Electronic Supporting Information

## Cyanide-bridged NiCr and alternate $\mathrm{NiFe} / \mathrm{NiCr}$ magnetic ultra thin films on functionalized

## $\mathbf{S i}(100)$ surface

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Fig. S1 Infrared spectra centered on the cyanide band of the precursor $\mathrm{K}_{3}\left[\mathrm{Cr}(\mathrm{CN})_{6}\right]\left(2129 \mathrm{~cm}^{-1}\right)$ and the bulk material $\mathrm{Ni}_{3}\left[\mathrm{Cr}(\mathrm{CN})_{6}\right]_{2}\left(2172 \mathrm{~cm}^{-1}\right)$.


Fig. S2 XPS spectra at the Cr 2 p , Ni2p and Fe2p edges for a) the reference $\mathrm{Ni}_{3}\left[\mathrm{Cr}(\mathrm{CN})_{6}\right]_{2}$ bulk compound, b) at the step Ni6 of NiCr SGS, c) at the step Ni6 of NiCr on NiFe SGS (sample G1), d) at the step Nil2 of alternated two cycles NiFe and two cycles NiCr growth (sample G2), e) at the step Ni6 of alternated one cycle NiFe and one cycle NiCr growth (sample G3), f) table with the energy values.
a)


b)

c)


d)



e)



f)

| Energies (eV) | Cr2p ${ }_{1 / 2}$ | Cr2p ${ }_{3 / 2}$ | Ni2p $\mathrm{p}_{1 / 2}$ | Ni2p $\mathrm{p}_{3 / 2}$ | $\mathrm{Fe} 2 \mathrm{p}_{1 / 2}$ | $\mathrm{Fe} 2 \mathrm{p}_{3 / 2}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $\begin{gathered} \hline \text { Bulk } \\ \mathrm{Ni}_{3} \mathrm{Cr}_{2} \end{gathered}$ | 587.3 | 577.9 | 873.6 | 856.5 | - | - |
| $\mathrm{Ni}-\mathrm{Cr}$ | 587.3 | 577.8 | 873.8 | 856.4 | - | - |
| $\begin{gathered} \begin{array}{c} \mathrm{Ni}-\mathrm{Cr} \\ \text { on } \mathrm{Ni}-\mathrm{Fe} \end{array} \end{gathered}$ | 587.4 | 578.0 | 873.8 | 856.2 | 721.5 | 708.7 |
| Alternated two cycles | 587.2 | 577.8 | 873.9 | 856.4 | 721.8 | 709.0 |
| Alternated one cycle | 587.2 | 577.9 | 874.0 | 856.3 | 721.7 | 708.9 |

Fig. S3 Alternated one cycle NiFe and one cycle NiCr growth ( 6 cycles) - sample G3: a) Evolution of the infra-red spectra centered on the cyanide band from the step Nil to the step Ni6 - IR spectra and peak area at each step. b) Atomic Force Microscopy image and cross section represented by the blue line on the image at the step Ni6. c) $\mathrm{M}=\mathrm{f}(\mathrm{T})$ curves at $\mathrm{H}=500$ Oe at the step Ni6 - plain circles: ZFC, open circles: FC.
a)

b)


c)


