Supplementary Materials for

Synthesis of centimeter-scale high-quality polycrystalline hexagonal boron nitride films from the Fe flux

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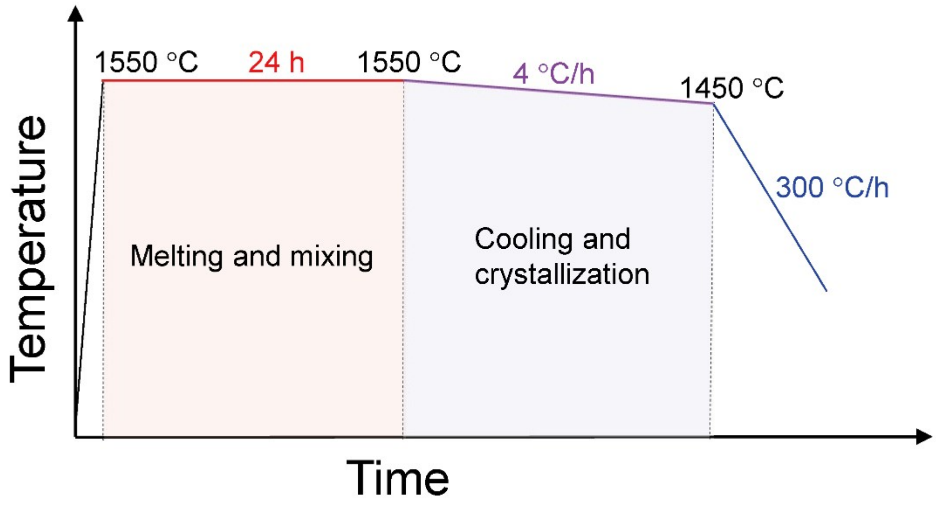
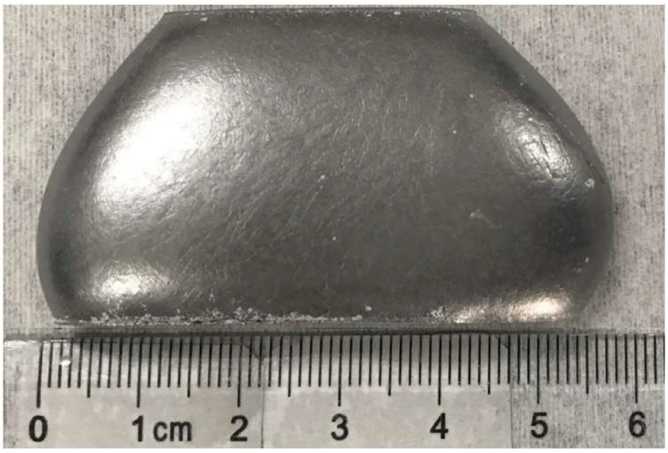
Supplementary Note 1: Laser beam radius

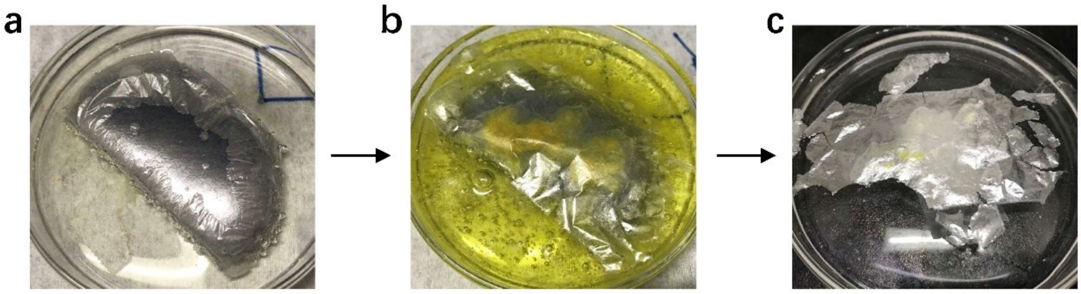
The radius of the laser beam spot (objective lens 100 ×) is an essential parameter for the thermal conductivity measurement, which was obtained by performing a micro-Raman scan across a smooth cleaved edge of an Au-covered Si substrate. Figure S6a-b displays the optical microscopy image of the Si edge and the corresponding Raman mapping of the Si Raman peak at 520.7 cm^{-1} with a step size of 0.1 μm. The measured intensity (*I*) is proportional to the total laser power incident on the sample. Figure S6c shows the measured *I* as a function of the distance (*x*) of the laser beam to the boundary. A Gaussian function $exp(-x^2/{r_0}^2)$ was used to fit the slope dI/dx to calculate r_0, which is estimated to be 0.31 ± 0.01 μm.



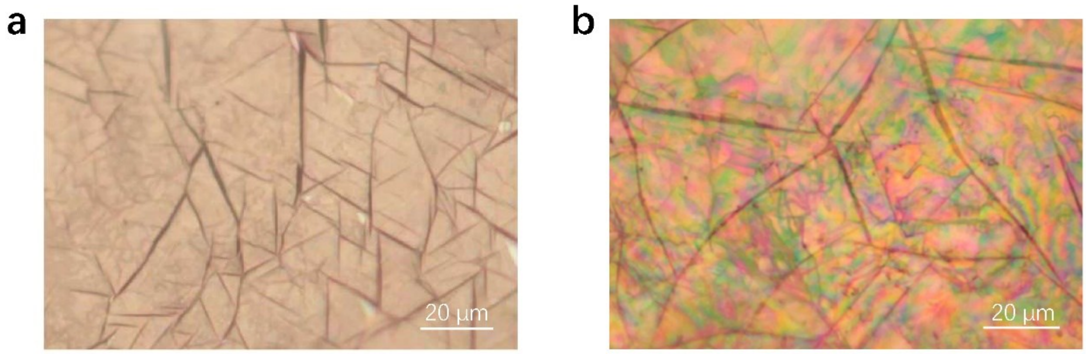
Supplementary Figure 1 Temperature profile for h-BN crystal growth. The sample zone keeps at 1550 °C for 24 hours (red line), and then cools down to 1450 °C at a rate of 4 °C/hour (purple line), followed by quickly quenching (300 °C/hour) to room temperature (blue line).



Supplementary Figure 2 Representative optical image of one Fe ingot covered with hBN films.

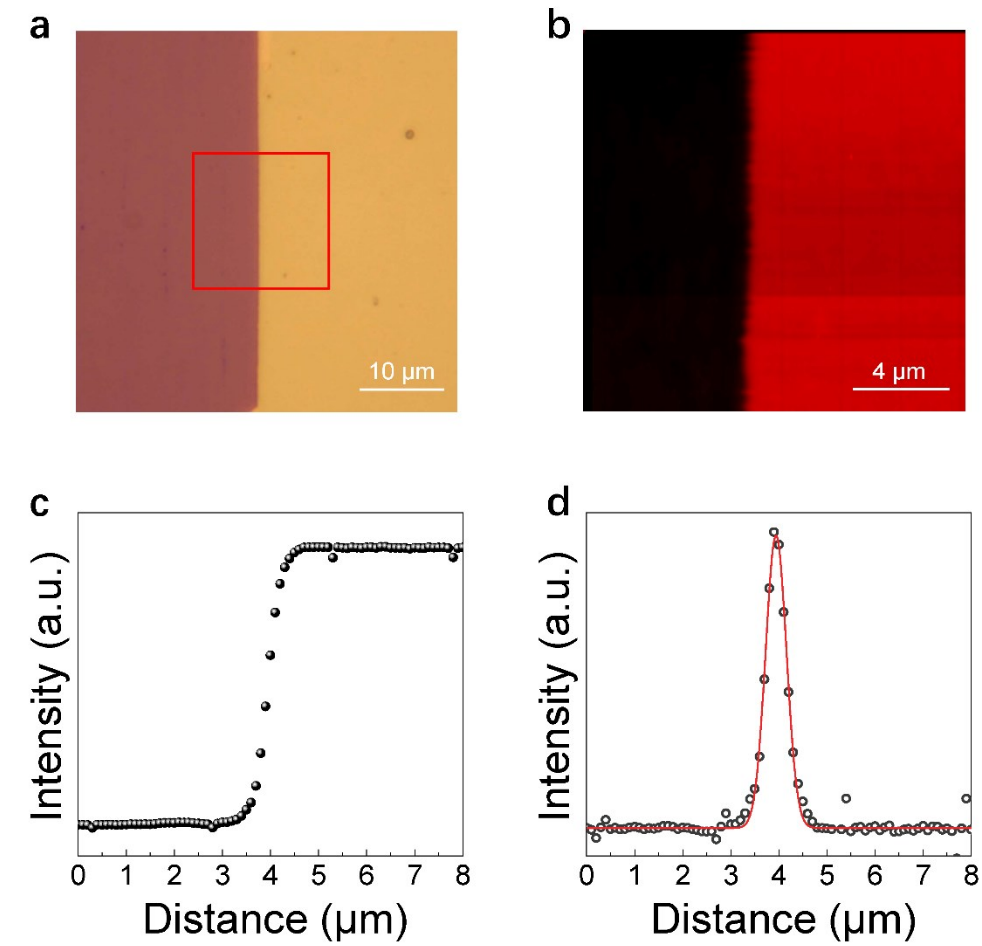


Supplementary Figure 3 Optical images of the exfoliation process. a, One Fe ingot covered with hBN film is immersed into the hydrochloride acid, and the hBN film starts to be exfoliated from the ingot immediately. **b,** The hBN film is wholly separated from the ingot about 1 hour later. **c,** The h-BN film is transferred and washed by the deionized water.

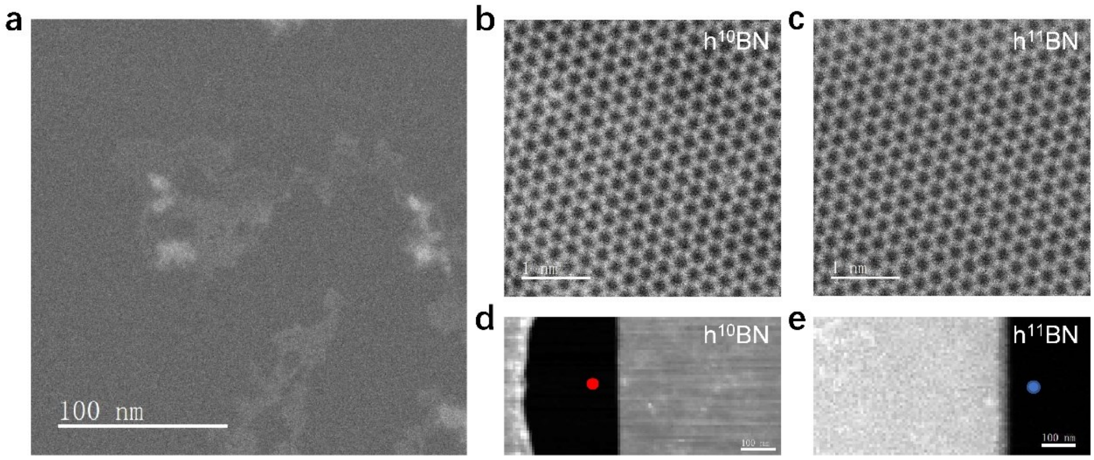


Supplementary Figure 4 Optical microscopy images of hBN films grown at different %B. **a,** h-BN films with 2.4% of %B, showing the uniform thickness; **b,** h-BN films with less than 2.4% of %B, showing uneven thickness-induced colorful surface.

Supplementary Figure 5 Optical image of the product with 25.3% of %B, showing original powder morphologies after the growth.



Supplementary Figure 6 Raman mapping of Si chip partially coated by Au and fitting results. a, Optical microscopy image of the boundary w/ and w/o Au coating on a Si substrate; **b,** Raman mapping of the Si peak at 520.7 cm^{-1} in the squared area in **a**; **c,** Intensity distribution of Si Raman peak across the boundary; **d**, Corresponding Gaussian fitting results of intensity distribution, suggesting 0.31 ± 0.01 μm of the radius of the laser beam spot.



Supplementary Figure 7 Illustration of the STEM-EELS acquisition method. a, HAADF image of $h^{11}BN$ at the large field of view; **b, c,** the unprocessed atomic resolution HAADF image of $h^{10}BN$ and $h^{11}BN$, respectively; **d**, **e**, the low-magnification HAADF image of $h^{10}BN$ and $h^{11}BN$ respectively. The red and blue disk in **d** and **e** are the acquisition positions of STEM-EELS corresponding to Fig. 3c.