Supplementary Information:

**Generation of clean iron nanocrystals on an ultra-thin SiO\textsubscript{x} film on Si(001)**

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**Fig. 1** Local Auger electron spectra acquired between the iron crystals on the ultra-thin SiO\textsubscript{x} film on Si(001) (red) and on an iron structure (blue) fabricated by local electron beam activation (60 nC) and an additional Fe(CO)\textsubscript{5} dosage time of 4 h 30 min (background pressure Fe(CO)\textsubscript{5} 3.0 \times 10^{-7} \text{ mbar}). The blue spectrum acquired on the Fe deposit is clearly dominated by the Fe\textsubscript{L\text{MM}} peaks, with only minor C\textsubscript{K\text{LL}} and O\textsubscript{K\text{LL}} peaks being visible. Considering the high surface sensitivity of AES and the waiting time of 9 days between the deposition experiment and the AES measurement, the traces of oxygen and carbon can be associated with adsorbed species from the residual gas, and thus represent the upper contamination level of the produced deposit, i.e., the bulk of the deposit presumably consists of pure iron. The red spectrum acquired between the iron crystals on the unirradiated ultra-thin SiO\textsubscript{x} film on Si(001) shows solely the O\textsubscript{K\text{LL}} transition; no carbon or iron are detected between the iron crystals, i.e., that deposition of the material is restricted to the active sites and the whole process is perfectly selective.
Fig. 2 (a) SEM image of iron deposit fabricated by local electron beam activation (240 nC) and additional Fe(CO)₅ dosage for 4h 30 min (background pressure Fe(CO)₅ 3.0 x 10⁻⁷ mbar). (b) Simulation of the BSE exit area on SiOₓ (0.5 nm) / Si(001) using the program Casino V2.42. The diameter of the BSE exit area was estimated to a maximum value of ~ 4.6 µm which corresponds to an area of which 99.99 % of the BSEs are emitted. This is in very good agreement with the experimental findings for high electron doses.