Interplay of magnetic states and hyperfine fields of iron dimers on MgO(001)

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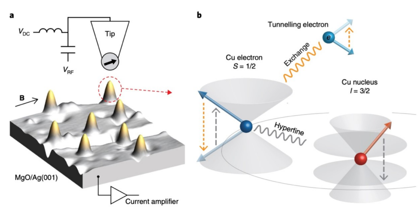
Abstract

Individual nuclear spin states can have very long lifetimes and could be useful as qubits. Progress in this direction was achieved on MgO/Ag(001) via detection of the hyperfine interaction (HFI) of Fe, Ti and Cu adatoms using scanning tunneling microscopy (STM). Previously, we systematically quantified from first-principles the HFI for the whole series of 3d transition adatoms (Sc–Cu) deposited on various ultra-thin insulators, establishing the trends of the computed HFI with respect to the filling of the magnetic s- and d-orbitals of the adatoms and on the bonding with the substrate. Here we take one step further by investigating the impact of the magnetic coupling between the dimer atoms on the HFI of Fe dimers on MgO(001) and its dependence on where the Fe atoms are located on the surface.

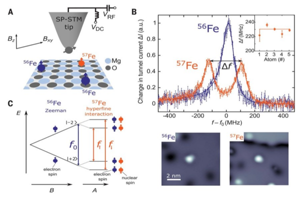
Probing hyperfine interactions atom by atom

Electrically controlled nuclear polarization of Cu on MgO/Ag(001)



Yang et al., Nat Nanotechnol **13**, 1120 (2018)

Hyperfine splitting for ^{57}Fe on MgO/Ag(001)

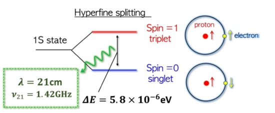


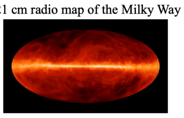
Willke et al., Science **362**, 336 (2018)

Current experiments limited to Ti, Fe and Cu adatoms on MgO

Hyperfine interactions

- Hyperfine interaction originates in the coupling between the electrons and the nuclear spins
- It can be used to detect the state of the nuclear spin





21 cm radio map of the Milky Way

Y. Oyama, PhD Thesis, SOKENDAI, Japan (2014)

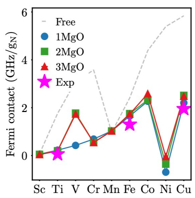
$$\hat{H} = \mathbf{S}\cdot\underline{A}(\mathbf{R})\cdot\mathbf{I}$$

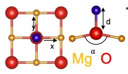
$$A_{ij} = a\,\delta_{ij} + b_{ij}\ (i,j = x,y,z)$$

Fermi contact — Dipolar interaction

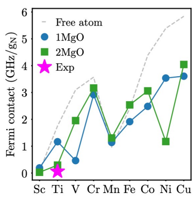
$$a = \frac{2P}{3}\, m_s(\mathbf{R}) \qquad b_{ij} = \frac{P}{4\pi}\int dr\, \frac{3r_ir_j - r^2\delta_{ij}}{r^5}\, m_s(\mathbf{r})$$

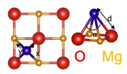
Hyperfine interaction on MgO





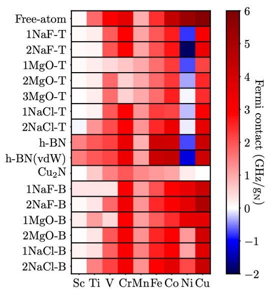
- V: change in valence
- Ni: antiparallel coupling between s and d spin moments



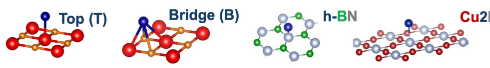


- Bridge-site leads to larger values
- Ni: the s-d coupling is parallel

Bird's eye view of hyperfine interactions



- Ni: the s-d coupling is antiparallel on top by contrast on bridge-site
- Bridge-site leads to larger values
- Top site: largest values on h-BN
- Maximum value for Cu on 2NaF, bridge-site



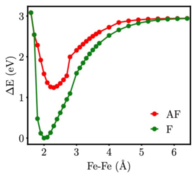
Shehada et al., Npj Comput. Mater. **7**, 87 (2021)

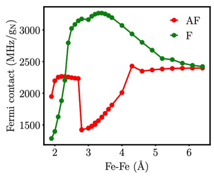
There is a correlation between the HFI and the magnetic state of Fe dimers?

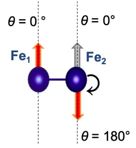
Density Functional Theory

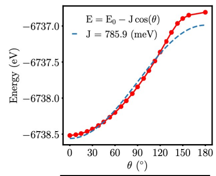
- Quantum Espresso
  Gianozzi et al., J Phys Condens Matter **21**, 395502 (2009)
- Pseudopotentials from the PSLibrary
  Dal Corso, Computational Materials Science **95**, 337-350 (2014)
- Projector augmented wave (PAW) method
  Blöchl, Phys Rev B **50**, 17953-17979 (1994)
- Exchange and correlation functional — PBE
  Perdew et al., Phys Rev Lett. **77**, 3865 (1996)
- Hyperfine parameters computed with GIPAW
  Pickard et al., Phys Rev B **63**, 245101 (2001)

Hyperfine interaction of isolated Fe dimers

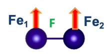




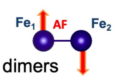




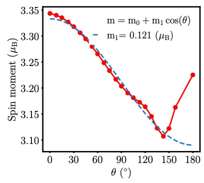
- Fe dimers prefer to be in F states

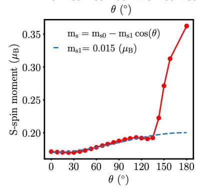


- HFI can be controlled by:



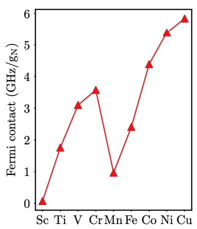
  - ✓Switching the magnetic state of the dimers
  - ✓Changing the distance between the atoms

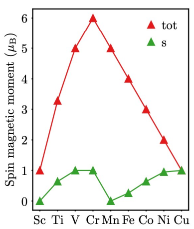




$$\hat{H} = \sum_i \mathbf{S}_i\cdot\left(\underline{A}_i^{(0)} + \sum_j \underline{A}_{ij}^{(1)}\,\mathbf{S}_i\cdot\mathbf{S}_j + \dots\right)\cdot\mathbf{I}_i$$

Hyperfine interaction isolated atoms

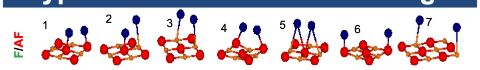


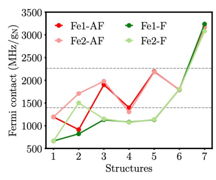


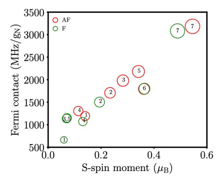
- The Fermi contact term is driven by spin moment from s electron at the nucleus

$$a = \frac{2P}{3}\, m_s(\mathbf{R})$$

Hyperfine interaction of Fe2/2MgO







- Fe dimers in the isolated and on MgO cases prefer to be in F states
- The HFI are stronger for the Fe dimers on top of Mg than for those on top of O similarly to adatoms
- The transition from F to AF phase leads to larger hyperfine interaction

Conclusions

Shehada et al. ArXiv:2202.00336

- We demonstrate the ability to substantially modify the HFI by atomic control of the location of the adatoms on the substrate
- The magnitude of the HFI can be controlled by switching the magnetic state of the dimers
- Individual nuclear spin states can have very long lifetimes and could be useful as: Magnetic bit for information storage and Qubits for quantum computing
- Our work has immediate implications in future STM investigations aiming at detecting and realizing quantum concepts hinging on nuclear spins

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