Electronic Supplementary Information (ESI): Magnetic clusters as efficient EY-like spin-scattering centre in graphene

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1 Dependence of the computed quantities on the Ni$_4$ cluster density

To check that the difference in cluster density between the theory and experiment does not affect our conclusions, we performed additional simulations by varying the graphene supercell sizes: 4×4 (corresponding to a density of $11.8 \cdot 10^{13}$ cm^{-2}), 6×6 ($5.3 \cdot 10^{13}$ cm^{-2}) and 7×7 ($3.9 \cdot 10^{13}$ cm^{-2}); their respective band structures are plotted in Fig S1. From this figure, it can be clearly seen that by increasing or decreasing the cluster density, the Ni$_4$ cluster always tranfer electrons to graphene (n-type doping), as observed from the shift of the Fermi level above the Dirac point. The calculated doping efficiency was found to be 0.064 e/cluster (4×4), 0.093 e/cluster 6×6) and 0.11 e/cluster (7×7), respectively. The estimated doping efficiency varies within the range of ± 0.02 from the values provided in the main text for the (5×5) supercell, which is 0.09 e/cluster. Similarly the average valence band spin splittings are found to be 39.6 meV (4×4), 39.1 meV (6×6) and 30.6 meV (7×7). The average conduction band spin splittings are found to be 23.5 meV (4×4), 17.1 meV (6×6) and 10.2 meV (7×7). Though the values vary slightly with the cluster density (which is similar to observations for small gold clusters in [1], the splittings of the valence band

are consistently larger than the conduction band ones accross all the simulated cluster densities, thereby maintaining a similar trend. Therefore, the variation in cluster densities should not drastically affect the main conclusions drawn in this work.
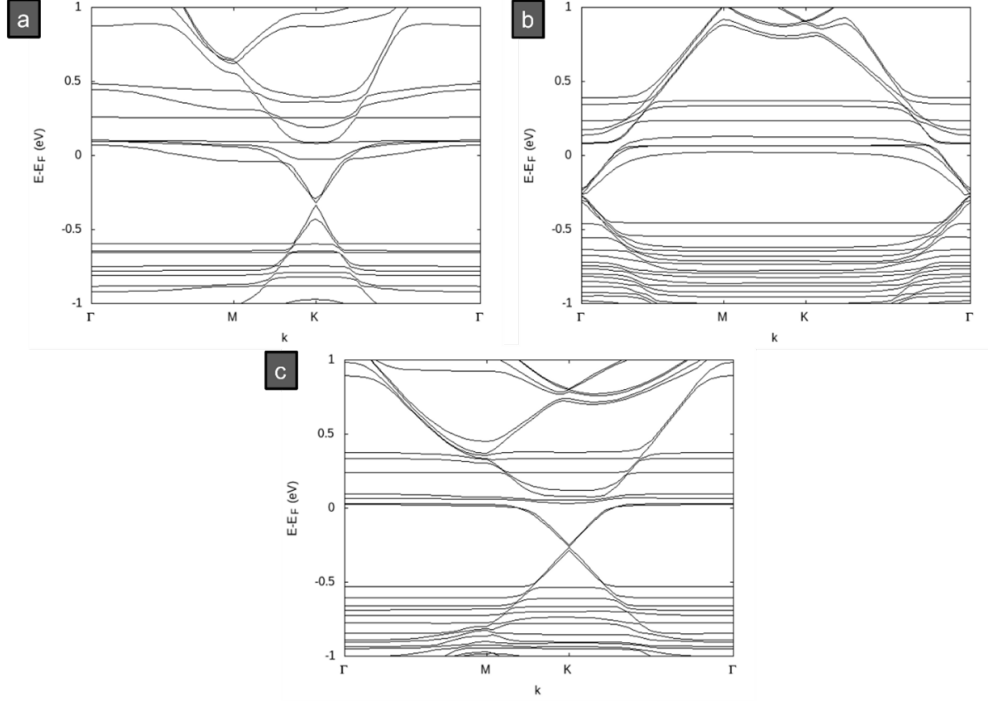

Fig. 1 Simulated non-collinear electronic structure of the Ni_4 cluster adsorbed on graphene for varying supercell sizes: (a) 4×4 (cluster density of $11.8 \cdot 10^{13}$ cm^{-2}), (b) 6×6 ($5.3 \cdot 10^{13}$ cm^{-2}) and (c) 7×7 ($3.9 \cdot 10^{13}$ cm^{-2}). In the case of the 6×6 supercell (multiple of 3n), the K and (K') points are folded to the Γ point of graphene.

References

[1] Keijers, W., Murugesan, R., Libeert, G., Scheerder, J.E., Raes, B., Brems, S., Gendt, S.D., Houssa, M., Janssens, E., Vondel, J.V.: Tuning the spintronic properties of graphene with atomically precise au clusters. Journal of Physics: Materials **4**(4), 045005 (2021)