Lanthanide atoms on surfaces: from single atom magnets to spin qubit candidates

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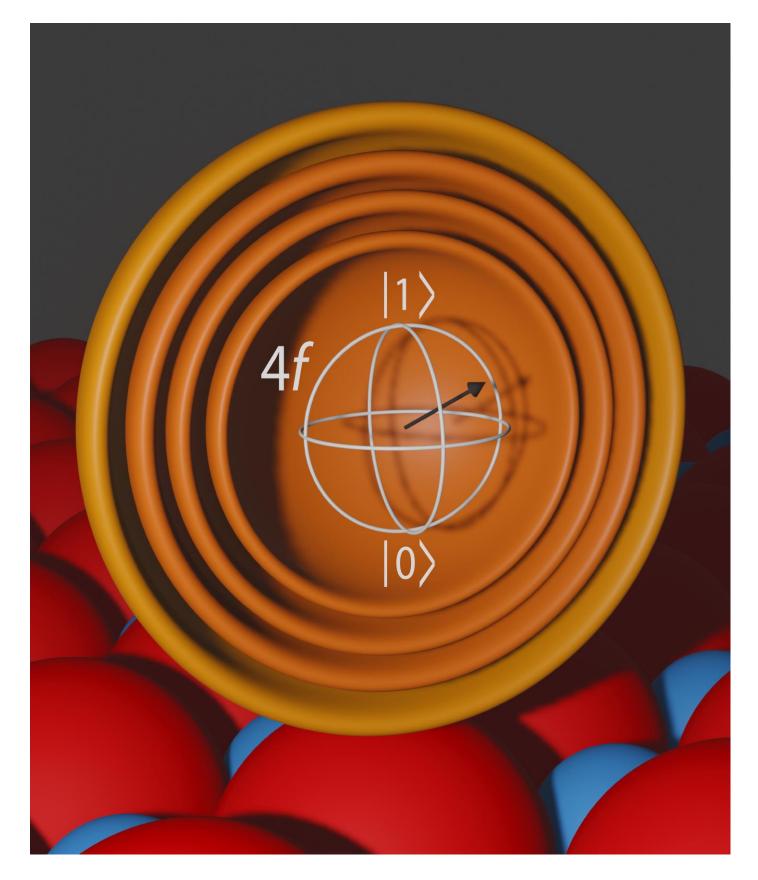
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Lanthanide atoms supported on suitable surfaces have been shown to exhibit long magnetic lifetimes, leading to magnetic remanence and large coercive fields at low temperature. These so-called single atom magnets are the smallest unit of matter that can be used to store and manipulate information [Science 352, 318 (2016)], hence, they represent the ultimate limit of downscaling for magnetic storage devices [Appl. Phys. Lett. 119, 160503 (2021)]. These properties also pave the way to their use as atomic-scale quantum logic units, i.e. qubit.

In the first part of this talk, I will present the properties of lanthanide atoms adsorbed on ultra-thin MgO films on Ag(100). Using X-ray absorption spectroscopy we found that ensembles of holmium (Ho) atoms on MgO/Ag(100) exhibit magnetic bistability and a magnetic lifetimes of thousands of seconds below 40 K [Phys. Rev. Lett. 124, 077204 (2020)]. For samples with an MgO thickness of 2 monolayers, it is possible to use spin-polarized scanning tunnelling microscopy (STM) to read and write the magnetic state of individual Ho atoms [Nature 543, 226 (2017)]. We found similar results for dysprosium (Dy) atoms on 2 ML of MgO, which in addition possess an extremely large perpendicular magnetic anisotropy (PMA) of 250 meV, i.e. twice the value observed for Ho [Nat. Commun. 12, 4179 (2021)].

In the second part, I will address the origin of the large PMA and magnetic bistability of these atoms. Exploiting the optical selection rules of the X-ray absorption transitions, we addressed the occupation and spin polarization of the valence orbital of lanthanide atoms on MgO/Ag(100). Combining our results with density functional theory, we inferred a transfer of charge through the oxide layer towards the underneath silver substrate [ACS Nano 15, 16162 (2021)]. This charge transfer occurs when the thickness of the MgO is below a critical value. For the case of dysprosium, this mechanism can be suppressed by increasing the MgO thickness above 6 ML. On such films, Dy atoms exhibit a neutral, free-atom electronic configuration with magnetic bistability at finite fields, but with no magnetic remanence and reduced anisotropy energy with respect to the same atoms adsorbed on 2 ML of MgO/Ag(100) [Nano Lett. 21, 8266 (2021)]. This result allowed us to conclude on the importance of the charge transfer mechanism to maximize the magnetic anisotropy of surface-adsorbed atoms.

In the last part, I will discuss the possible use of lanthanide atoms as surface spin qubits. To allow for quantum coherent operations, an atom should have a suitable ground state that can efficiently interact with microwave fields. While Ho and Dy single atom magnets do not exhibit this property, we found that erbium (Er) and thulium (Tm) atoms on MgO/Ag(100) possess the required characteristics [Phys. Rev. B 107, 045427 (2023)]. Further studies using electron-spin resonance STM will allow us to address their performance in coherent operations and their potential for the realization of future atomic scale quantum devices.



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