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Supplemental Material: Influence of vanadium-doping on the magnetism of FeCo/SiO₂ nanoparticle

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Figure 1: Representative x-ray diffraction pattern of V-doped FeCo/SiO₂ nanoparticles. The reflections are broadened significantly, indicative of the amorphous structure of the core/shell nanoparticles.

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Figure 2: Magnetization as a function of field for the FeCo/SiO2 (3 nm) sample. Three representative hysteresis loops are shown taken at T = 10, 25, and 300 K. The curve at 300 K was taken well above HC, where HC and MR are zero.



Figure 3: Magnetization as a function of field for the FeCo/SiO2 (4 nm) sample. Three representative hysteresis loops are shown taken at T = 10, 25, and 300 K. The curve at 300 K was taken well above HC, where HC and MR are zero.

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Figure 4: Magnetization as a function of field for the FeCo/SiO2 (6 nm) sample. Three representative hysteresis loops are shown taken at T = 10, 25, and 300 K. The curve at 300 K was taken well above HC, where HC and MR are zero.



Figure 5: Magnetization as a function of field for the V-doped FeCo/SiO2 sample. Three representative hysteresis loops are shown taken at T = 10, 25, and 300 K. The curve at 300 K was taken well above HC, where HC and MR are zero.

The calculated anisotropy from the coercivity and saturation magnetisation at 5 K were; 4.82x106 J/kg, 4.62x106 J/kg, 4.82x106 J/kg, and 1.16x106 J/kg for the FeCo/SiO2(3 nm), FeCo/SiO2(4 nm), FeCo/SiO2(6 nm), and V-doped FeCo/SiO2 core/shell nanoparticles respectively. However, it is necessary to note that these values for the anisotropy are an estimate only as it is problematic to mass normalize the saturation magnetization correctly as the total amount of magnetic material was not known. That is, it is not known precisely how much of the magnetic interfacial phase has formed. The observed trend of the anisotropy (e.g. increased K with silica shell

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thickness for the undoped FeCo/SiO2 nanoparticles) is in agreement with anisotropy values obtained from Vogel-Fulcher analysis of the in-phase AC susceptibility (presented in Desautels et. al., Langmuir, 31,2879, 2015).

Unlike the prototypical exchange biased system of CoO/Co core/shell nanoparticles [W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957)] where the antiferromagnetic (AF) CoO shell has formed due to the surface oxidation of the Co cores in atmosphere, we postulate that the AF vanadium oxide has formed due to the oxidation of V in the presence of SiO₂. In addition, as the AF CoO in the CoO/Co core shell nanoparticles forms a uniform shell, in our system, the distribution of vanadium oxide at the interface is unknown. That is, does the vanadium oxide form a uniform shell, or are there pockets of vanadium oxide? The answer to these questions requires a detailed analysis of the interfacial layer using high resolution TEM (with simulations) of FeCo/SiO2 nanoparticles doped with differing amounts of vanadium. We are working currently towards addressing these questions.