Electronic Supplementary Information

Deep learning-assisted analysis of HRTEM images of crystalline nanoparticles

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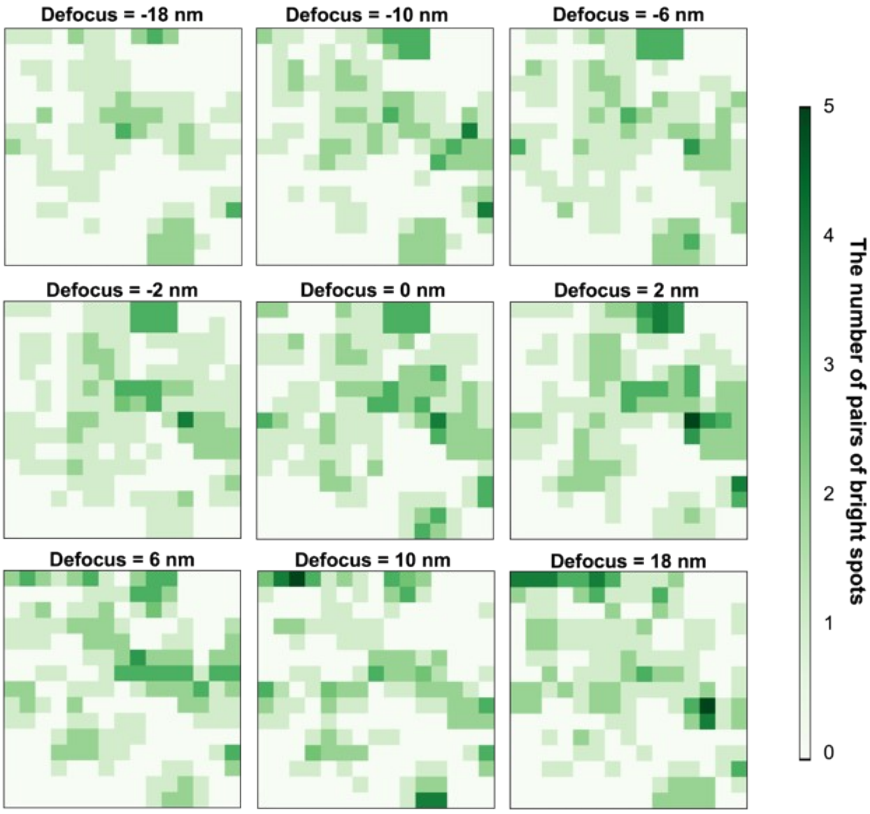
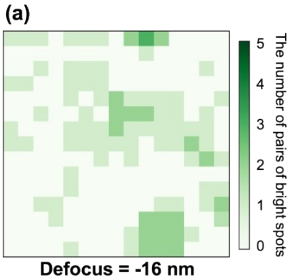


Figure S1. The crystallinity profile of HRTEM images captured at various defocus values, where the defocus ranging from -18 nm to 18 nm.



(b)

[[5, 1, 1]]	[[5, 1, 1]]	[[5, 1, 1]]	0	[[3, 1, 1]]	[[3, 1, 0]]	[[3, 1, 0]]	0	[[2, 2, 0], [1, 1, 3]]	[[4, 4, 0], [2, 2, 0], [2, 2, 2]]	[[4, 4, 0], [2, 2, 0]]	0	0	0	0
0	0	0	0	0	0	0	0	0	[[2, 2, 0]]	0	0	0	0	0
0	0	[[4, 4, 2]]	0	[[4, 0, 0]]	[[4, 0, 0]]	[[4, 0, 0]]	0	0	0	0	0	0	[[5, 1, 1]]	0
[[4, 4, 2]]	[[4, 4, 2]]	0	0	[[4, 0, 0]]	[[4, 0, 0]]	[[4, 0, 0]]	0	0	0	0	0	0	0	0
0	0	0	0	0	0	[[4, 0, 0]]	[[5, 1, 1], [1, 3, 1]]	[[4, 2, 2]]	[[4, 2, 2]]	[[4, 2, 2]]	[[4, 2, 2]]	0	0	[[3, 1, 1]]
0	0	[[3, 1, 1]]	0	0	[[3, 1, 1]]	0	[[4, 4, 1], [2, 0, 3]]	[[5, 1, 1], [1, 3, 1]]	[[4, 4, 1], [2, 0, 3]]	[[4, 2, 2]]	[[4, 2, 2]]	0	[[4, 4, 2]]	0
[[3, 1, 1]]	0	0	0	[[3, 1, 1]]	[[3, 1, 0]]	[[3, 1, 1]]	[[5, 2, 0], [3, 1, 1]]	[[4, 2, 2]]	[[4, 2, 2]]	[[5, 1, 0]]	[[5, 1, 1]]	[[4, 2, 2]]	[[4, 2, 2]]	0
[[3, 1, 1]]	[[3, 1, 1]]	0	0	[[3, 1, 0]]	[[3, 1, 0]]	[[3, 1, 0]]	[[3, 1, 0]]	[[4, 2, 2]]	0	[[5, 1, 1]]	[[5, 1, 1]]	[[4, 4, 2], [1, 0, 5]]	[[4, 2, 2]]	0
0	0	0	0	0	0	[[3, 1, 0]]	0	[[4, 0, 0]]	0	0	0	[[4, 2, 2]]	[[5, 2, 0], [1, 1, 3]]	[[4, 2, 2]]
0	0	0	0	[[3, 1, 1]]	0	0	0	0	0	0	0	0	0	0
0	0	0	0	[[3, 1, 1]]	0	0	0	0	0	0	0	0	[[3, 1, 0]]	[[3, 1, 0]]
0	0	[[3, 1, 1]]	[[3, 1, 1]]	0	0	0	0	0	0	[[5, 1, 1]]	[[5, 1, 1]]	0	[[3, 1, 0]]	[[3, 1, 0], [2, 2, 2]]
0	0	0	[[3, 1, 1]]	0	0	0	0	0	[[4, 4, 0], [2, 2, 0]]	[[4, 4, 0], [2, 2, 0]]	[[4, 4, 0], [2, 2, 0]]	0	0	0
0	0	0	0	0	0	0	0	[[5, 1, 1]]	[[4, 4, 0], [2, 2, 0]]	[[6, 1, 1], [2, 2, 1]]	[[4, 4, 0], [2, 2, 0]]	[[5, 1, 1]]	0	0
0	0	0	0	0	0	0	0	[[3, 2, 1]]	[[4, 4, 0], [2, 2, 0]]	[[4, 4, 0], [2, 2, 0]]	[[4, 4, 0], [2, 2, 0]]	0	0	0

Figure S2. The crystal planes distribution of the HRTEM image of γ-Fe_2O_3 nanoparticle at a defocus of -16 nm.

ESI-1. Standard Database of crystallographic parameters

In order to better traverse and compare the calculation results of the images, an Excel database is established based on the standard PDF card (source from The Inorganic Crystal Structure Database (ICSD)), which include the space groups, lattice constants, and inter-plane distances for different indexed planes. The standard PDF cards used for HRTEM image of γ-Fe_2O_3 nanoparticle in our method is PDF#39-1346 and PDF cards used for HRTEM image of iron oxide nanoparticle with mix phases are PDF#46-1436, PDF#19-0629, PDF#44-1415, PDF#29-0713, PDF#13-0087, PDF#34-1266, PDF#33-0664.

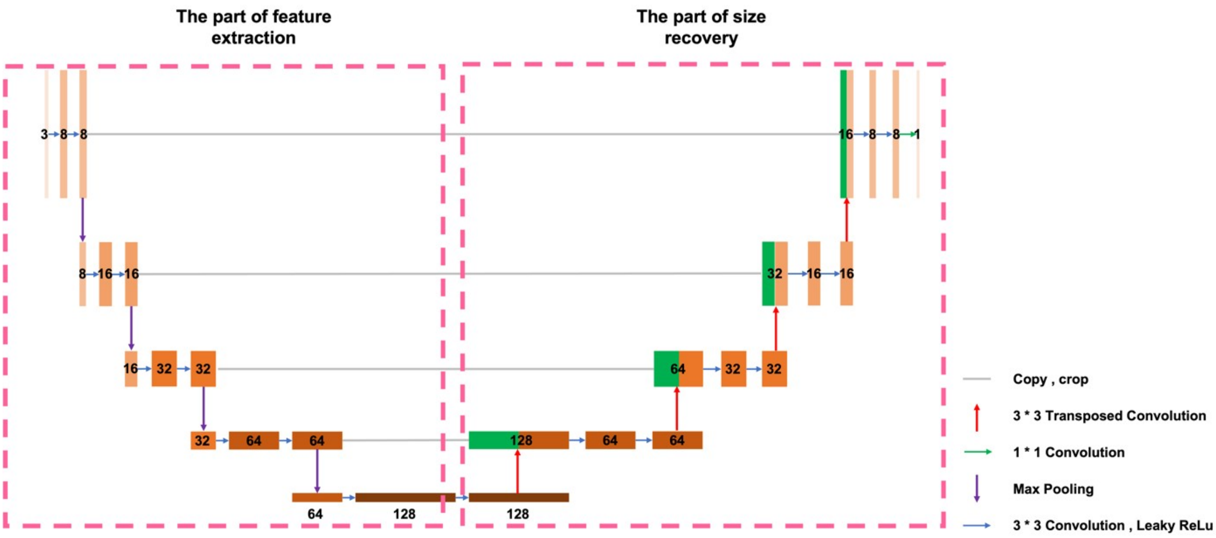


Figure S3. The architecture of U-net deep learning model, which is used to binarize the cropped FFT image.

ESI-2. Calculation method of lattice spacing

In order to facilitate comparison with standard databases, it is necessary to convert the pixel distances (unit: pixels) in the predicted FFT images to lattice spacing (unit: nm) in the HRTEM image. The side length *d* of FFT image represents the reciprocal of the nanometer length corresponding to 1 pixel in the HRTEM image (*p* nm/pixel, which obtained from .dm3 file of HRTEM images). Taking a 2048*2048 image with 256*256 moving window as an example (Fig. S2), the 256 pixels in FFT images corresponds to (1/ 0.016765363) nm^{-1}. The lattice spacing *q* is calculated as:

$$q = \frac{I}{d * p} \qquad (1)$$

where, *I* means the half of the pixel distance between a pair of centrosymmetric bright spots, and *q* means the lattice spacing corresponding *I*. As a result, the lattice spacing is calculated by pixel count and unit conversion in the FFT image.

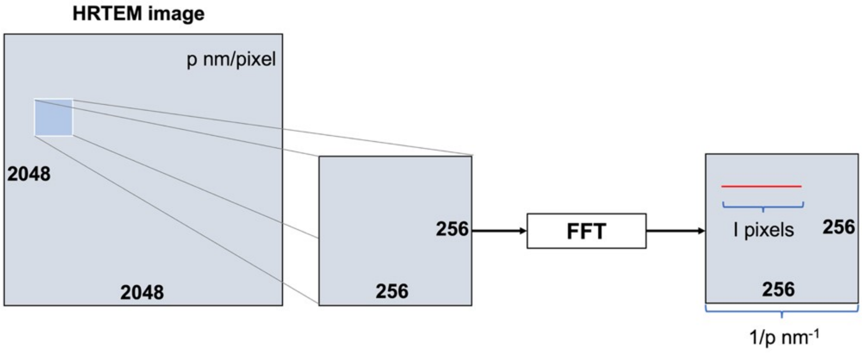


Figure S4. Calculation method of lattice spacing in predicted FFT images. Take a 256*256 moving window in 2048*2048 image as example.

ESI-3. Theoretical angle between two crystal planes in different space groups

For the crystal planes *(h_1, k_1, l_1)* and *(h_2, k_2, l_2)* in different space group, the interplanar angle β is calculated as:

(i) The Orthorhombic:

$$\beta = \arccos\left(\frac{\frac{h_1h_2}{a^2} + \frac{k_1k_2}{b^2} + \frac{l_1l_2}{c^2}}{\sqrt{\left(\frac{h_1^2}{a^2} + \frac{k_1^2}{b^2} + \frac{l_1^2}{c^2}\right)\left(\frac{h_2^2}{a^2} + \frac{k_2^2}{b^2} + \frac{l_2^2}{c^2}\right)}}\right) \qquad (2)$$

(ii) The Cubic:

$$\beta = \arccos\left(\frac{h_1h_2 + k_1k_2 + l_1l_2}{\sqrt{\left(h_1^2 + k_1^2 + l_1^2\right)\left(h_2^2 + k_2^2 + l_2^2\right)}}\right) \qquad (3)$$

(iii) The Hexagonal:

$$\beta = \arccos\left(\frac{h_1h_2 + k_1k_2 + \frac{1}{2}(h_1k_2 + h_2k_1) + \frac{3a^2}{4c^2}l_1l_2}{\sqrt{\left(h_1^2 + k_1^2 + h_1k_1 + \frac{3a^2}{4c^2}l_1^2\right)\left(h_2^2 + k_2^2 + h_2k_2 + \frac{3a^2}{4c^2}l_2^2\right)}}\right) \qquad (4)$$

(iv) The Tetragonal:

$$\beta = \arccos\left(\frac{\frac{h_1h_2 + k_1k_2}{a^2} + \frac{l_1l_2}{c^2}}{\sqrt{\left(\frac{h_1^2 + k_1^2}{a^2} + \frac{l_1^2}{c^2}\right)\left(\frac{h_2^2 + k_2^2}{a^2} + \frac{l_2^2}{c^2}\right)}}\right) \qquad (5)$$

where *a, b, c* in the formulas represents the lattice constant corresponding to different kind of space group.