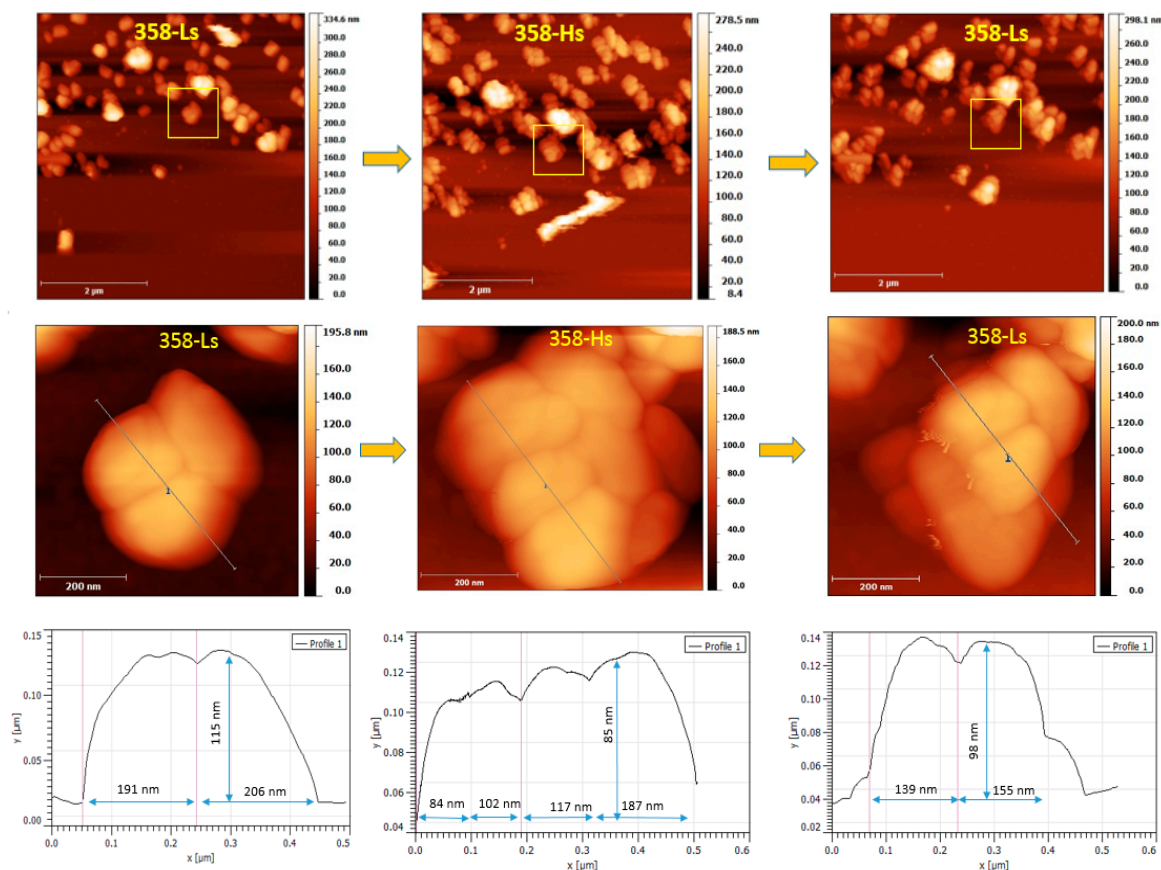
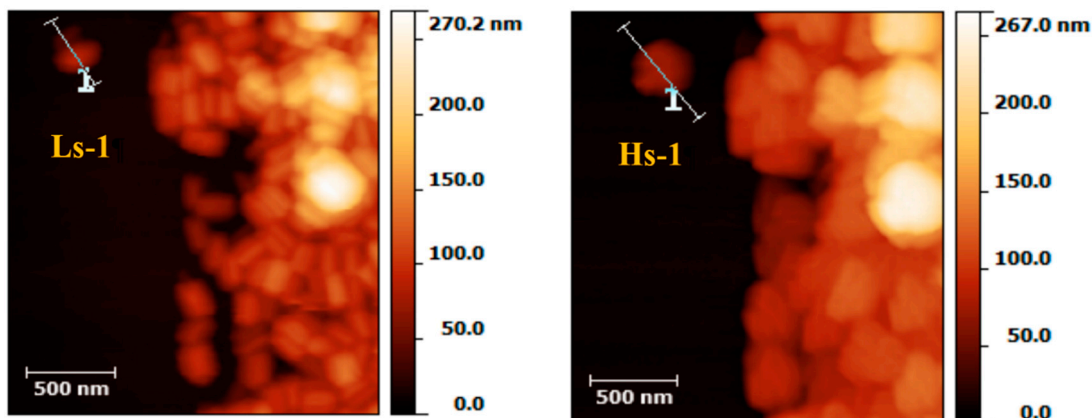


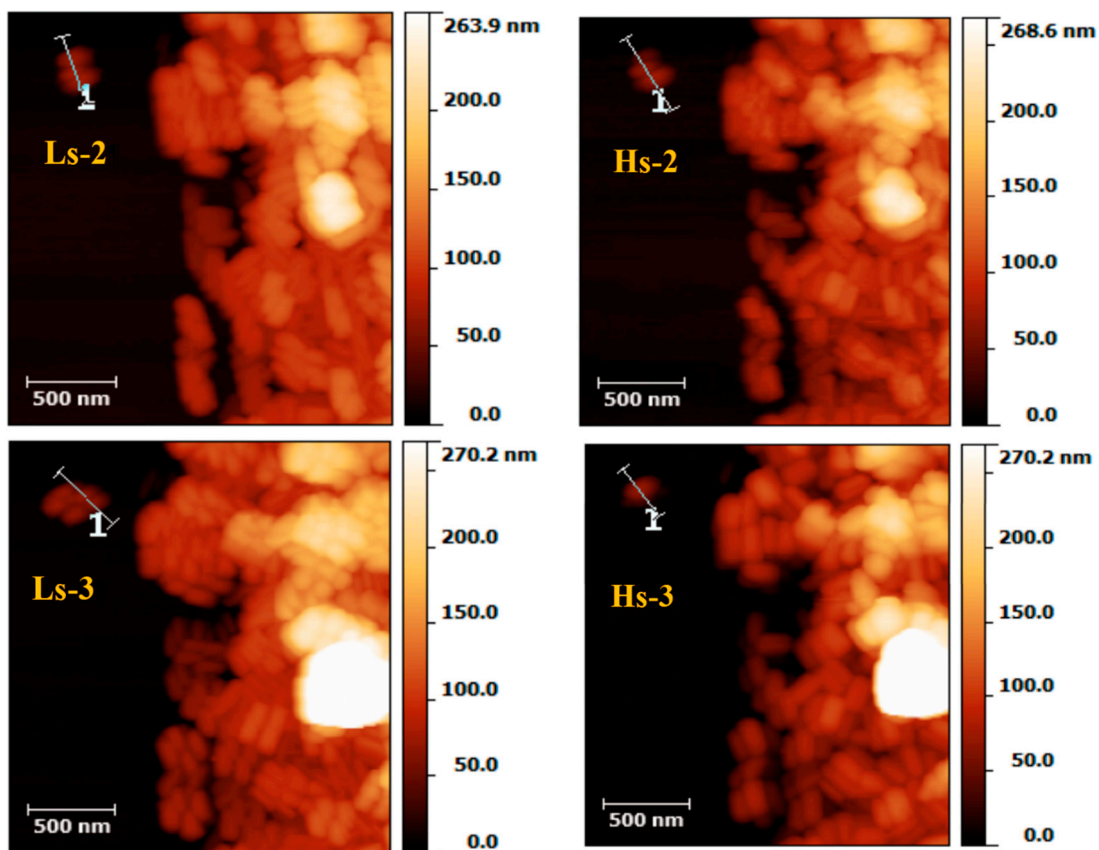
# Supplementary Materials: *In-situ* AFM Imaging of Microstructural Changes Associated with the Spin Transition in [Fe(Htrz)<sub>2</sub>(Trz)](BF<sub>4</sub>) Nanoparticles

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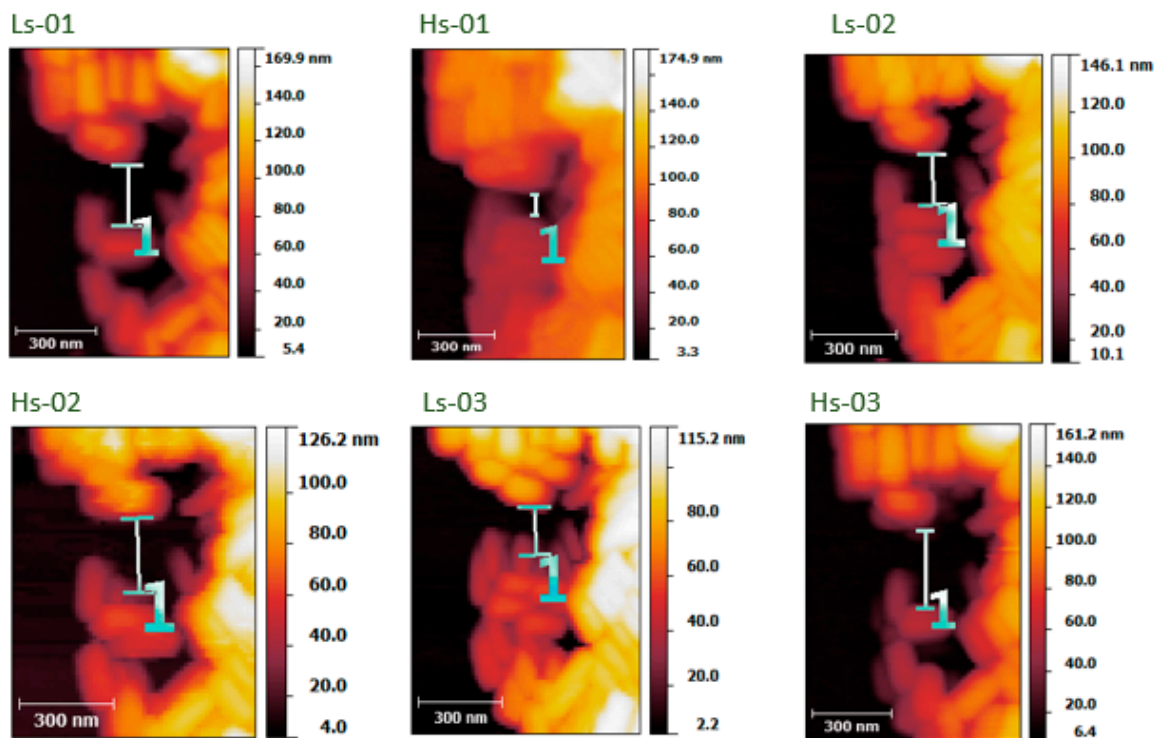


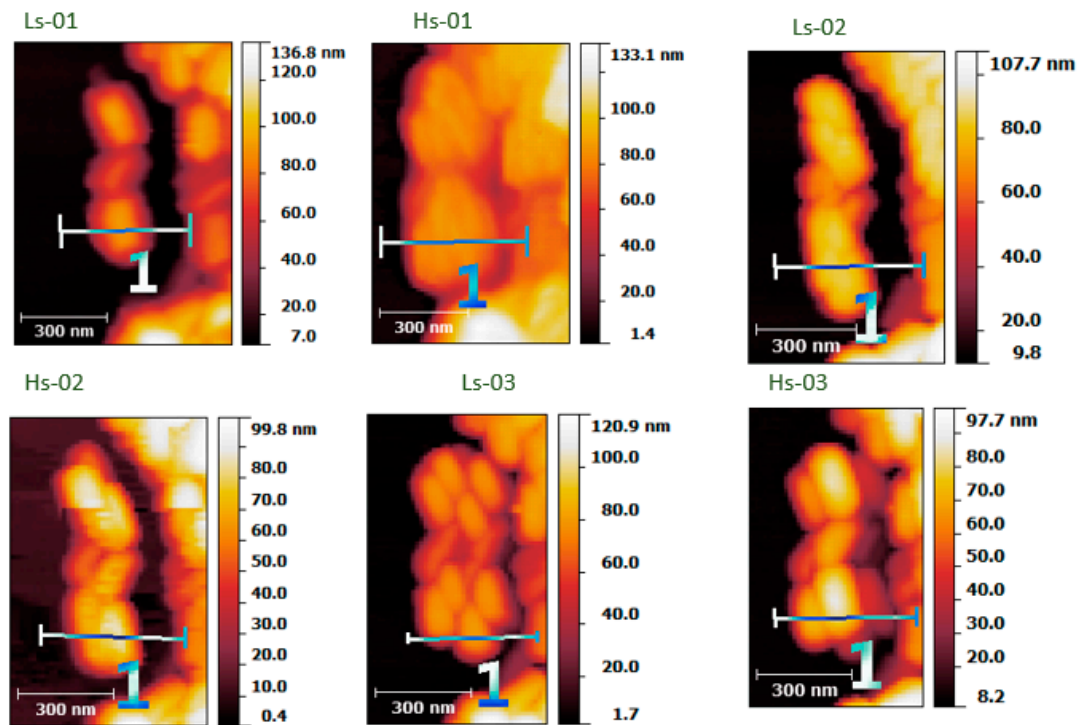
**Figure S1.** AFM height images and cross-sections of [Fe(trz)(Htrz)<sub>2</sub>](BF<sub>4</sub>) nanoparticles acquired in different spin states at 358 K over a complete thermal cycle. (These data were extracted from the same experiment as Fig. 1 in the main text.) Images were acquired using a Cypher ES (Oxford Instruments) AFM and OMCLAC160TS-R3 probes (Olympus,  $f = 300$  kHz,  $k = 26$  N/m, Al reflex coating).



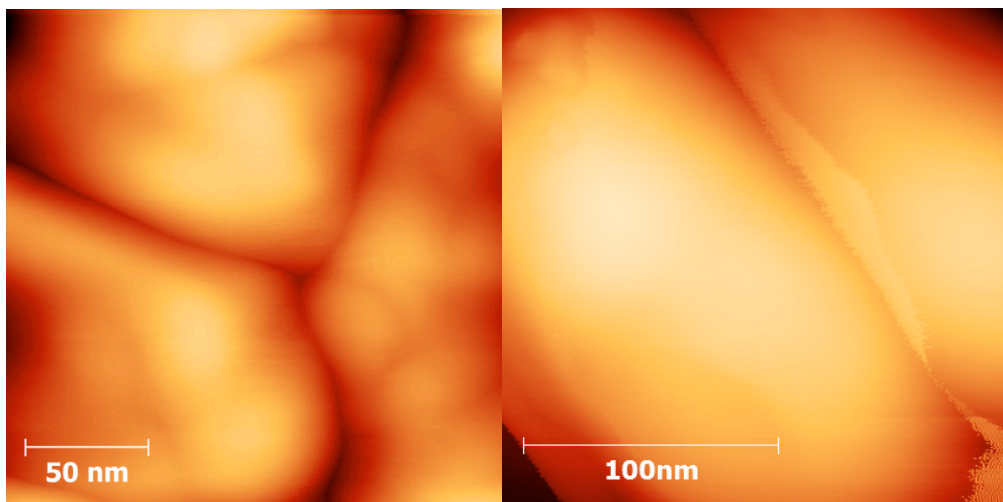


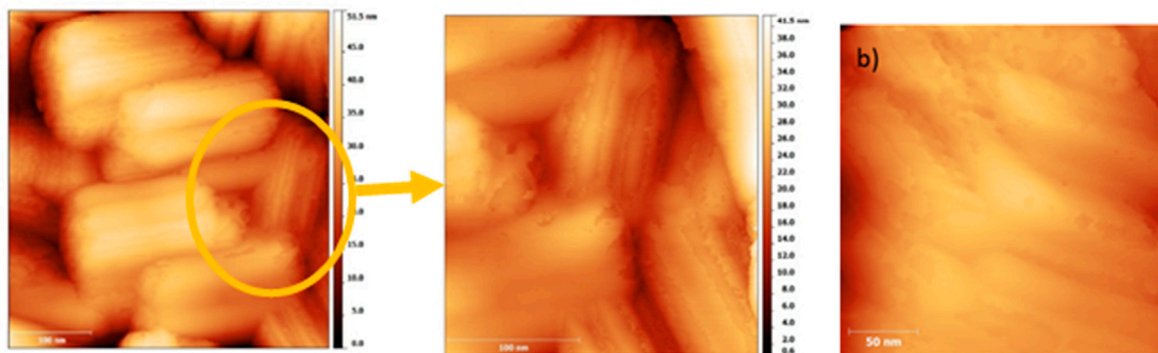
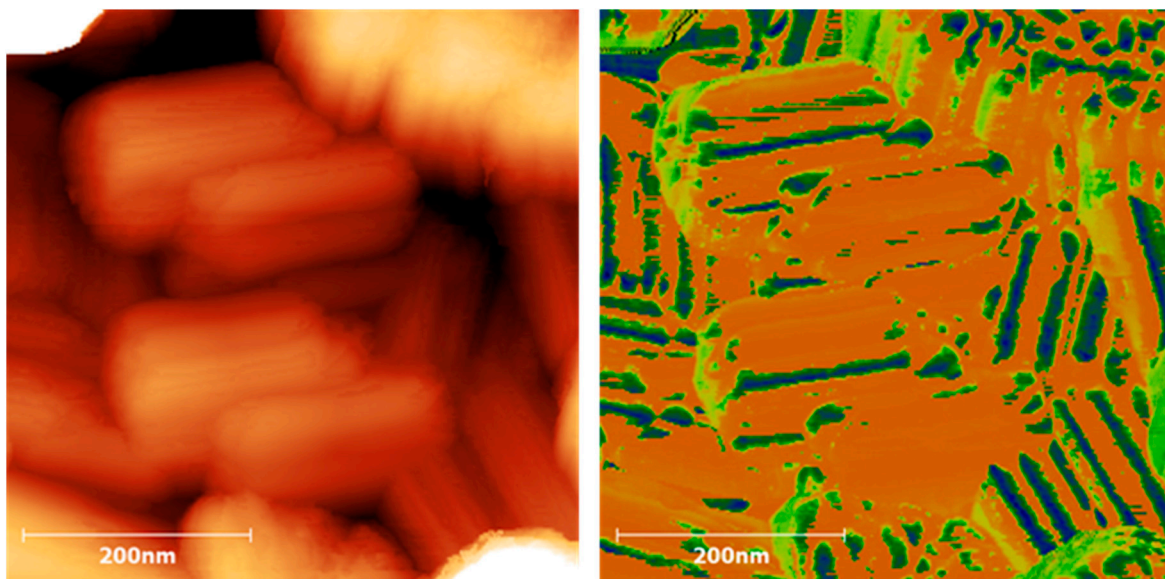
**Figure S2.** AFM height images of  $[\text{Fe}(\text{trz})(\text{Htrz})_2](\text{BF}_4)$  nanoparticles in the LS (left) and HS (right) states over three successive thermal cycles. Images were acquired using a Dimension Icon (Bruker) instrument and MPP-11120-10 probes (Bruker,  $f = 300 \text{ kHz}$ ,  $k = 40 \text{ N/m}$ , Al reflex coating).





**Figure S3.** AFM height images of [Fe(trz)(Htrz)<sub>2</sub>](BF<sub>4</sub>) nanoparticles in the LS and HS states over three successive thermal cycles. (These data were extracted from the same experiment as Figure S2.)





**Figure S4.** Surface degradation of  $[\text{Fe}(\text{trz})(\text{Htrz})_2](\text{BF}_4)$  particles following 8 thermal switching cycles: topography and phase images. The first two images were acquired on the fresh sample. Data were acquired using a Cypher ES (Oxford Instruments) AFM and ARROW-UHF-AuD (Nanoworld,  $f = 2$  MHz,  $k = 6$  N/m, Au reflex coating).

