphitic structure during heating (a, b). (c), high resolution images showing merging of the graphitic structure with a domain edge (marked by white arrows). The black arrow shows an area where a small cage like graphitic nuclei merged without discontinuity.

Figure S6: Trapped structures before and after reheating.

References