Soft magnetism in single phase Fe_3Si thin films deposited on $SrTiO_3(001)$ by pulsed laser deposition – Supplementary information

S1. RSM and rocking scans in Fe_3Si films

Reciprocal space maps across Fe_3Si(202) and STO(111) reflections as well as rocking scans are shown in Fig. S2. Fe_3Si(202) reflections for the samples prepared in a temperature range between 200 ºC and 400 ºC show a broad profile, related with a high degree of mosaicity in the deposited samples, which is also indicated by the broad and irregular lineshape of the rocking scans. Nevertheless, a much larger coherence of the Fe_3Si crystallographic domains is obtained for a deposition temperature of 600 ºC, observing a well-defined Lorentzian shape of the rocking scan. Nevertheless, this is accompanied by the formation of Fe phases and much more particulate film.

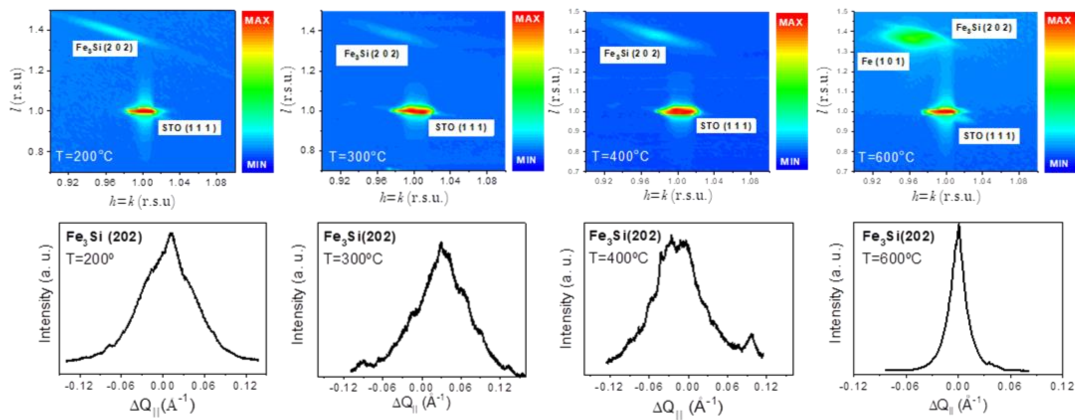

Fig. S1. (Top) Reciprocal space maps across STO(111) and Fe_3Si(202) reflections. (Bottom) Rocking scans around Fe_3Si(202) reflection.

In plane and out of plane lattice parameters are collected in table S1. Fe3Si grows fully relaxed regardless of the substrate temperature during the deposition, and variations between in plane and out of plane values are inside the experimental error, indicating a cubic symmetry.

Table S1: Calculated lattice constants for Fe3Si films deposited at different temperature

T_s	In plane (a)	Out of plane(c)
200C	(0.569± 0.003) nm	(0.566 ± 0.004) nm
300C	(0.567 ± 0.003) nm	(0.568 ± 0.004) nm
400C	(0.566 ± 0.003) nm	(0.568 ± 0.004) nm
600C	(0.566 ± 0.003) nm	(0.566 ± 0.004) nm

S2. In-situ RHEED characterization and ex-situ XRR

The crystalline growth of the Fe_3Si was *in-situ* monitored by reflected high energy electron diffraction (RHEED). The electron beam was oriented along the STO [110] crystallographic axis (Fig. S1.a). When comparing samples deposited at 200ºC and 400ºC we can see a transition from a stripe like pattern, characteristic of a planar growth, to a dot pattern with faint stripes, related with a transition to an island-like growth, as later demonstrated by HR-TEM.. At 600 ºC the apparition of poly-crystal intensity rings can be observed in the last cycles of deposition. Nevertheless, XRD does not show a significant presence of polycrystalline phase.

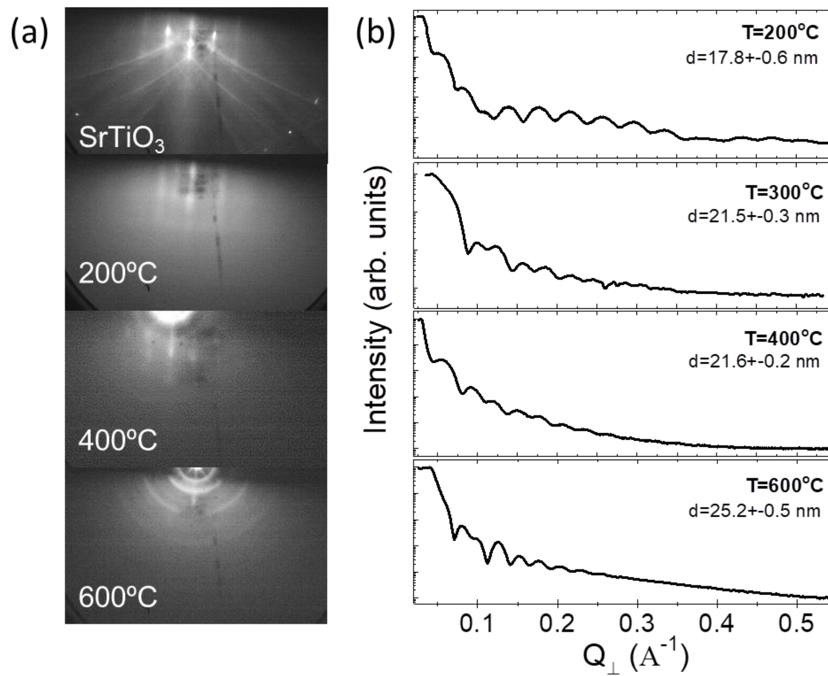

Fig. S2. (a) RHEED diffraction pattern recorded for (from top to bottom) as-inserted STO(001) substrate, 43 cycles of Fe_3Si deposition at 200 ºC, 400 ºC and 600 ºC. (b) XRR measurements for samples deposited at 200 ºC, 300 ºC, 400 ºC and 600 ºC.

X-ray reflectivity (XRR) measurements show the presence of Kiessig fringes for all deposited films. Given the strong dependence of this interference phenomena with surface roughness, the presence of intensity oscillations is indicative of flat interfaces. A faster decay of the oscillations with incident angle is observed as the growth temperature is increased. This evolution is more likely to be related with an increase of surface and

interface roughness when deposition temperature is increased, which is consistent with a transition from planar to granular growth, as observed by HR-TEM. Fe_3Si thickness was estimated from oscillation analysis obtaining values of 17.8(6), 21.5(3), 21.6(2) and 25.2(5) nm for samples deposited at 200, 300, 400 and 600 ºC, respectively, which is in good agreement with nominal thicknesses.

S3. EDX in Fe_3Si films deposited at 600ºC

In Fig. S3 STEM-HAADF cross-section images of the sample deposited at the highest temperature (600 ºC) are shown. The variations of density along the film indicate the formation of a particulate film. Also, element sensitive analysis indicates an inhomogeneous distribution of Fe and Si. The results indicate a higher concentration of Fe within the clusters meanwhile Si occupies the regions in between, mainly forming oxides. Analysis of different cluster areas show the presence of pure Fe clusters as well as Fe_3Si in agreement with XRD results.

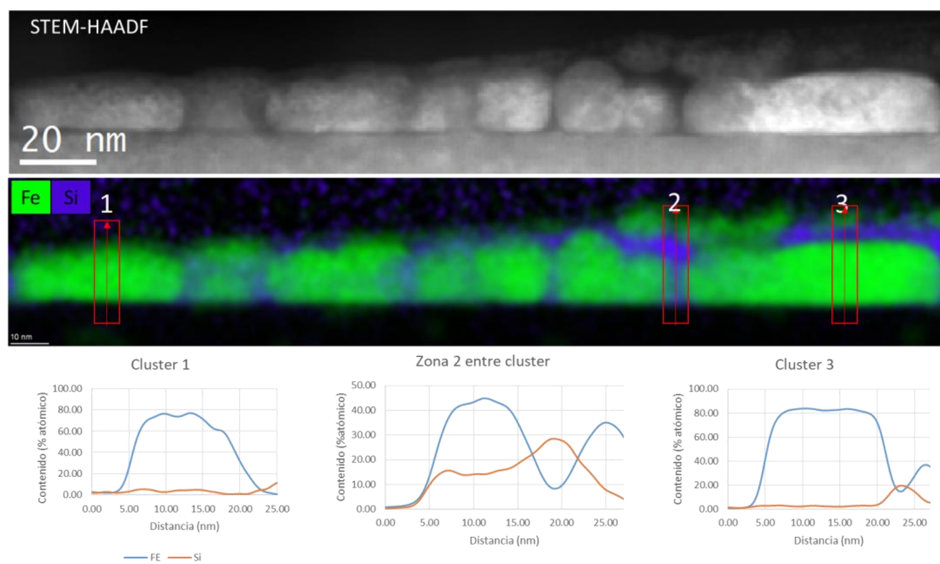

Fig. S3: From top to bottom: STEM-HAADF, element selective EDX images overlapping Fe-K and Si-K and EDX analysis in the framed regions for the sample deposited at 600ºC.