

Supporting Information

Nanoparticles-induced morphological transformation in block copolymer-based hybrid nanocomposites

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The shape, mean size, and size distribution of the NPs had been characterized by TEM (Tecnai G2 F20, FEI). **Fig. S1a** shows the TEM image of the iron oxide nanoparticles. The nanoparticles are spherical with an average diameter of ~ 5.5 nm. The size distribution histogram of NPs was estimated from counting of 100 individual particles. The estimated polydispersity (PD) was $0.09 \sigma/D$, where σ is the standard deviation and D is the diameter of a particle. The volume-averaged SAXS measurement (GALAXI, JCMS) was used for a more precise determination of the size and size distribution of the nanoparticles. The measured and fitted curves are shown in **Fig. S1c**. The fitting of the radially averaged data was done with the SasView software package and was refined with respect to the spherical form factor. The size and size distribution obtained from the refinement is ~ 5.6 nm (PD: 0.13). The dimensions obtained with the refinement of the SAXS data is in good agreement with the TEM result.

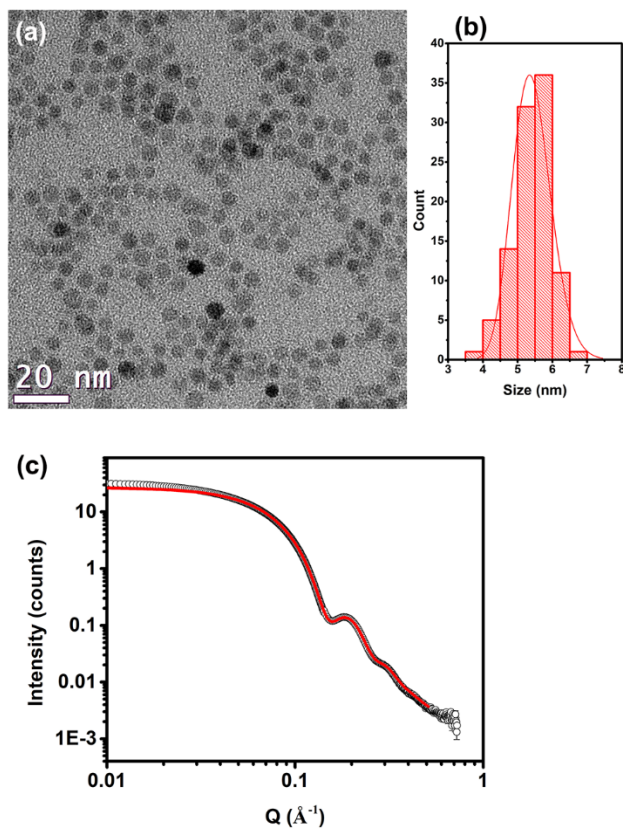


Fig. S1. (a) TEM image and (b) corresponding size distribution histograms of iron oxide nanoparticles. (c) The 1D SAXS spectra of the nanoparticles in solution was radially averaged. A refinement of the spherical form factor is presented by the red solid line.

The surface and topographical properties of the patterned substrate were characterized with AFM (MultiMode8, Bruker) and SEM (SU8000, HITACHI) here. From microscopy results (**Fig. S2a** and **b**), the variations of pitch size and some facet junctions which cannot be avoided come out randomly across the surface of the quasi-periodic saw-toothed substrate. The patterned substrate has an averaged width of the saw-tooth of ~ 140 nm, and the averaged amplitude (29.6 nm) was calculated based on the angular relationship between different planes. **Fig. S2c** shows GISAXS scattering data where the incident beam is parallel to the saw-tooth patterns. The averaged width of ~ 146 nm is obtained by indexing Bragg peaks. **Fig. S2d** shows the experimental GISAXS data with the stripes perpendicular to the incident beam. The majority of scattered intensity is confined in the off-specular line, as expected for this geometry. All GISAXS and GTSAXS data were collected in perpendicular geometry to avoid strong scattering from the patterned substrates that can hinder the scattering from self-assembled nanocomposite thin films.

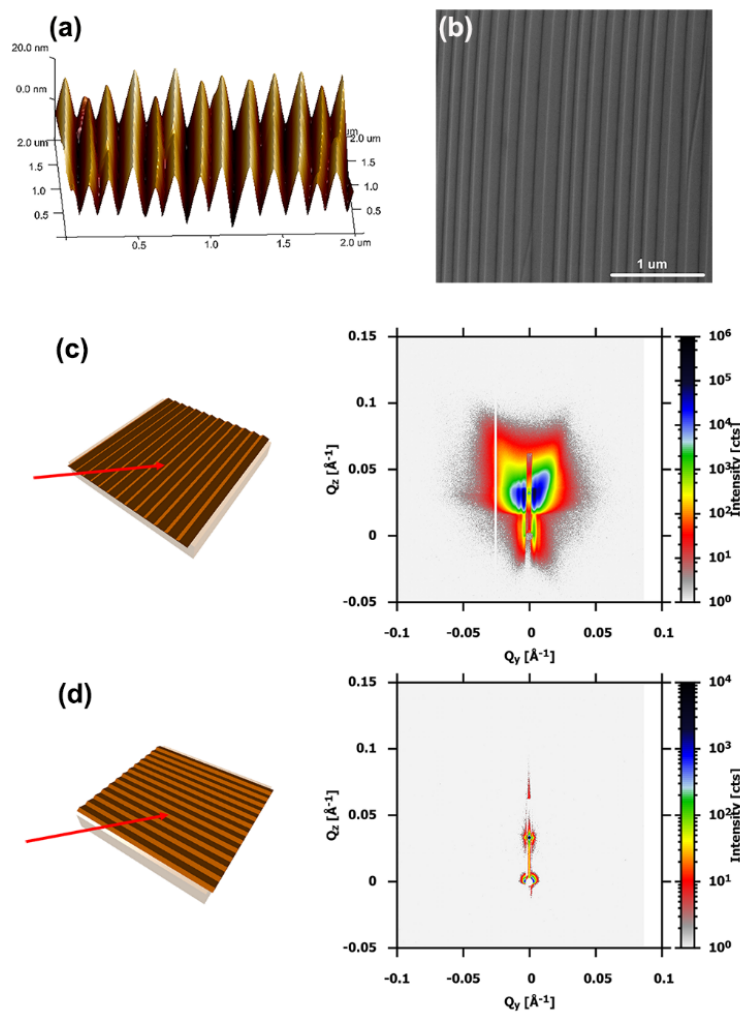


Fig. S2. (a) The 3D AFM image and (b) SEM image of the faceted sapphire substrate annealed at 1500 °C for 24 hours. The scan size for AFM was $2 \times 2 \mu\text{m}^2$. GISAXS scattering intensity collected with the incident beam (c) parallel and (d) perpendicular to the saw-tooth patterns at 0.2° angle of incidence.

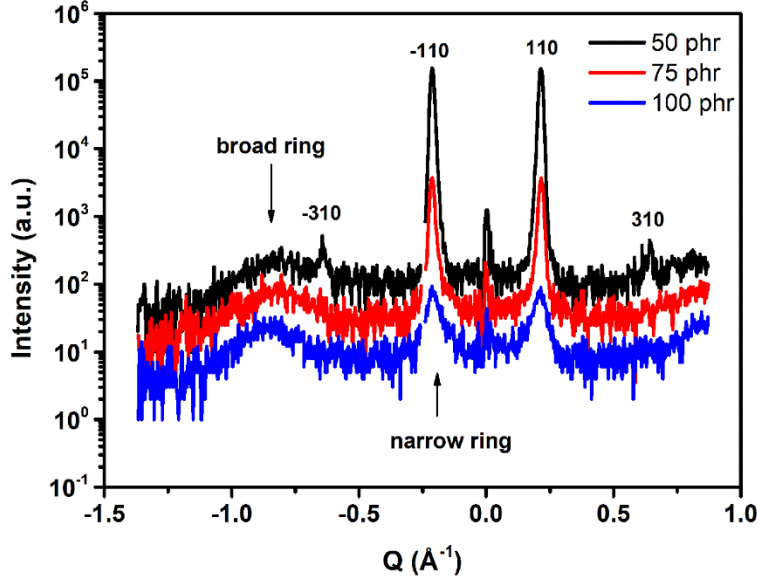


Fig. S3. The linecuts along Qy direction at $Q_z = -0.015 \text{ \AA}^{-1}$, crossed (110) and (-110) Bragg spots, for GTSAXS patterns of 50, 75 and 100 phr.

Distortion factor is defined as the area ratio between different incorporated NPs concentrations to the reference point. We take the area at 5 phr as the reference point.

$$\text{Distortion factor} = A_x/A_{5 \text{ phr}} \quad (1.1)$$

$$A_x = a_x * b_x \quad (1.2)$$

where x is the concentration of NPs, a_x and b_x are the corresponding lattice parameters. As two lattice parameters are taken into account, the distortion factor can illustrate the change of unit cell effectively. The increasing of the distortion factor means the expanding of the unit cell.

The correlation length (ξ) is obtained by fitting the first-order Bragg spot (110) and first-order Bragg peak in GISAXS along Qy direction with Voigt function. The Voigt function is a

probability distribution given by a convolution of Lorentz and Gaussian distribution functions. Gaussian distribution function is used to describe the primary beam. The correlation length is calculated by $\xi = 2\pi/FWHM$, where FWHM is full width at half maximum of Voigt function. During the peak fitting, w_G (the full width at half maximum Gaussian distribution function) is fixed at 0.00125 \AA^{-1} which is the instrumental resolution value.

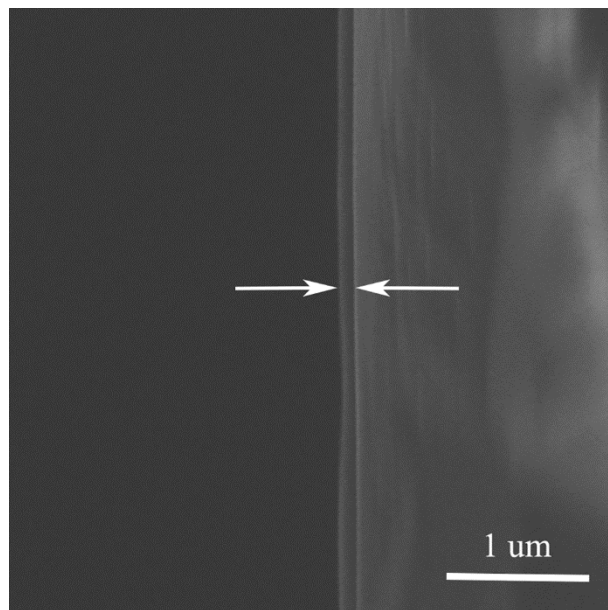


Fig. S4. Cross-section SEM image of the sample. The film thickness is pointed out with arrows.