Supplemental Document



## **All-optical spin injection in silicon investigated by element-specific time-resolved Kerr effect: supplement**

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## All-optical spin injection in silicon investigated by element specific time-resolved Kerr effect: supplemental document Section I. Sample synthesis and characterization.

The sample was prepared and characterized under ultra-high vacuum conditions at the endstation of the VUV-Photoemission beamline (Elettra Sincrotrone Trieste). The Si substrate was cut from a p-doped Si wafer (B dopant,  $0.05 \Omega$ cm resistivity) with (111) surface termination. After prolonged thermal annealing at 700 K, the substrate was flash-annealed to 1520 K until a sharp 7×7 surface reconstruction appears in the low-energy electron diffraction (LEED) pattern. The topmost photoemission spectrum in Fig. S1 (black line) refers to this preparation step. The surface was held at a temperature of  $1050$  K and exposed to  $100$  L of NH<sub>3</sub> to produce an ultra-thin layer of crystalline  $Si_3N_4(0001)$ . This reaction is known to be self-limiting at the thickness corresponding to two bilayers  $(< 1$  nm), which display an  $8 \times 8$  surface reconstruction in the LEED pattern [1]. The formation of the  $Si<sub>3</sub>N<sub>4</sub>$  layer is accompanied by the presence of the NIs level and the shifted components in the  $Si2p$  and  $Si2s$  levels (red spectrum in Fig. 1S). Ni was grown on the  $Si_3N_4(0001)$  surface kept at liquid nitrogen temperature, to favor the formation of a continuous film. As shown in Fig. S1 (green spectrum), the Ni film thickness of 7 nm is sufficient to suppress almost completely the signal from the  $Si<sub>3</sub>N<sub>4</sub>(0001)$ . The Ni film displays no LEED pattern. Finally, an Ag film of 2 nm was deposited on Ni, again using liquid nitrogen temperature to obtain a uniform coverage (blue spectrum of Fig. S1). This was particularly important to prevent the oxidation of Ni, as the sample was removed from the growth chamber for the magnetic measurements.



Fig. S1: Photoemission spectra taken at the various stages of the sample growth.

Section II. TEM characterization of the sample.

Details on the film nanostructure were provided by high-resolution transmission electron microscopy (HRTEM) using a JEOL 2010 UHR field emission gun microscope operated at 200 kV with a measured spherical aberration coefficient Cs of  $0.47 \pm 0.01$ . A representative bright-field HRTEM image of the Ni/ $Si_3N_4/Si(111)$  cross-section is shown in Fig. S2. The  $Si_3N_4$  layer is about 0.7 nm-thick and homogenously extends on top of the bare Si (111) substrate with atomically flat and sharp interfaces. In the close proximity of  $Si<sub>3</sub>N<sub>4</sub>$  interface in the semiconductor, a limited interdiffusion of Ni atoms can give rise to a nickel silicide layer with a calculated stochiometry corresponding to NiSi<sub>2</sub>. The silicide layer was not uniform in all the probed regions of the sample, but its thickness was always observed to be below 3 nm.



Fig. S2: Cross-sectional HRTEM image of Ni/ $Si<sub>3</sub>N<sub>4</sub>/Si$  (111) nanostructure.

## Section III. Kerr rotation calculation

Fig. S3 displays the calculated Kerr rotation for bulk Ni sample.

Calculation procedure: First, the real components of the dielectric tensor for Ni have been calculated with DFT package WIEN2k [2] and then extended to the imaginary parts using the Kramers-Kronig relations, making sure that they converged to zero at higher energies. Then using the magneto-optical medium boundary matrix formation [3, 4], the Kerr rotation in longitudinal geometry at 45° angle of incidence was calculated. In the Figure S3, we have highlighted the Ni  $M_{2,3}$  (red) and Si  $L_{2,3}$  (blue) edge. The calculations further support the fact that no magneto-optical effect arising from Ni M-edge is present at the Si L-edge.



Fig. S3: Kerr rotation for a bulk Ni sample.

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