

Supporting Information: Anisotropy Control in Magnetic Nanostructures through Field-assisted Chemical Vapor Deposition

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Experimental Details

Substrate preparation

Previously cut (5 mm x 5 mm) silicon substrates (100) were purchased from CrysTec[®]. Possible organic residues were removed using 10 minutes of ultrasonication in *iso*-propanol.

FTO substrates (Sigma Aldrich) were cut in 10 mm x 15 mm and rinsed using a specific cleaning protocol. In detail, the substrates were subsequently ultrasonicated for 10 minutes in a Hellmanex[®] solution, deionized water, acetone and *iso*-propanol. In between each washing step, the samples were previously rinsed with the upcoming solvent.

Iron film deposition

For all processes, 0.20(1) mL of Fe(CO)₅ were transferred into a custom-made precursor reservoir, connected to the setup and stored under inert conditions until deposition. Afterwards, the substrate was fixed to the heater by silver paste and the chamber was evacuated after drying of the paste. Subsequently, the sample was heated to 300(5) °C and the reservoir was opened. In case of a field assisted process, the magnetic field was applied before opening the precursor reservoir. The pressure was adjusted to 0.40(3) mbar. After 3 minutes, the reservoir was closed and the samples were cooled to room temperature in vacuum without a specific cooling rate. In case of field assisted processing, the magnetic field was applied until the samples reached 200 °C and subsequently switched off. All samples were stored at air.

Oxidation of anisotropically grown iron nanostructures on FTO

The samples were annealed for 2 h at 750 °C under ambient conditions with a specific heating rate of 750 °C/h. Afterwards, the samples were cooled to room temperature without a specific cooling rate.

Materials Characterization

The powder X-ray diffraction (XRD, STOE-STADI MP) patterns were measured in reflection mode using Mo K α ($\lambda = 0.07094$ nm) radiation. Although no additional signals were detected within the resolution range of the experiment, EDX analysis revealed the presence of oxygen in the sample. This indicates the formation of a non-stoichiometric amorphous passivation layer, initialized by the exposure of the sample to air. The surface morphologies of the specimens were characterized using a Zeiss Leo Supra-35 scanning electron microscope, operated at 5kV and a FEI Magellan 400, operated at 3 kV. Relative compositions were analyzed using an Oxford Xmax 80 EDS-system mounted on a FEI Nova NanoSEM 650 operated at various acceleration voltages. Hematite EDX spectrum was simulated using *hyperspy*.¹ TEM specimen characterization was performed on a FEI Tecnai G2 Spirit at 120 kV. Isothermal magnetization measurements were performed using PPMS-VSM (Quantum Design PPMS-14), at room temperature, under applied magnetic field up to 3T. The measurements were done in the perpendicular and parallel directions in respect to the applied field used during the synthesis. Demagnetization factor was not taken into consideration for these measurements. Electrochemical measurements were performed in a flat three-electrode electrochemical cell using a saturated calomel electrode (SCE) as the reference, a Pt wire as the counter electrode and 1 M NaOH as the electrolyte (pH = 13.6). Linear sweep voltammetry (10 mV/s) was carried out in a potential range from -1 to 1 V vs. SCE using a potentiostat (PAR, Versa state IV) in the dark and under simulated solar illumination (Xe lamp, 150 W, Oriel with a Schott KG-3 filter). Potentials with respect to the reversible hydrogen electrode (RHE) scale were calculated using the Nernst equation (1).

$$E_{RHE} = E_{SCE} + E_{SCE}^0 + 0.059pH \quad (1)$$

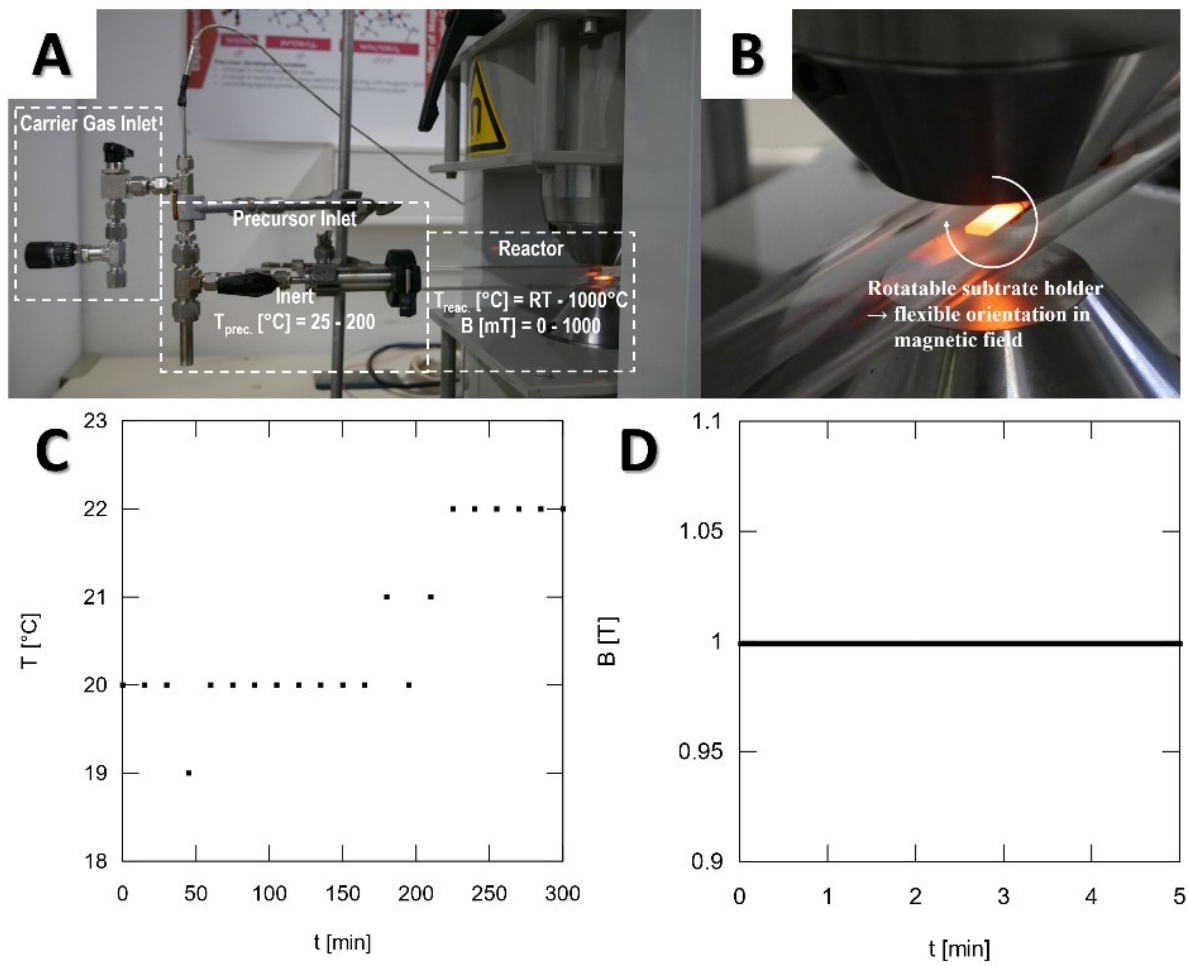


Figure S1: **A** and **B** illustrate the used setup, consisting of an electromagnet, rotatable heater, and precursor inlet. The time dependent thermal radiation induced heating and magnetic field stability are visible in **C** and **D**, respectively.

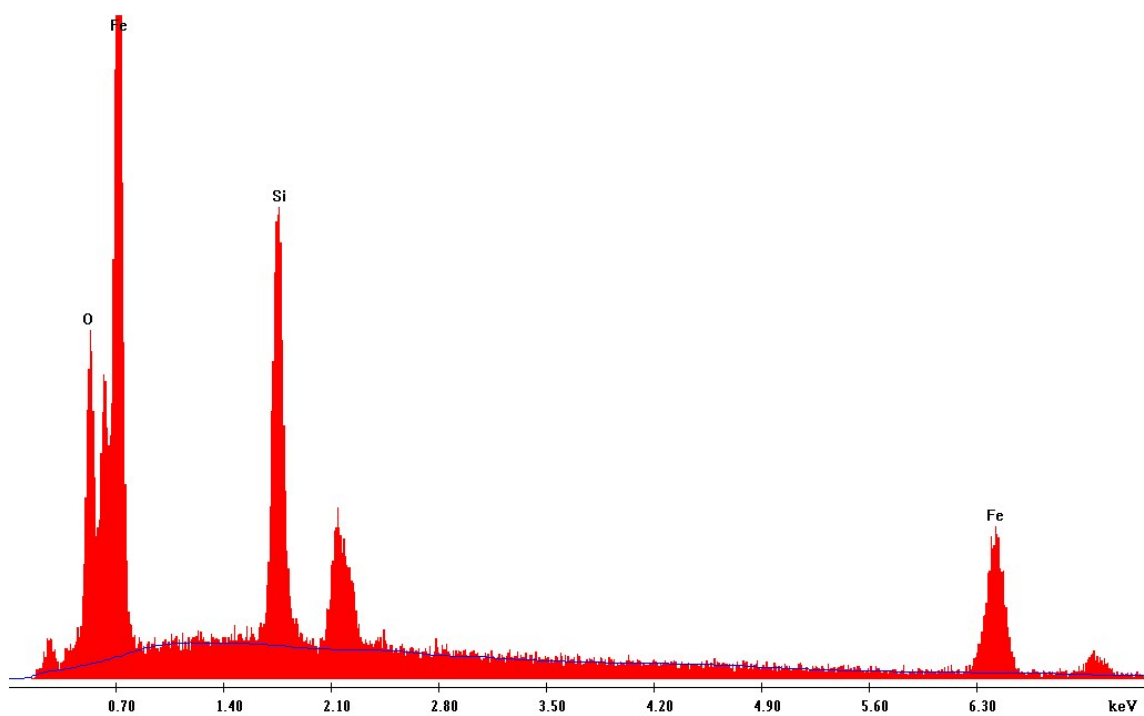


Figure S2: EDX analysis of an iron film deposited in 1 T magnetic field flux density.

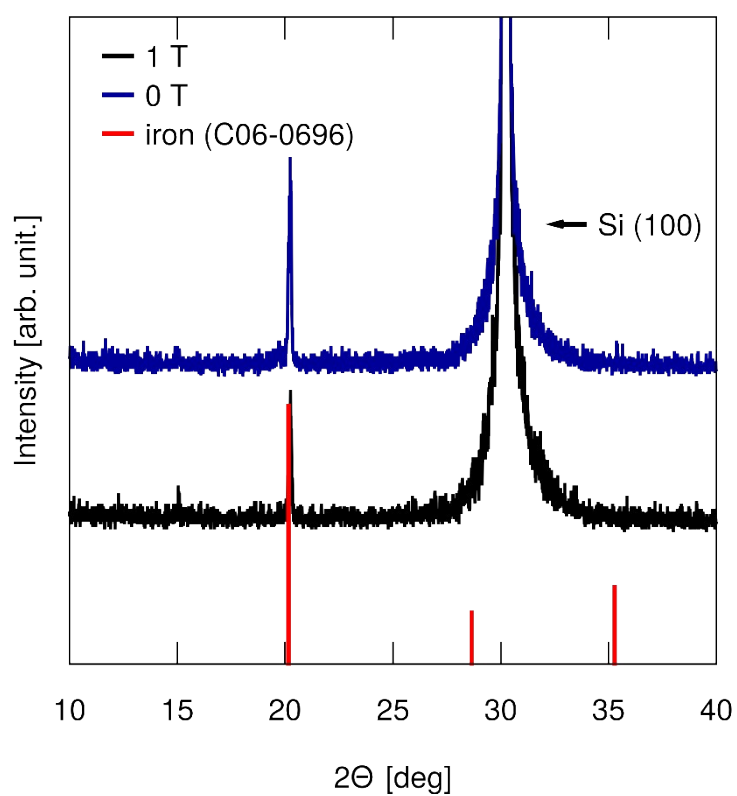


Figure S3: XRD analysis of deposited iron films. Exclusively, signals for iron and the substrate were detected.

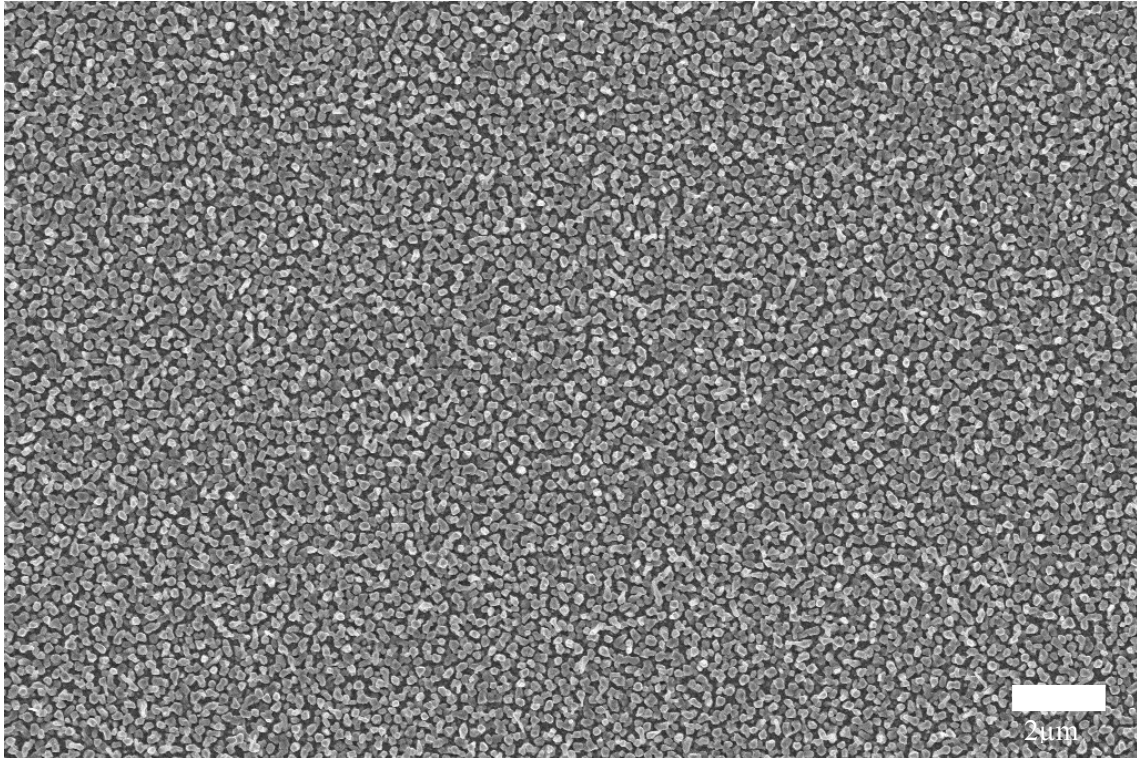


Figure S4: SEM micrograph demonstrating the homogeneity of deposited anisotropic structure formation for 1.00 T field flux density

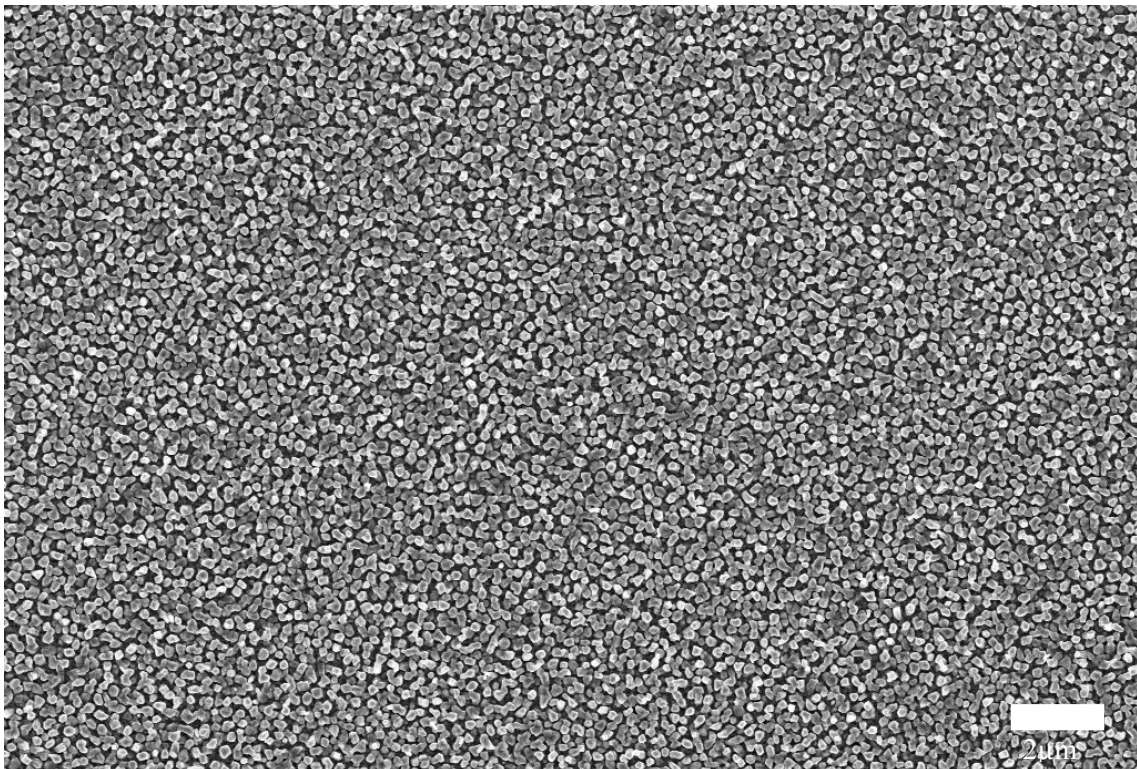


Figure S5: SEM micrograph demonstrating the homogeneity of deposited anisotropic structure formation for 0.50 T field flux density

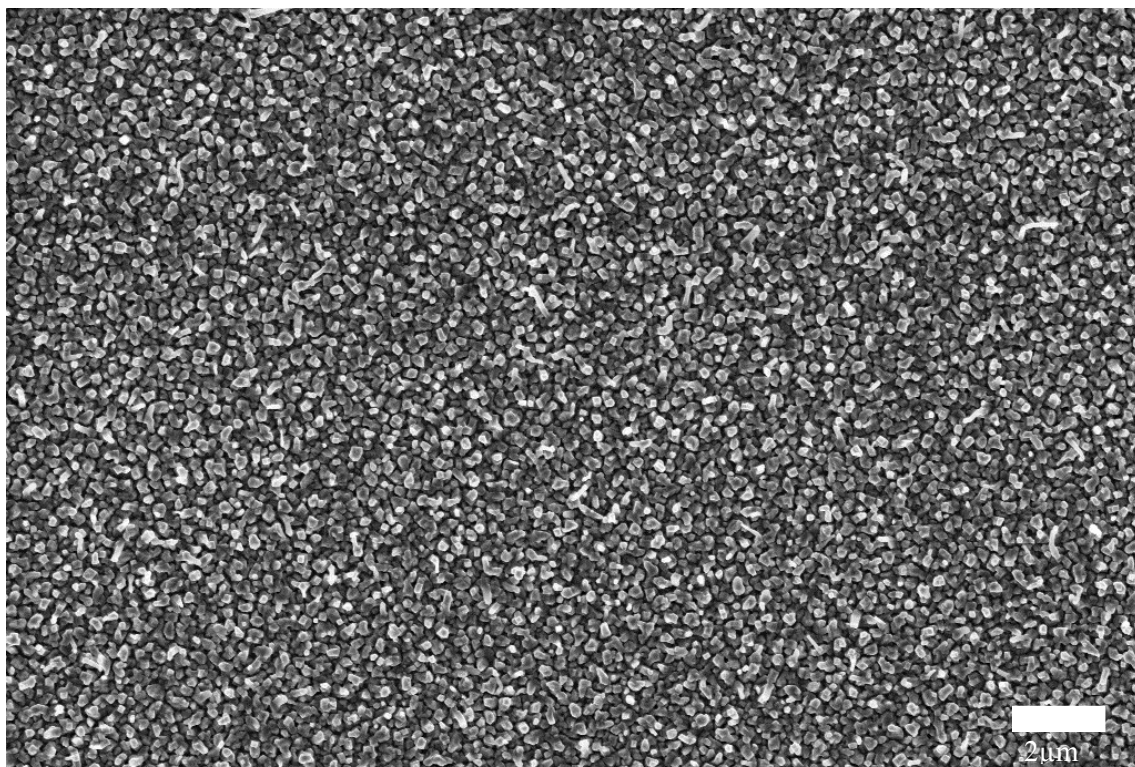


Figure S6: SEM micrograph demonstrating the homogeneity of deposited anisotropic structure formation for 0.25 T field flux density

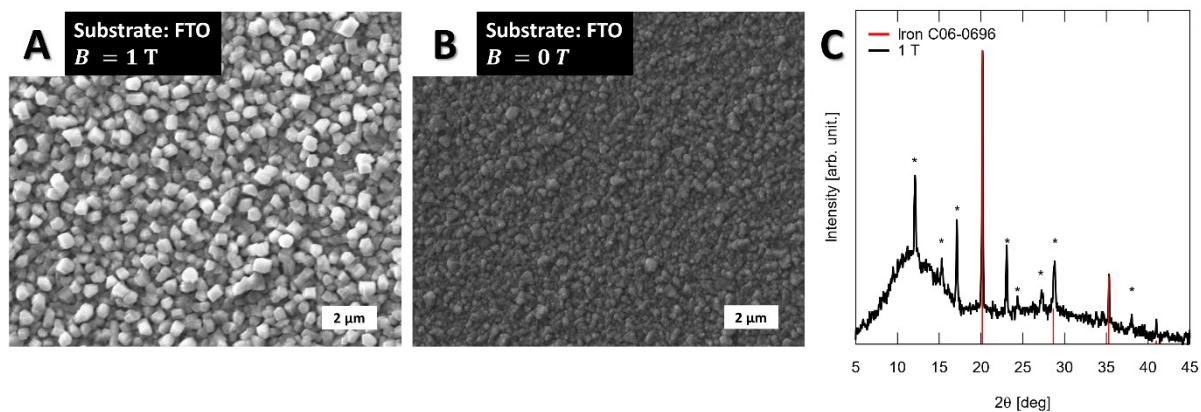


Figure S7: SEM micrographs of iron film formation on FTO in 1.00 T (A) and 0.50 T (B) field flux density. A representative XRD demonstrates the phase pure formation of iron films (FTO substrate reflections are labelled as '*').

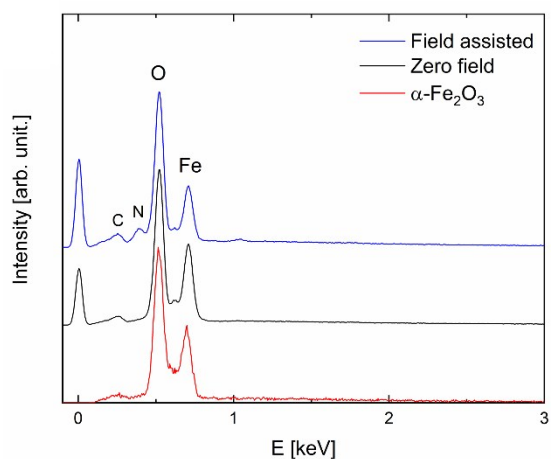


Figure S8: EDX spectra obtained with 5 kV excitation voltage.

References

1. F. de la Peña, T. Ostasevicius, V. T. Fauske, P. Burdet, P. Jokubauskas, M. Sarahan, D. Johnstone, M. Nord, J. Taillon, J. Caron, K. E. MacArthur, A. Eljarrat, S. Mazzucco, T. Furnival, E. Prestat, M. Walls, G. Donval, B. Martineau, L. F. Zagonel, A. Garmannslund, T. Aarholt, C. Gohlke, I. Iyengar, *hyperspy: HyperSpy 1.0.1* 2016, <http://dx.doi.org/10.5281/zenodo.58841>.