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Supplementary Figure 1: (a) Variation of Au thickness with the sputtering deposition time and (b) XRD pattern of 10 nm Au nano-particles. *Asterisk* in the XRD patterns denotes the (*111*) peak position of Au.



Supplementary Figure 2: Surface roughness profiles of (a) 4 nm, (b) 10 nm and (c) 20 nm Au thin films obtained from AFM images. Inset shows the values of average (Ra). and root mean square (Rq) roughness.



Supplementary Figure 3: (a) cross-sectional TEM image of CFO deposited at room temperature on 2.5 nm Au nano-dots and (b) CFO deposited on 10 nm Au underlayer. (c) and (d) are the corresponding magnetization (M) versus applied magnetic field (H) plots of CFO thin film deposited at room temperature on 2.5 nm Au nano-dots and 10 nm Au underlayer respectively. Magnetization measurements were carried out in perpendicular (gray M-H plots) and in-plane (red M-H plots) directions. (e) remanent magnetization (Mr) and remanance ratio (R = Mr/Ms) of CFO deposited at room temperature on varying Au under-layers.

PNR is a technique that can accurately measure the magnetic and chemical depth-profile of thin film samples along the surface normal direction to the substrate. This can be obtained by determining the depth-dependence of the local in-plane scattering lenght density (SLD). PNR data can be obtained for four neutron spin crosssections, namely R++, R--, R+- and R-+, where "+" and "-" signs represent the polarisation directions of the neutron spin with respect to the external magnetic field, which could be either parallel or anti-parallel direction. Out of four neutron spin cross-sections; R++ and R-- are the non-spin-flip (NSF) reflectivity channels. In addition, the nuclear (chemical) structure as a function of thickness is determined by fitting the oscillating fringes of the NSF reflectivity data. The fitting models are built based on both the nuclear scattering length density (NSLD) and the magnetic scattering length density (MSLD) profiles, where NSLD and MSLD reflect the chemical and magnetic depth profile of the sample, respectively.¹⁻⁴ The NSLD of a particular volume-element is the product of the bound coherent scattering length (b_{nuc}) of a particular element and the atomic density (N) of that isotope within the volume-element. If the material is made up of several elemental constituents, the total SLD is the sum of the individual elemental-SLDs. In addition, due to the magnetic dipole-dipole interaction between the neutron and the atom's magnetic moment, magnetized atoms carry a magnetic scattering length +/- b_{mag} that resulting in a MSLD, where the +/- signs denots the direction of polarized neutron beam with spin aligned parallel/anti-parallel to the external magnetic field. The magnetic scattering length is directly proportional to the atom's total magnetic moment. The total local neutron SLD of the alloy consisting of elements 1, 2, 3... is, therefore, given by the: $SLD_{total} = SLD_{total nuc} +/- SLD_{total mag} =$ $(b_{nuc 1} + b_{mag 1}) \times N_1 + (b_{nuc 2} + b_{mag 2}) \times N_2 + \dots$



Supplementary Figure 4: PNR results for 4 nm Au nano-dot/Substrate where (a) NSF reflectivity and NSA data and (b) the NSLD models obtained from the best fit parameters. Fitting model consist a structure; Si/SiO₂ (450 nm)/Au (4 nm). There is no oscillation of reflection intensity in the measured Q range. NSA data suggest that there is no ferri/ferro-magnetism in the sample. Since Au/Air interface in not sharp, the surface roughness of 1 nm is considered to fit the decay of the reflectivity.



Supplementary Figure 5: Schematic view of unrelaxed Au (*111*) and $CoFe_2O_4$ (*311*) interface with four different possible $CoFe_2O_4$ surface terminations. Brown, blue, green and red spheres represent Au, Co, Fe and O respectively.

Supplementary Table 1. The relative total density functional energy of the configurations presented in Supplementary Table 1. The energies were presented with respect to the most stable structure.

Configuration	(a)	(b)	(c)	(d)
Energy (eV)	3.0191	0	11.9959	10.1063

We constructed four different Au(111)/CoFe₂O₄(311) interfaces for which the unrelaxed structures are shown in Supplementary Figure 5. These configurations differ by the termination surface of $CoFe_2O_4$. The $CoFe_2O_4$ at the interface of structure (a) is terminated with O atoms only, the one in (b) is terminated with O, Fe and Co, the one in (c) is terminated Co and Fe only and the one in (d) is terminated with O, Fe and Co with a different morphology from that of (b). As shown in Supplementary Table 1, we found that configuration (b) had the lowest total energy resulting in a most stable configuration therefore it used to investigate the nature of the chemical bonds between Au and Co/Fe/O. As shown in Figure 5, relaxation cuases significant surface reconstruction.

Verticle growth of CFO

In the initial process where nucleation for the formation of stable atomic clusters needs to happen; crystallized Au nano-dots provide preferred nucleation sites for CFO through a heterogeneous or secondary nucleation process.^{5,6} Particularly in the early stage, nucleation sites/events are randomly created and spread over the

surface of Au nano-dots with every cycle of CFO deposition. Every subsequent cycle of CFO deposition adds mass to the already formed nuclei providing a pathway to CFO crystal for its crystal growth through macroscopic dimensions. Here the crystallization process can be in accordance to the Avrami- Johnson-Mehl equation:⁷ $\chi(t) = 1 - \exp[-(t-t_0)^3/t_c^3]$ where $\chi(t)$ is the crystalline fraction, t_c is the characteristic crystallization time, and t_0 is incubation time *(time required for the formation of the first crystallites with a critical size)*. The already formed Au nucleation sites could facilitate the formation of the defined crystalline structure of CFO nuclei whereas the materials landing on amorphous substrate/region generally remain amorphous. Moreover, CFO-crystallites could grow in an upward direction and relatively faster compared to amorphous CFO. In this stage, atoms can migrate to crystalline phase from amorphous phase. The increased number of nuclei facilitate the film growth process and eventually become crystalline due to the growth of the crystallite nuclei.⁸ At a certain stage of deposition, the sputtered film surface is rich in crystallized CFO-crystallites. Therefore, new nucleation sites cannot be created, possibly resulting in the cease of crystallite growth in radial outward direction. Hereafter, CFO crystallized in vertical direction (*perpendicular to substrate plane*) and crystallization proceed further forming a columnar structure (scheme 1c).

Supplementary references

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