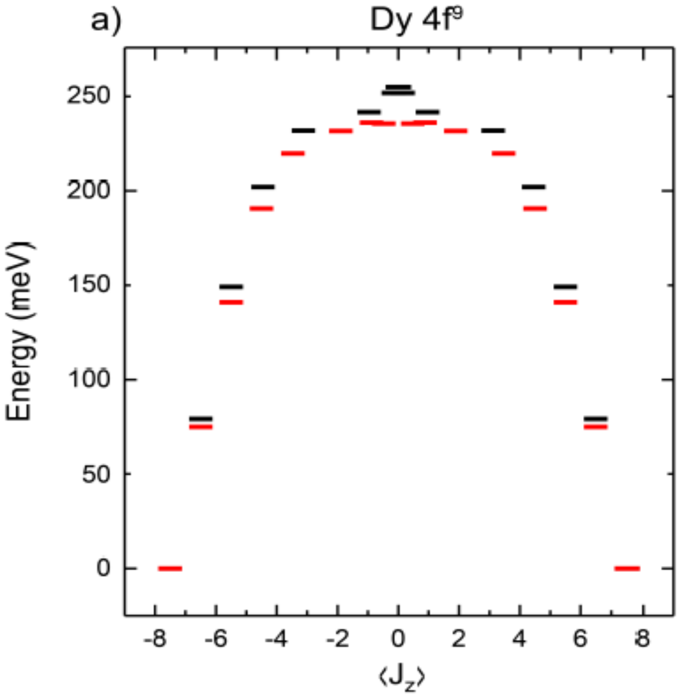
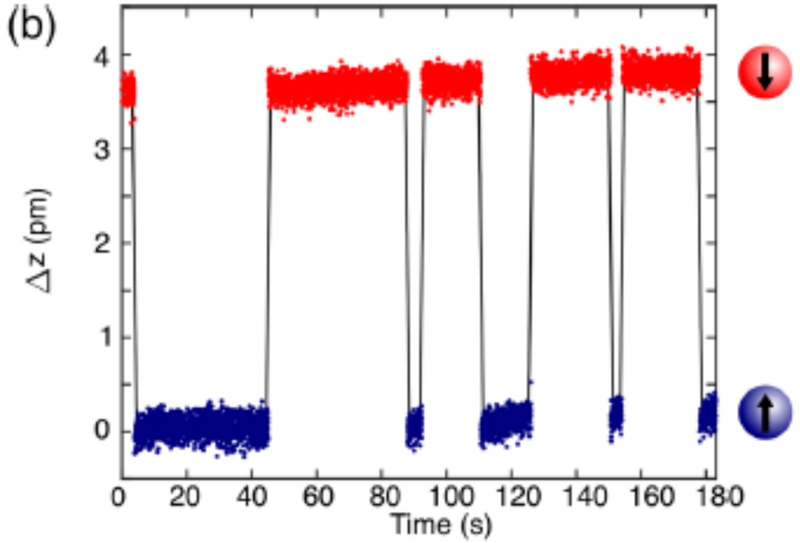
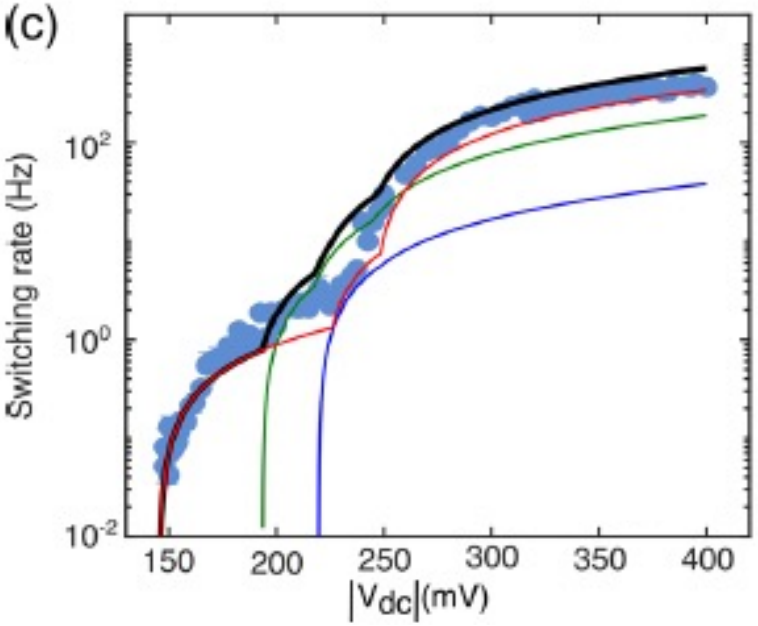
9.1 Spin excitation of Dy atoms on MgO

Combining DFT and multiplet calculations, we deduce the energy scheme for a Dy atom adsorbed on top of an O atom on the MgO surface, shown in figure a) (consider only the black levels).
For experimental confirmation, we perform an experiment with STM at T = 0.7 K. To do that we place the STM tip on top of a Dy atom and we acquire the telegraph signal as a function of V_{bias} as shown in figure b). Each time that V_{bias} matches one of the energy levels, an increase in the switching rate in the telegraph signal is observed (see fig. c).

Describe which kind of transitions are responsible for the accelerations seen in figure c) and motivate why these transitions are possible.





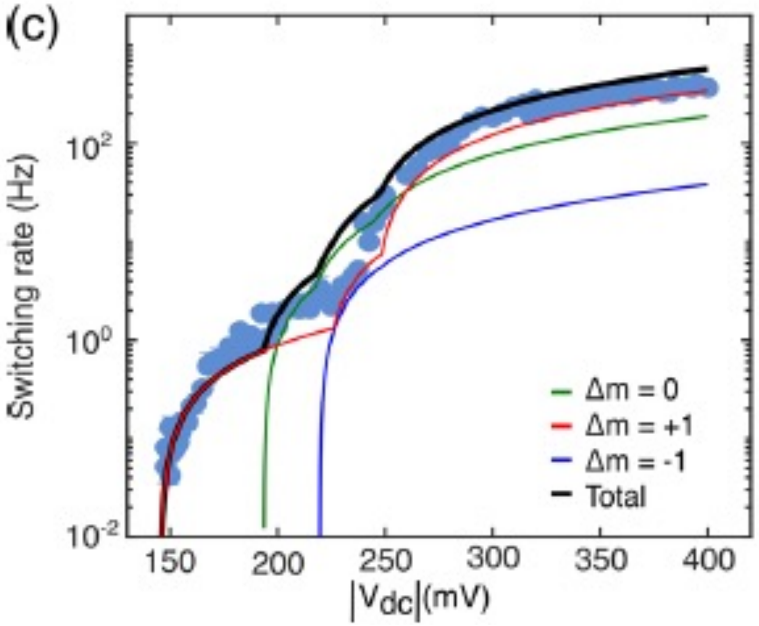


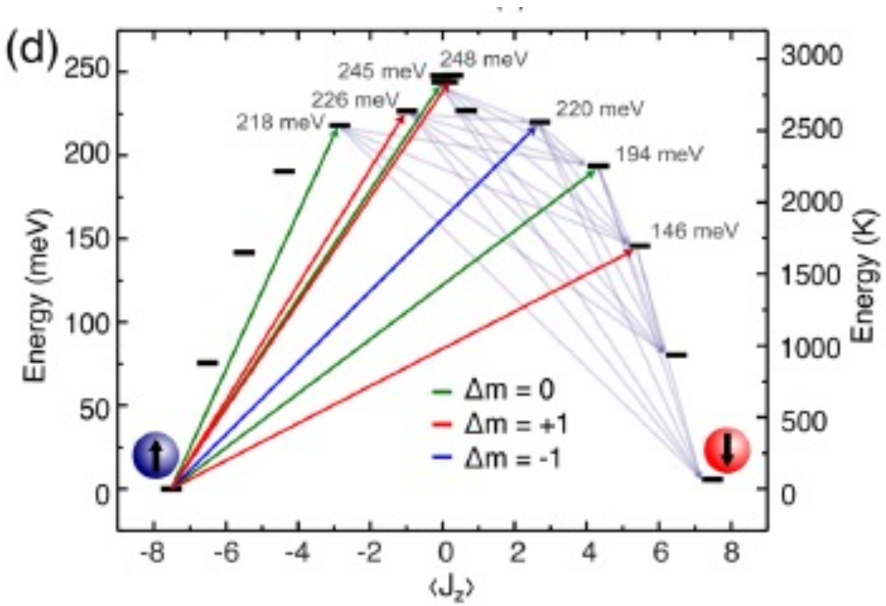
Adapted from https://doi.org/10.1038/s41467-021-24465-2

9.1 Spin excitation of Dy atoms on MgO - Solution

The possible transitions are the ones shown in the figures below

Note that also the transition to the first excited doublet (75 meV) is possible, but in that cases the spin lifetime is so long, or equivalently the switching rate so small, that it is not possible to observe it in the SP-STM experiment





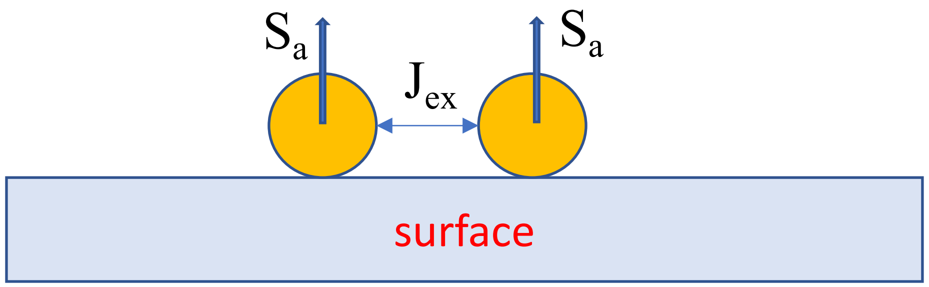
Adapted from https://doi.org/10.1038/s41467-021-24465-2

9.2 Spin excitations in exchange-coupled atoms

Consider two atoms on a surface. For simplicity we assume that each atom has a spin $S_a=1/2$ which can point up or down. Using the standard notation |S,m>, each atom can then assume the states |1/2, 1/2> or |1/2, -1/2>.

With an STM tip we approach the two atoms to form a dimer.

An exchange coupling couples the two spins: the dimer is then described by an Hamiltonian of the form $H = -2J_{ex}\,\mathbf{S_a}\,\mathbf{S_a}$, where J_{ex} is the exchange constant.

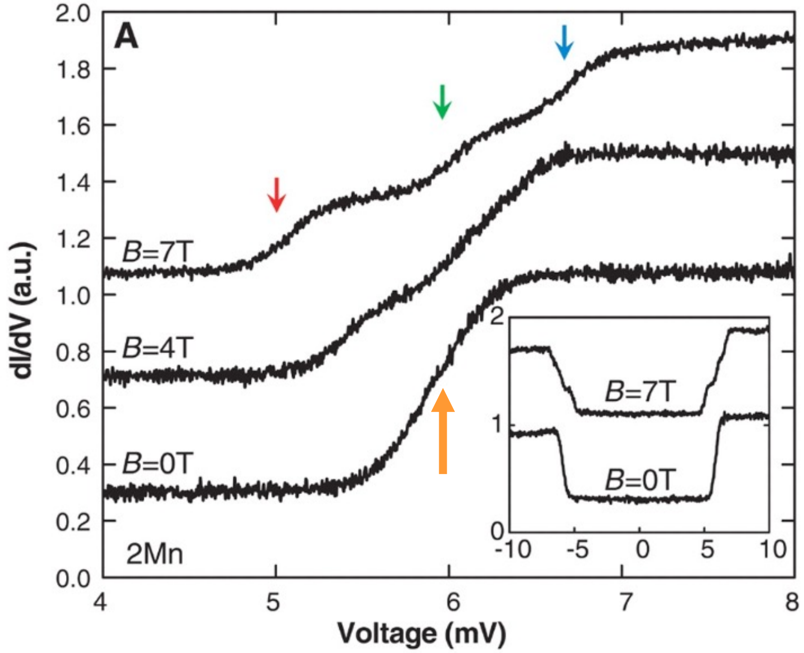


1) What is the total spin S_d of the dimer? Assuming J_{ex} negative (antiferromagnetic coupling) and B = 0, describe the ground and excited states of the dimer and their energy separation.

2) We go with the STM tip on top of the dimer and we perform an IETS experiment. The result is shown in the figure here on the side.

Why at B = 0 only one excitation (orange arrow) is observed while in field three excitations (red, green, and blue arrows) are observed? Use the experimental data to estimate the exchange constant J_{ex}.

IETS spectra at different magnetic field B on a Mn dimer



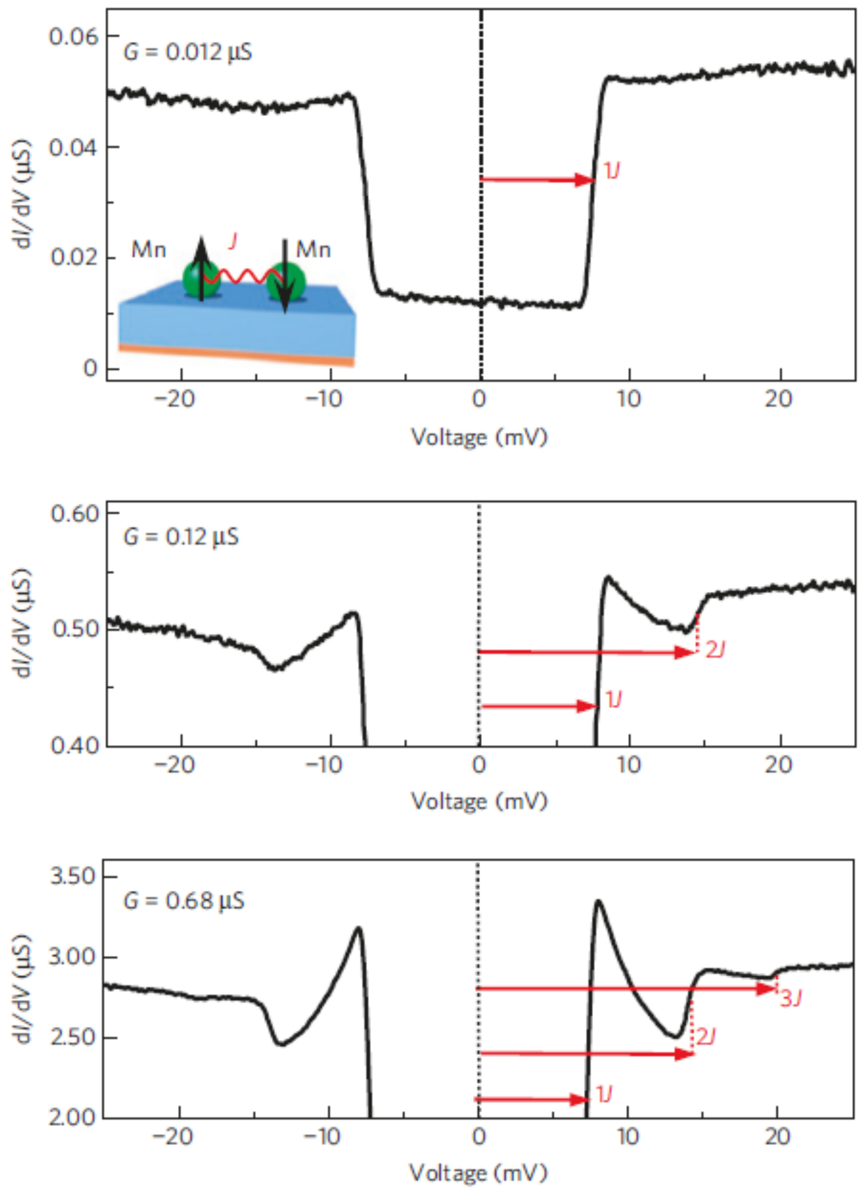
Adapted from : C. F. Hirjibehedin et al., Science 312, 1021 (2006) https://doi.org/10.1126/science.1125398,

9.2 Spin excitations in exchange-coupled atoms

3) We now want to see the effect of the tunneling current on the IETS spectra. The results are shown in the figure on the side. G is the conductance, proportional to the tunneling current.

Is the assumption of S=1/2 a good assumption? How the description changes if we take the correct value of S=5/2 for the Mn atom spin?

Note that in the figure the authors use the convention $J = 2J_{exc}$



Adapted from : C. F. Hirjibehedin et al., Science 312, 1021 (2006) https://doi.org/10.1126/science.1125398,

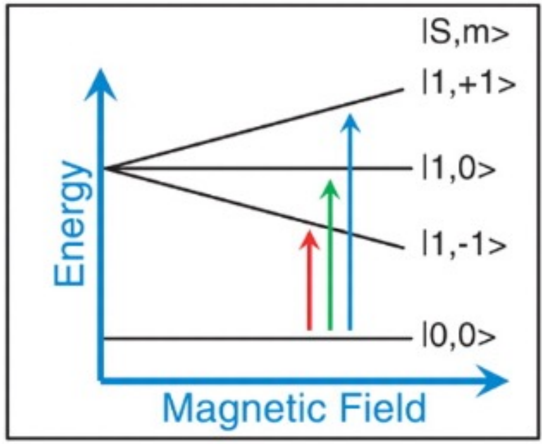
9.2 Spin excitations in exchange-coupled atoms - Solution

1) The atom spins in the dimer can be:
parallel to give a dimer spin $S_d = 1$ corresponding to the triplet |1,-1>, |1,0> and |1,1> (ferromagnetic coupling) or
antiparallel to give a dimer spin $S_d = 0$ corresponding to the singlet |0,0> (antiferromagnetic coupling).

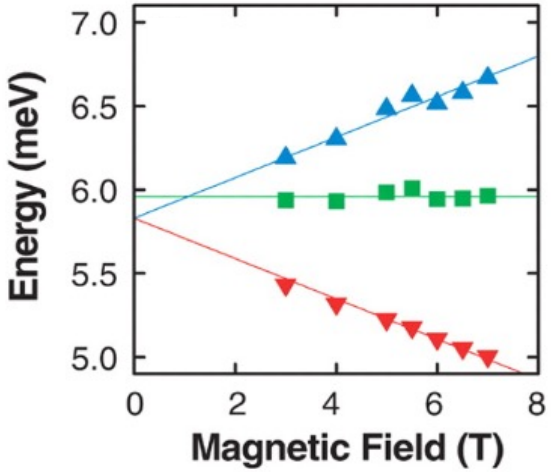
At B = 0 the ground state is given by the singlet |0,0> and the excited state by the three states of the triplet (|1,m> m = -1, 0, 1) which are degenerate in energy. The energy separation between the singlet and the triplet is equal to $2J_{ex}$.

The spin *s* of the tunneling electron, while scattering with the spin of the dimer, undergoes a change of $\Delta\sigma = \pm 1, 0$. To conserve the total moment, all the transitions in the sketch are possible for the dimer spin: red ($\Delta m = -1$), green ($\Delta m = 0$) and blue ($\Delta m = +1$).

expected



measured



2) At B = 0 the three states of the triplet are degenerate in energy and then only the transition corresponding to the orange arrow is observed.

In an external magnetic field, the Zeeman energy splits the three states of the triplet.

The exchange constant can be estimated by the position of the step in the IETS spectrum acquired at B = 0, which is roughly 6 meV.
This corresponds to $J_{ex} = 3$ meV

9.2 Spin excitations in exchange-coupled atoms - Solution

3) We remind that 2 $\boldsymbol{S}_a \cdot \boldsymbol{S}_a = \boldsymbol{S}_d^2 - \boldsymbol{S}_a^2 - \boldsymbol{S}_a^2 = \boldsymbol{S}_d^2 - 2\boldsymbol{S}_a^2$

If we assume that S_a=1/2, the exchange energy corresponding to $H_{exc} = -2J_{exc}\boldsymbol{S}_a \cdot \boldsymbol{S}_a$ can have only the values -3/2 J_{exc} for S_d= 0 and 1/2 J_{exc} for S_d= 1.

→ only one transition should be seen in the IETS corresponding to $E_{exc}(S_d = 1) - E_{exc}(S_d = 0) = 1/2\, J_{exc} - (-3/2\, J_{exc}) = 2J_{exc}$

Additional transitions are possible only when we consider the correct description with S_a=5/2.

In this case $\boldsymbol{S}_d = \boldsymbol{S}_a + \boldsymbol{S}_a$ can assume all the integer values between S_a- S_a and S_a+ S_a ,i.e. S_d =0, 1, 2, 3, 4, 5.

As a consequence 2 $\boldsymbol{S}_a \cdot \boldsymbol{S}_a = \boldsymbol{S}_d^2 - 2\boldsymbol{S}_a^2$ can assume the following values:

$S_d = 0 \quad \Rightarrow\ 2\,\boldsymbol{S}_a \cdot \boldsymbol{S}_a = -2S_a^2$

$S_d = 1 \quad \Rightarrow\ 2\,\boldsymbol{S}_a \cdot \boldsymbol{S}_a = 2\ - 2S_a^2$

$S_d = 2 \quad \Rightarrow\ 2\,\boldsymbol{S}_a \cdot \boldsymbol{S}_a = 6 - 2S_a^2$

$S_d = 3 \quad \Rightarrow\ 2\,\boldsymbol{S}_a \cdot \boldsymbol{S}_a = 12 - 2S_a^2$

$S_d = 4 \quad \Rightarrow\ 2\,\boldsymbol{S}_a \cdot \boldsymbol{S}_a = 20 - 2S_a^2$

$S_d = 5 \quad \Rightarrow\ 2\,\boldsymbol{S}_a \cdot \boldsymbol{S}_a = 30 - 2S_a^2$

Then we should observe the following transitions:

$\Delta_{10} = 2J_{exc} = J;$

$\Delta_{21} = 4J_{exc} = 2J;$

$\Delta_{32} = 6J_{exc} = 3J;$

$\Delta_{43} = 8J_{exc} = 4J;$

$\Delta_{54} = 10J_{exc} = 5J;$

Experimentally we see only the first three. The reason is that to see them we need to use tunneling currents high enough to keep the excited states populated (spin pumping) against spin-phonon and spin-conduction electron scattering events that tend to relax the system to the ground state. Then, the higher we go in the excited state index, the smaller is the population of the state and then smaller is the excitation intensity