

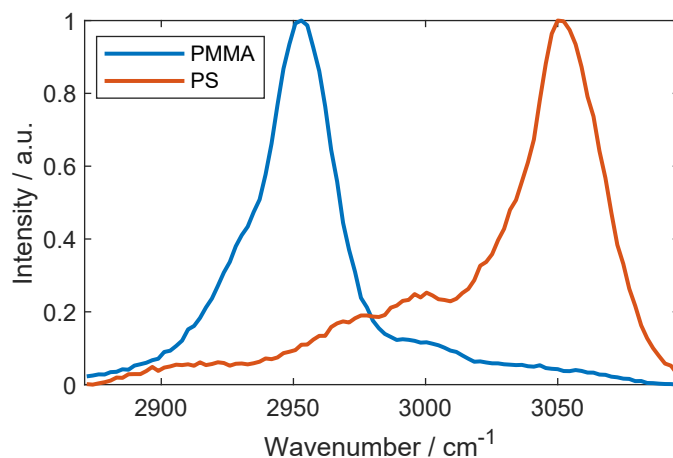
## Supplementary Information

### Nanoscale Bond-selective Imaging by Computational Fusion of Atomic Force Microscopy and Coherent anti-Stokes Raman Scattering Microscopy

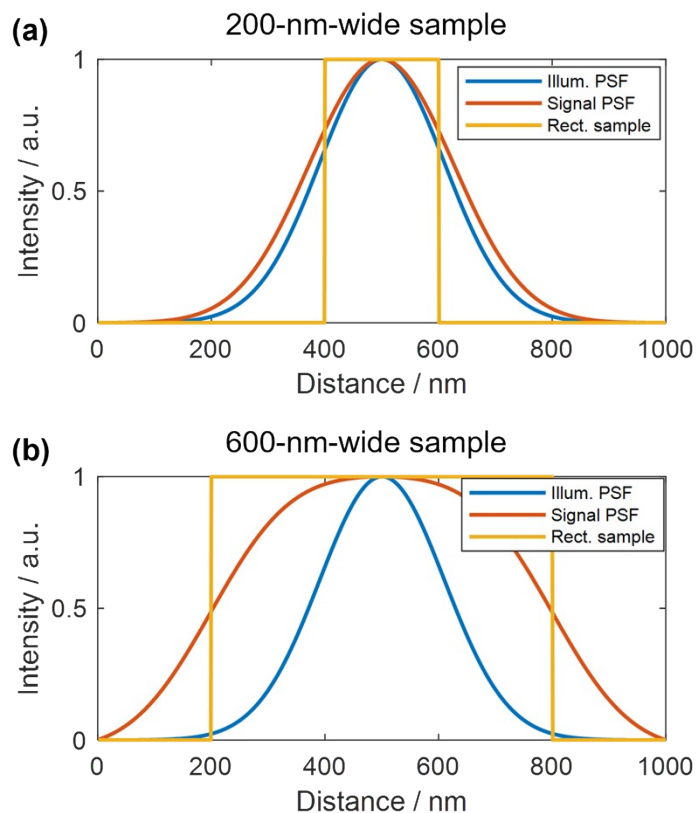
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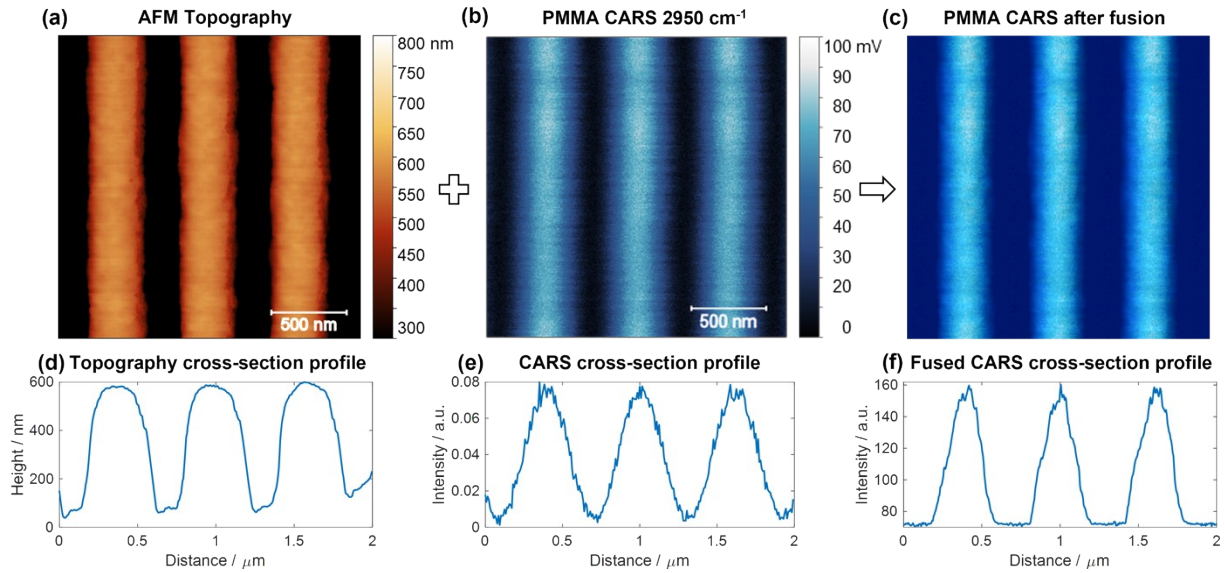
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**Figure S1.** CARS spectra collected on PMMA and PS film. The peaks of 2950 cm<sup>-1</sup> for PMMA and 3050 cm<sup>-1</sup> for PS were used as a guide for the frequency selection of the CARS images in Figure 3. The wavelengths of the pump and Stokes beams were 793 nm and 1040nm in this measurement.



**Figure S2.** Simulated point spread functions (PSFs) of the CARS illumination laser fields and CARS signal on a cuboid PMMA striped nanostructure. (a) The width of each striped structure is 200 nm. The incident excitation field is a Gaussian profile, and the convolution with a 200-nm rectangular structure yields a Gaussian-shaped PSF with a broader full-width-half-maximum (FWHM). (b) One example of a 600-nm-wide cuboid structure is included as a comparison. The convolved CARS signal PSF shows a relatively flat top with a broad falling edge.



**Figure S3.** One demonstration of the distorted fusing results caused by inappropriate parameter selection for the AFM topography image. As shown in (a), the color scale starts from 300 nm, which leads to an information loss of the stripe edges during the Intensity recalculation process. The resulting fused image only has partial morphological features transferred and therefore shows a narrow and sharp shape of each PMMA striped structure.