Supporting Information:

Laser-Rewriteable Ferromagnetism at Thin Film Surfaces

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1. Transmission Electron Microscopy

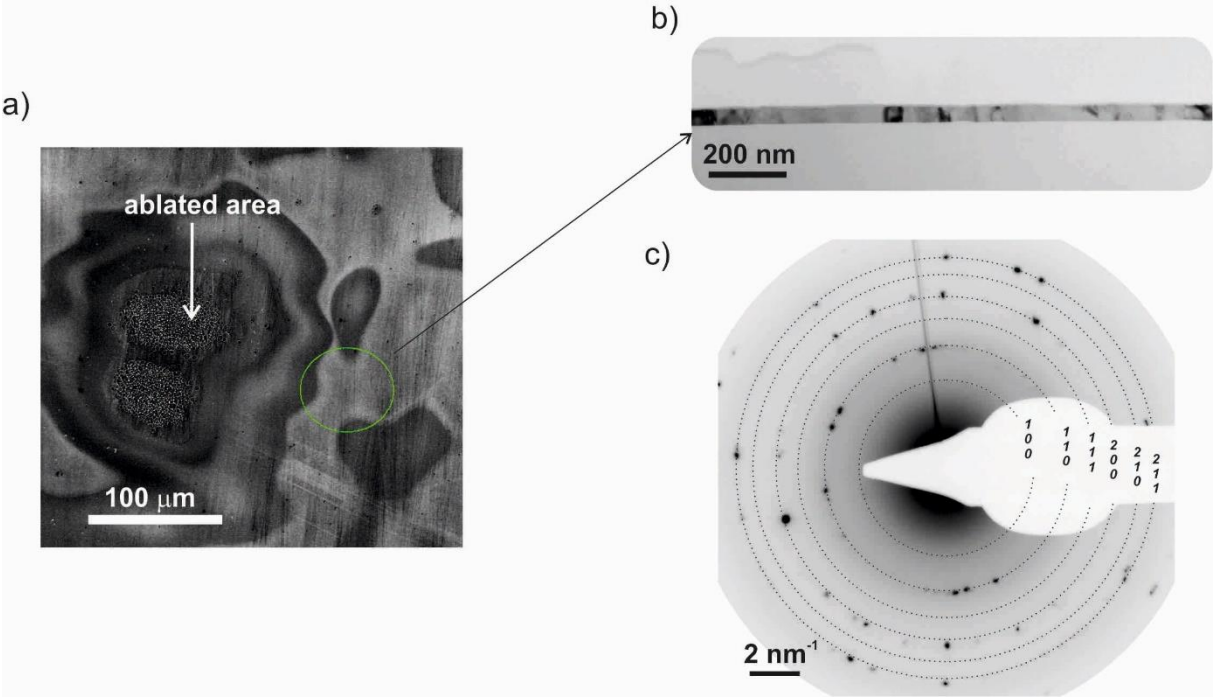

Figure S1: Cross-sectional TEM preparation and analysis of laser-treated $Fe_{60}Al_{40}$ deposited on oxidized silicon. a) A TEM lamella was extracted from a region known to be ferromagnetic and close to the ablation zone. The lamella was prepared by *in situ* lift-out using a Focused Ion Beam device. **b)** Bright-field TEM imaging shows an intact 40 nm thick polycrystalline layer. **c)** Selected-area electron diffraction (SAED) confirms its microstructure to be A2 $Fe_{60}Al_{40}$ *via* the absence of 100, 111, and 210 superstructure reflections. See Ref. 18 for an SAED pattern of a B2 $Fe_{60}Al_{40}$ film for comparison.

Experimental Details:

TEM lamella preparation of the laser-irradiated $Fe_{60}Al_{40}$ film was done by in situ lift-out using a Zeiss Crossbeam NVision 40 system. A protective cap layer was deposited beginning with electron beam assisted carbon-based precursor decomposition and subsequently followed by Ga focused ion beam (FIB) assisted Pt-based precursor decomposition. Afterwards, the TEM lamella was prepared using a 30 keV Ga FIB with adapted currents. The lamella was transferred to a 3 post copper lift-out grid (Omniprobe) using a Kleindiek micromanipulator. To minimize sidewall damage, Ga^+ ions with an energy limited to 5 keV were used for the final thinning of the TEM lamella to electron transparency. TEM investigations were performed using an image C_s-corrected Titan 80-300 microscope (FEI). Besides TEM bright-field imaging, selected-area electron diffraction was done to analyse the microstructure of the laser-treated $Fe_{60}Al_{40}$ film. Prior to TEM analysis, the specimen mounted in a double-tilt analytical holder was placed for 10 s into a Model 1020 Plasma Cleaner (Fischione) to remove organic contamination.

2. Spatial distribution of X^2

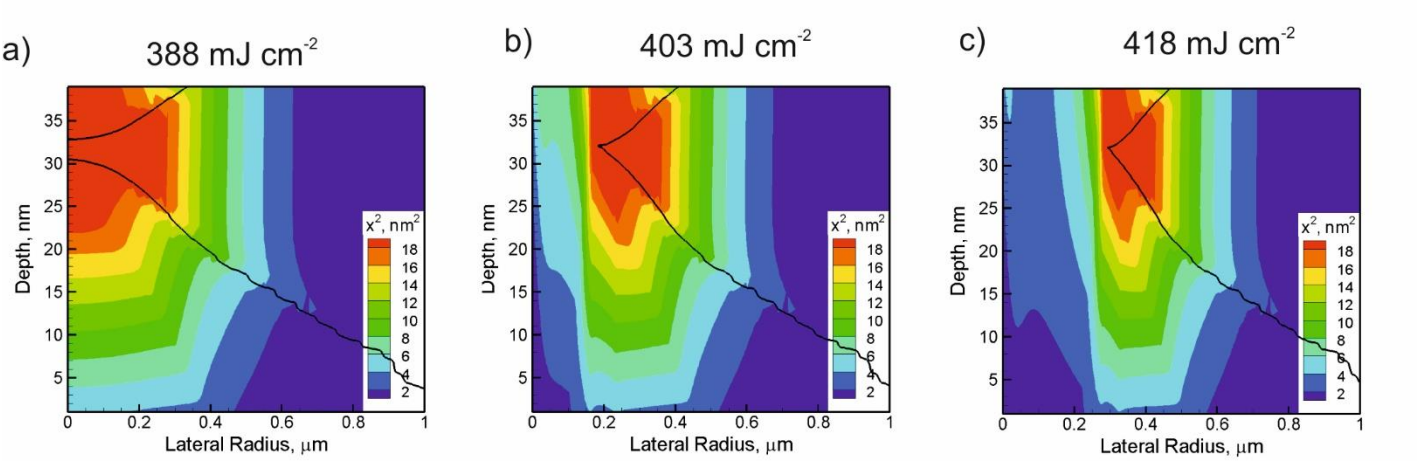

Figure S2: Spatial variation of X^2. The spatial variations have been calculated for single pulse laser irradiation with fluences of **a)** 388 **b)** 403 and **c)** 418 mJ cm^{-2}. The black line shows the extent of the melting fronts during the process.