

Atomic-scale magnetometry with a mobile molecular quantum sensor

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Recent advances in scanning probe techniques rely on the chemical passivation of the probe-tip termination with single molecules weakly connected to the metallic apex. Valuable information, otherwise inaccessible with a metallic tip, can then be gathered, notably offering to image the skeletal structure and bonding of single molecules. The undeniable success of this approach opens the tantalizing prospect of introducing spin sensitivity through the probe-tip termination with a magnetic molecule. Here, we use a tip decorated by a single nickelocene molecule [1,2] and exploit it as a spin-sensitive mobile sensor. Nickelocene behaves as a two-level spin system where the uniaxial magnetic anisotropy (noted D) separates the magnetic ground state $|S = 1, M = 0\rangle$ from the excited state $|S = 1, M = \pm 1\rangle$. This nickelocene-terminated tip, or Nc-tip, offers the unique possibility of producing electrically-driven spin excitations on the tip apex. We show that when the Nc-tip is within 150 pm from a magnetic object, its spin excited spectrum is modified by the presence of an exchange interaction. Magnetic information may be probed with lateral and vertical atomic-scale resolution. Virtually all systems may be addressed with this detection scheme, including ferromagnetic surfaces, as we exemplify for a single Fe atom on Cu(100) and for surface atoms within a cobalt island grown on Cu(111).

[1] M. Ormaza *et al.*, Nat. Commun. **8**, 1974 (2017)

[2] M. Ormaza *et al.*, Nano Lett. **17**, 1877 (2017)