JÜLICH
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Molecular magnetism goes spintronic

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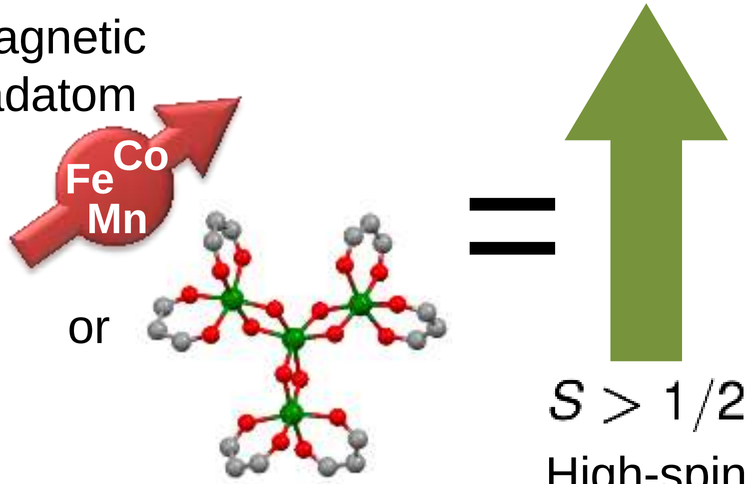
13th  December 2013, Santa Barbara

Alexander von Humboldt
Stiftung/Foundation

FNP
Foundation for Polish Science

Superparamagnets: why magnetic anisotropy?

Magnetic adatom



or

Single-molecule magnet (SMM)

=

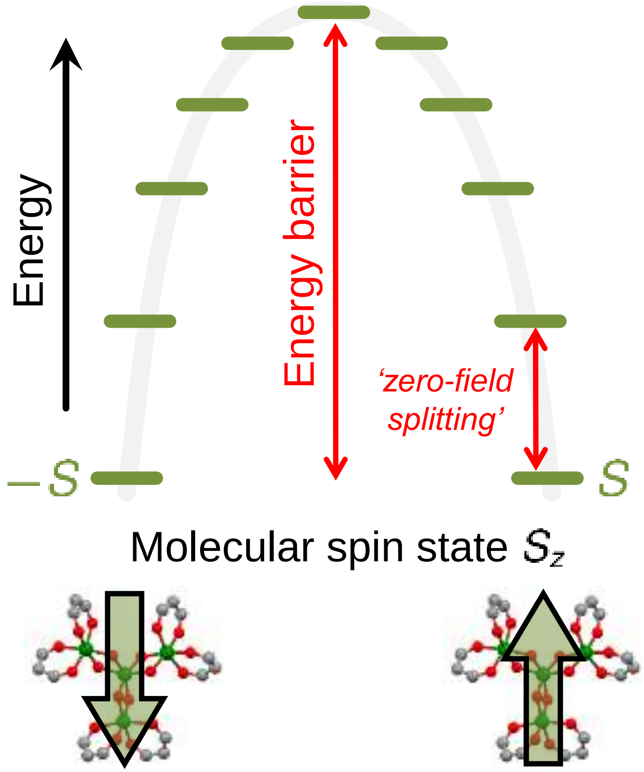
$S > 1/2$

High-spin ground state

Uniaxial magnetic anisotropy

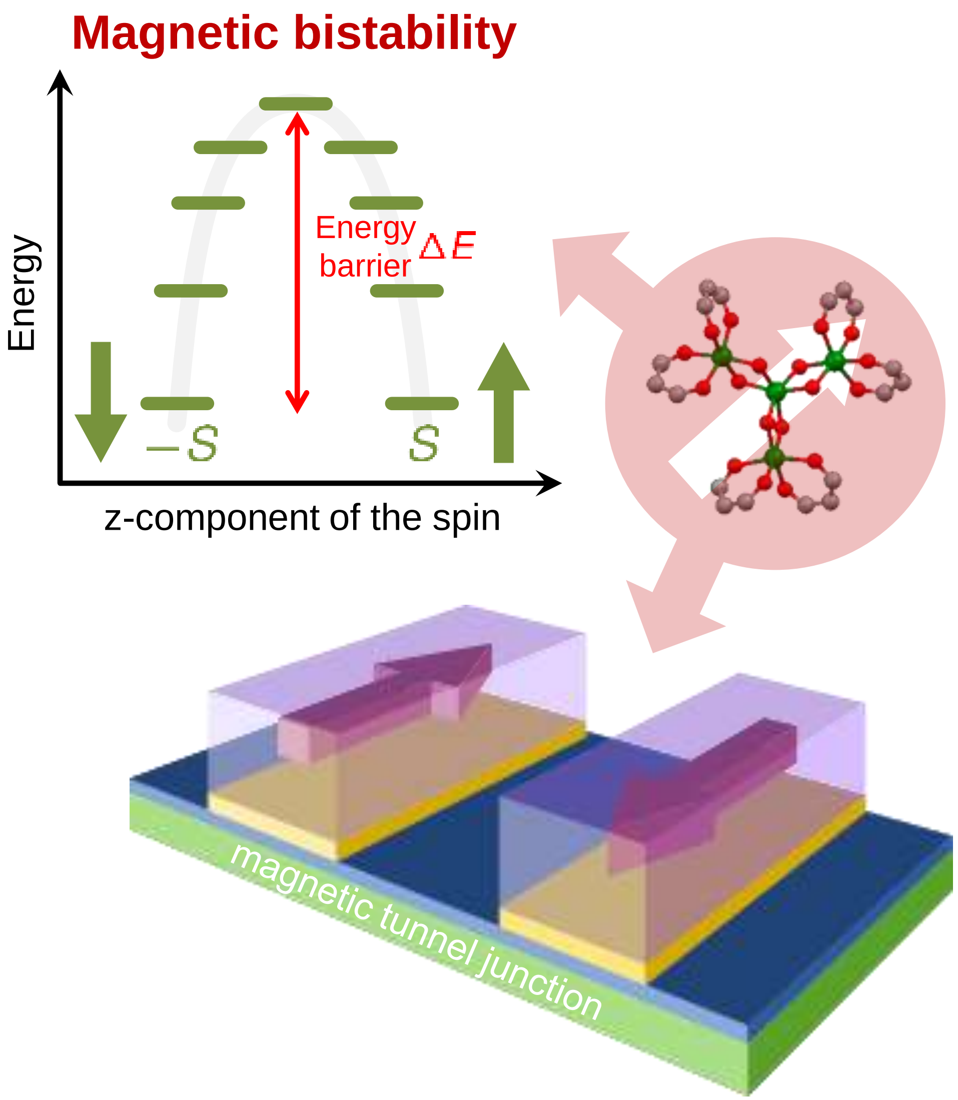
= preferential orientation along a specific direction

Magnetic bistability



Magnetic bistability = prerequisite for a building block of a memory cell

Motivation: why spin-polarized transport?



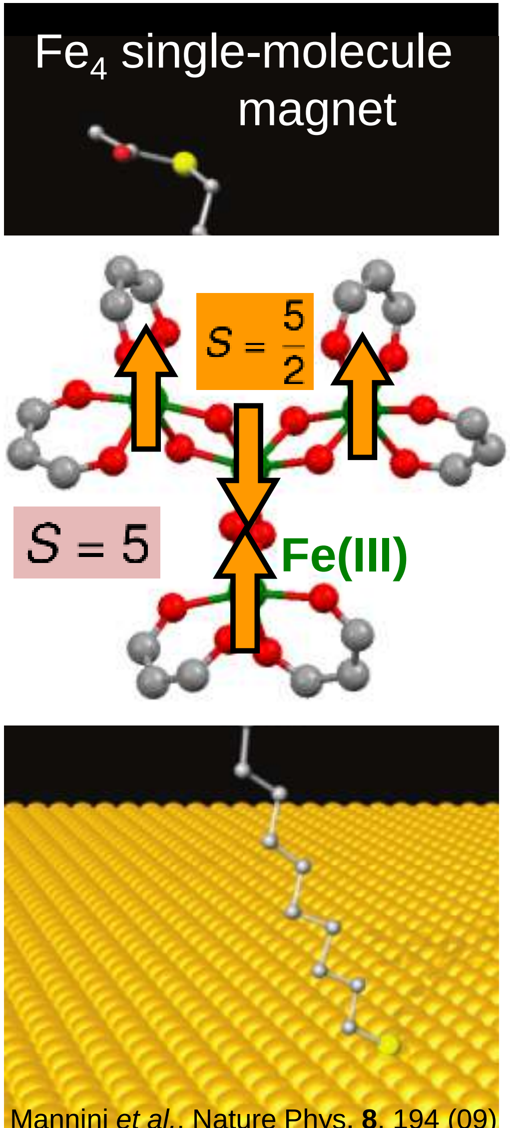
IDEA: use *spin-polarized* currents to control magnetic state of SMM

CHALLENGES:

- synthesis of SMMs with large energy barriers
- stability of SMM's magnetic properties when connecting to a electronic/spintronic circuit

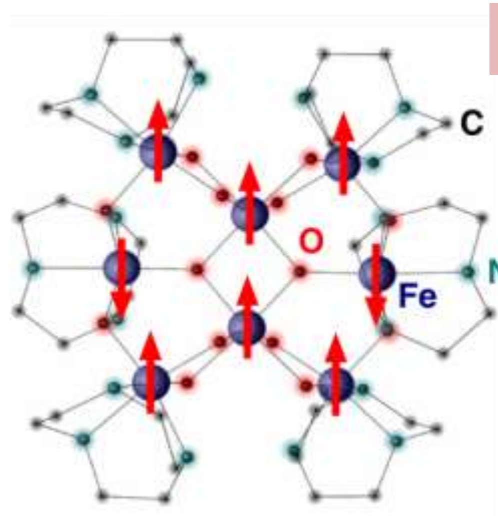
QUESTION: can one *generate* the effect of molecular magnetism, i.e. magnetic anisotropy, in a nano-scopic spintronic system **without** intrinsic anisotropy?

Molecular magnets: magnetism at nanoscale

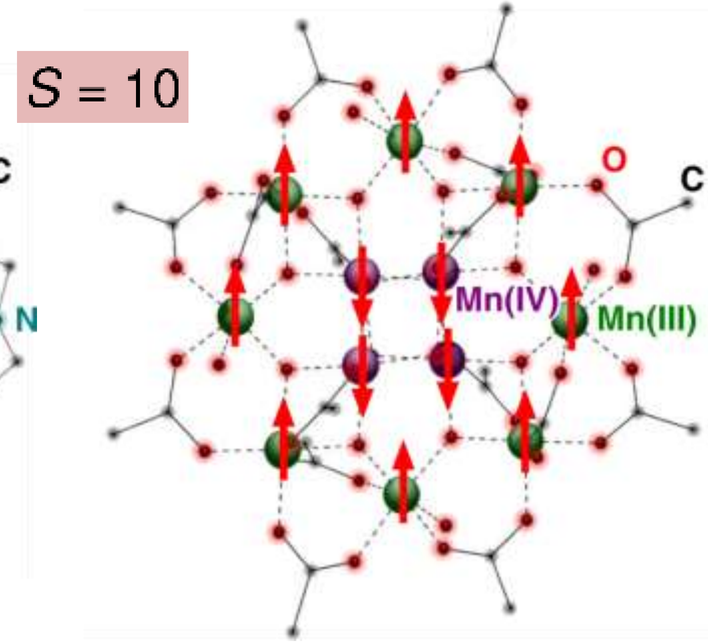


Mannini *et al.*, Nature Phys. **8**, 194 (09)

Nanoscopic systems, whose magnetism stems from strong interactions between their magnetic constituents



$[Fe_8O_2(OH)_{12}(tacn)_6]^{8+}$

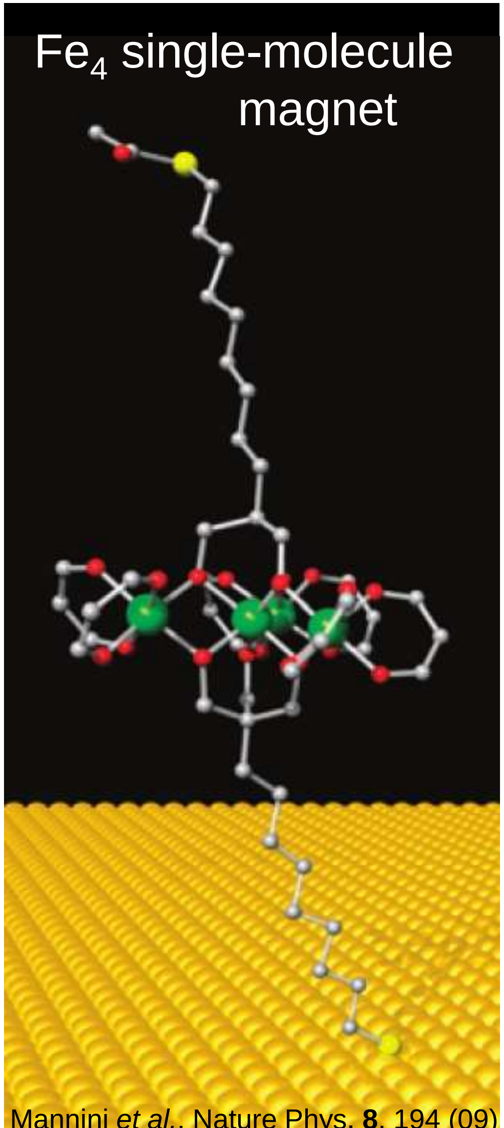


$[Mn_{12}O_{12}(RCOO)_{16}(H_2O)_4] \cdot Y$

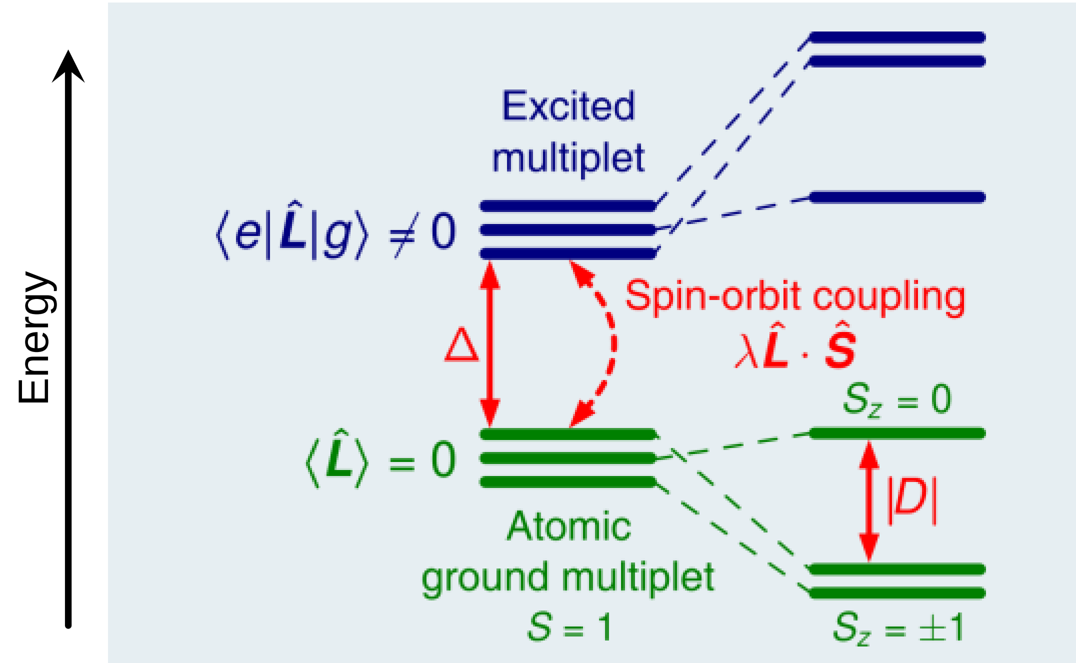
Uniaxial anisotropy constant: { **SMMs** $|D| \lesssim$ tens of μeV; **magnetic adatoms** $|D| \lesssim$ a few meV }

Aromi & Brechin, Struct. Bond. **122**, 1 (06);
Gatteschi et al., *Molecular nanomagnets*, OUP 2006

The origin of magnetic anisotropy



Mannini *et al.*, Nature Phys. **8**, 194 (09)



Effective *giant-spin* Hamiltonian:

$$\mathcal{H}_{\rm eff} = \boldsymbol{S}\cdot\boldsymbol{D}\cdot\boldsymbol{S} = D\left[S_z^2 - \tfrac{1}{3}S(S+1)\right] + E(S_x^2 - S_y^2)$$

z-z spin-quadrupole moment: Q_{zz}

$$D_{ij} = -\lambda^2 \frac{\langle g|L_i|e\rangle\langle e|L_j|g\rangle}{\Delta}$$

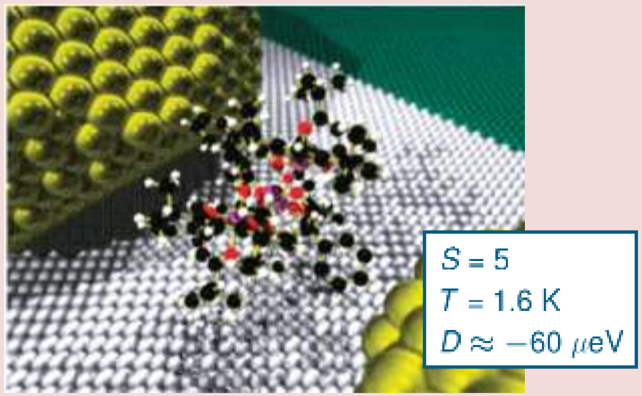
$$D = \tfrac{3}{2}D_{zz}$$

$$E = \tfrac{1}{2}(D_{xx} - D_{yy})$$

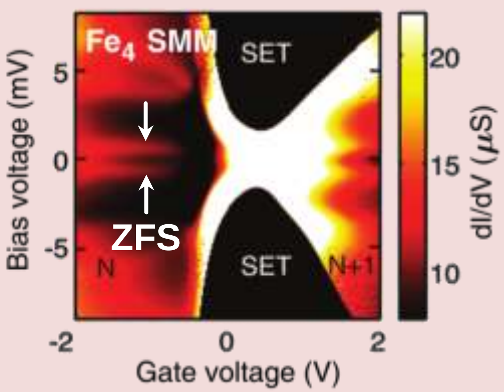
Kahn, *Molecular magnetism*, VCH
Gatteschi *et al.*, *Molecular nanomagnets*, OUP

Electronic transport through molecular magnets

Molecular junction

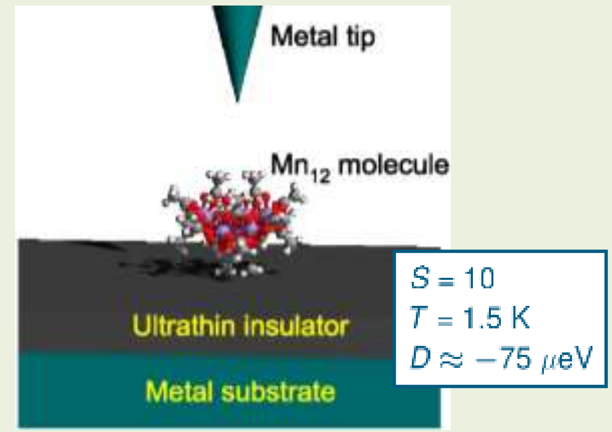


Zyazin *et al.*, Nano Lett. **10**, 3307 (10)

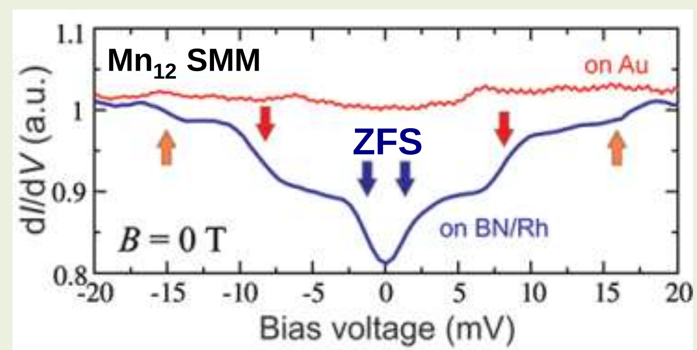


	SMM	S	T (K)	D (μeV)
Heersche *et al.*, PRL **96**, 206801 (06)	Mn_{12}	10	1.5-3	− 56
Jo *et al.*, Nano Lett. **6**, 2014 (06)	Mn_{12}	10	≲0.3	− 70-85
Burzuri *et al.*, PRL **109**, 147203 (12)	Fe_4	5	1.9	− 56

Scanning tunneling microscope



Kahle *et al.*, Nano Lett. **12**, 518 (12)



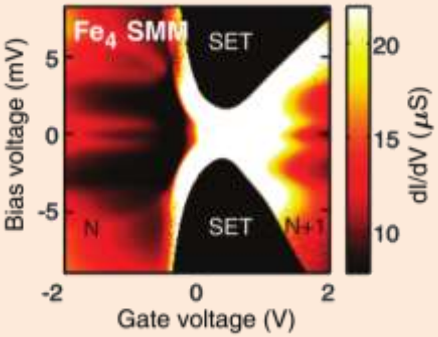
	SMM	S	T (K)
Voss *et al.*, PRB **78**, 155403 (08)	Mn_{12}	10	300

Is tuning of magnetic anisotropy possible?

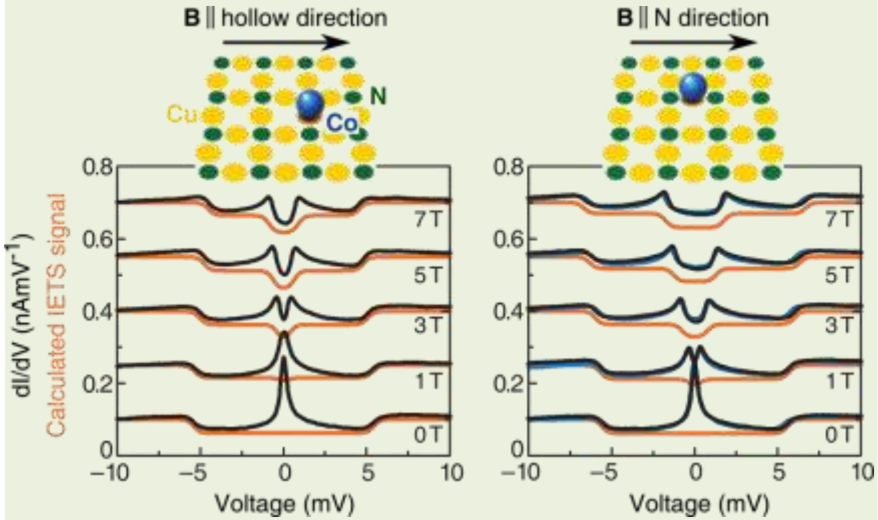
electric field

Zyazin *et al.*, Nano Lett. **10**, 3307 (10)

Burzuri *et al.*, PRL **109**, 147203 (12)

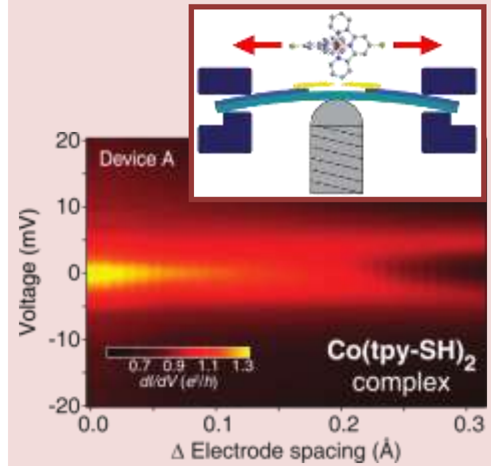


modification of local environment

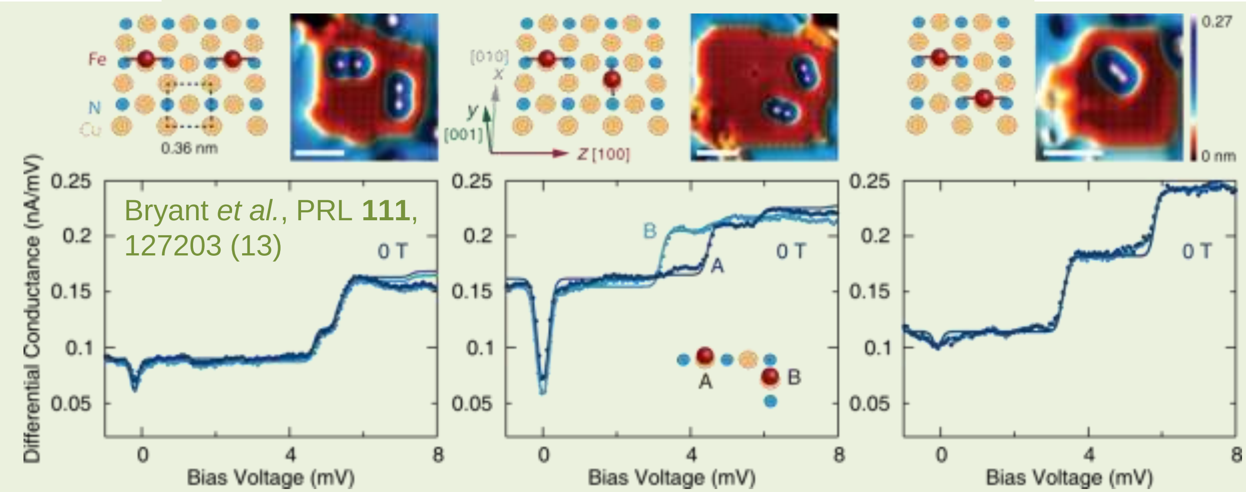


Otte *et al.*, Nature Phys. **4**, 847 (08)

mechanical straining

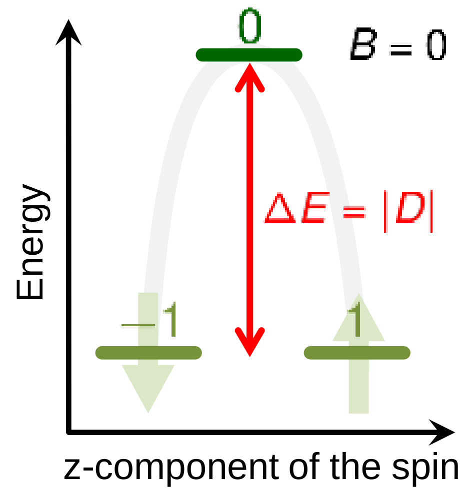


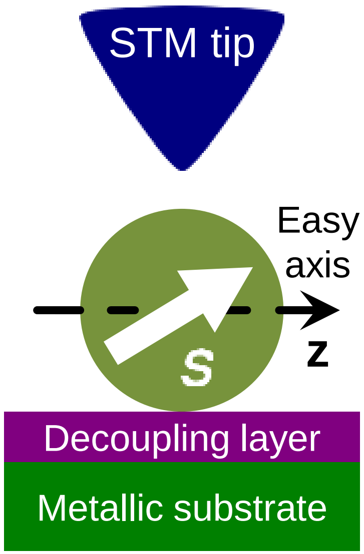
Parks *et al.*, Science **328**, 1370 (10)

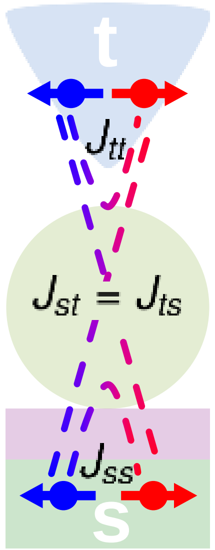


Bryant *et al.*, PRL **111**, 127203 (13)

Identification of spin-anisotropy from transport spectra







nanomagnet: the *giant-spin* Hamiltonian

$$\mathcal{H}_{\text{eff}} = BS_z + DQ_{zz}$$

$$\mathcal{H}_{\text{eff}}|m\rangle = E_m|m\rangle \quad \text{and} \quad S_z|m\rangle = m|m\rangle$$

electron tunneling processes: the Appelbaum Hamiltonian

$$\mathcal{H}_{\text{tun}} = \sum_{qkk'\alpha}\Big\{ T_{\text{d}}\, a^{q\dagger}_{k\alpha} a^{\overline{q}}_{k'\alpha} + \sum_{q'\beta} J_{qq'}\, \boldsymbol{\sigma}_{\alpha\beta}\cdot \boldsymbol{S}\, a^{q\dagger}_{k\alpha} a^{q'}_{k'\beta} \Big\}$$

direct tunneling

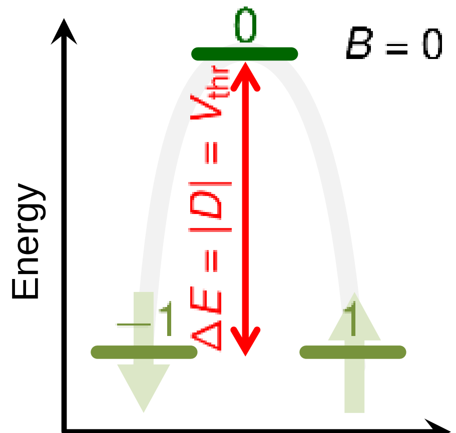
elastic/inelastic scattering

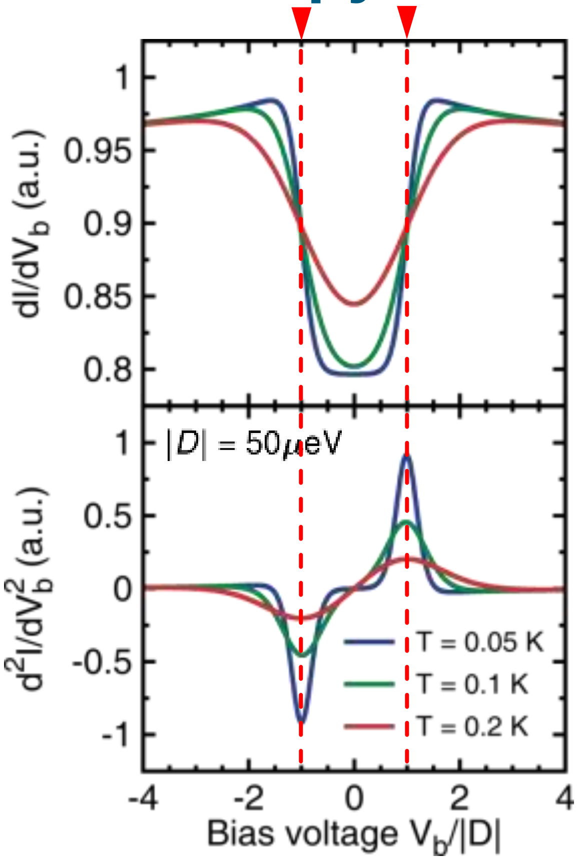
$$S_- a^{q\dagger}_{k\uparrow} a^{q'}_{k'\downarrow} + S_+ a^{q\dagger}_{k\downarrow} a^{q'}_{k'\uparrow} + S_z\left[a^{q\dagger}_{k\uparrow} a^{q'}_{k'\uparrow} - a^{q\dagger}_{k\downarrow} a^{q'}_{k'\downarrow}\right]$$

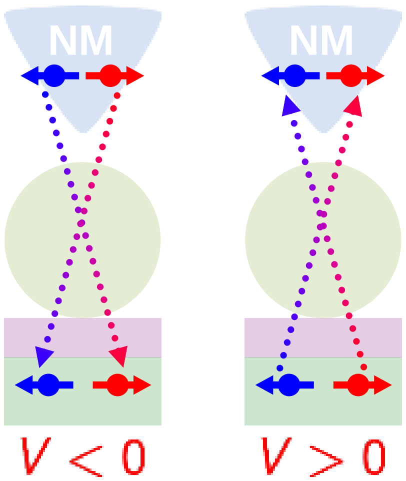
MM & Barnaś, PRB **75**, 134425 (07); PRL **111**, 046603 (13)

Identification of spin-anisotropy from transport spectra

weak coupling





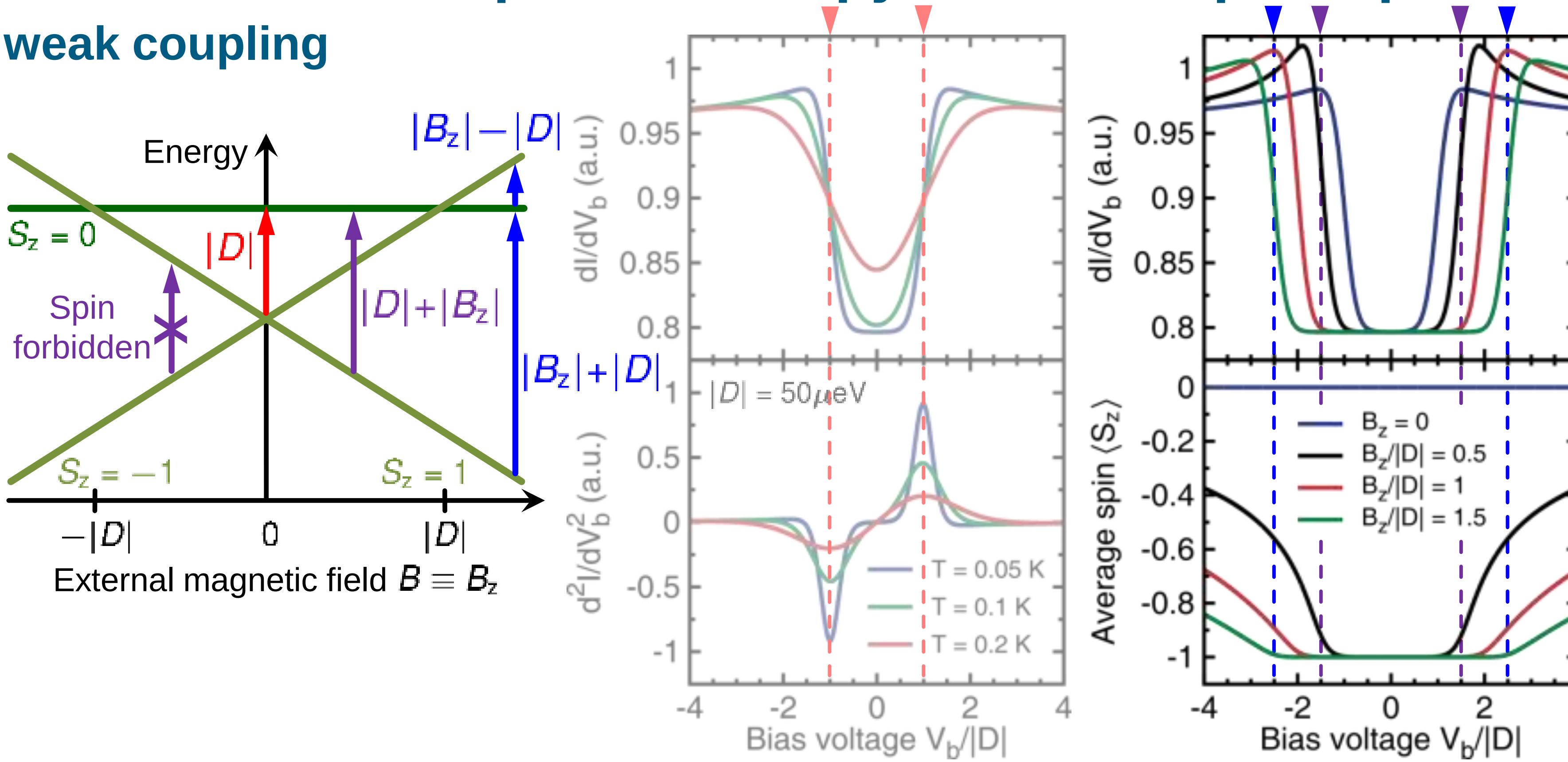


- at low T and V_b the two metastable ground states $|\pm S\rangle$ occupied with equal probabilities
- for $|V_b| \approx V_{thr} \equiv D(2S-1)$ the **inelastic** transport processes become activated
- **symmetric** inelastic conduction electron-spin scattering processes

MM & Barnaś, PRB **75**, 134425 (07); PRL **111**, 046603 (13); see also Fransson *et al.*, Bode *et al.*, Delgado *et al.*, Elste & Timm;
Experiments: Hirjibehedin *et al.*, Science **312**, 1021 (06); Jo *et al.*, Nano Lett. **6**, 2014 (06); Zyazin *et al.*, Nano Lett. **10**, 3307 (10); Kahle *et al.*, Nano Lett. **12**, 518 (12)

Identification of spin-anisotropy from transport spectra

weak coupling



- at low T and V_b the two metastable ground states $|\pm S\rangle$ occupied with equal probabilities
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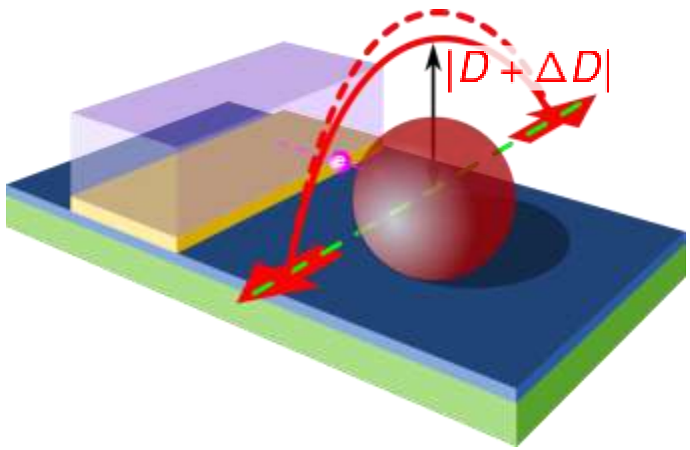
MM & Barnaś, PRB **75**, 134425 (07); PRL **111**, 046603 (13); see also Fransson *et al.*, Bode *et al.*, Delgado *et al.*, Elste & Timm;
Experiments: Hirjibehedin *et al.*, Science **312**, 1021 (06); Jo *et al.*, Nano Lett. **6**, 2014 (06); Zyazin *et al.*, Nano Lett. **10**, 3307 (10); Kahle *et al.*, Nano Lett. **12**, 518 (12)

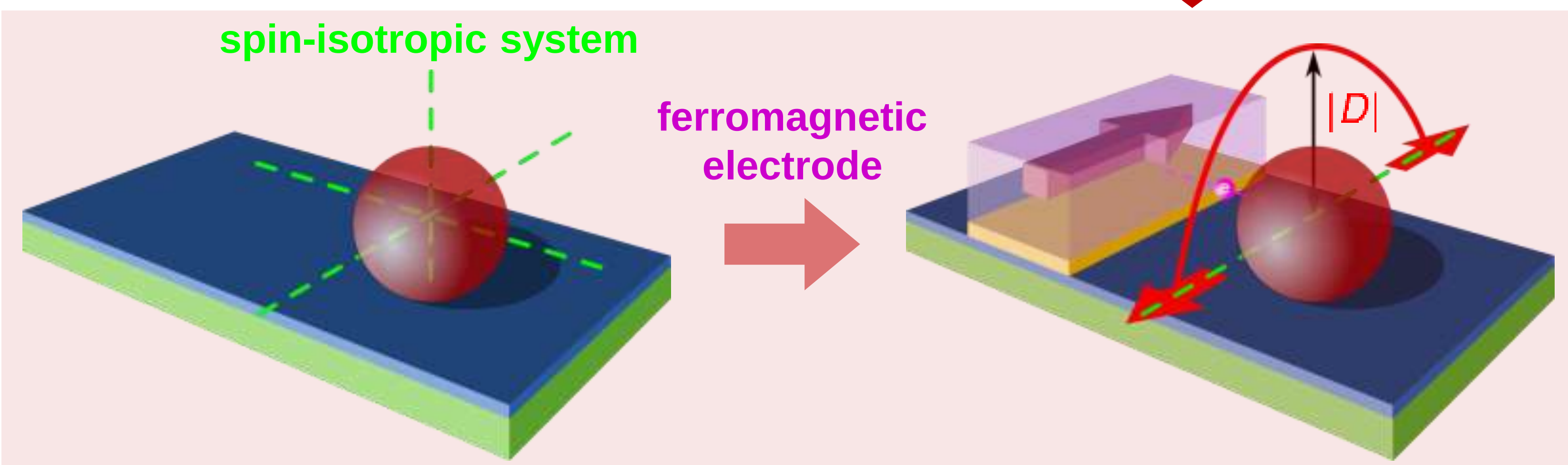
Molecular magnetism goes spintronic

- let's increase the strength of **tunnel coupling** and allow **charge fluctuation**
- **intrinsic magnetic anisotropy + charge fluctuations = anisotropy renormalization**

MM, Weymann & Barnaś, PRB **86**, 245415 (12)



Can one generate spin-anisotropy in a nanoscopic spintronic system **without** intrinsic anisotropy?



MM, Hell & Wegewijs, Nature Phys. **9**, 801 (13)

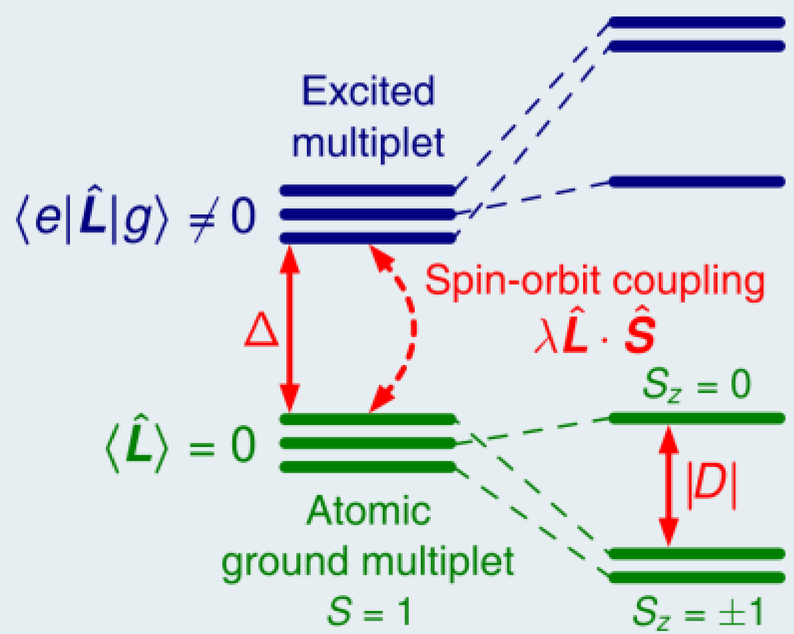
Atomic vs. spintronic origin of magnetic anisotropy

➡ Effective *giant-spin* Hamiltonian: $\mathcal{H}_{\text{eff}} = BS_z + DQ_{zz}$

z-z spin-quadrupole moment:

$$Q_{zz} = S_z^2 - \tfrac{1}{3}S(S+1)$$

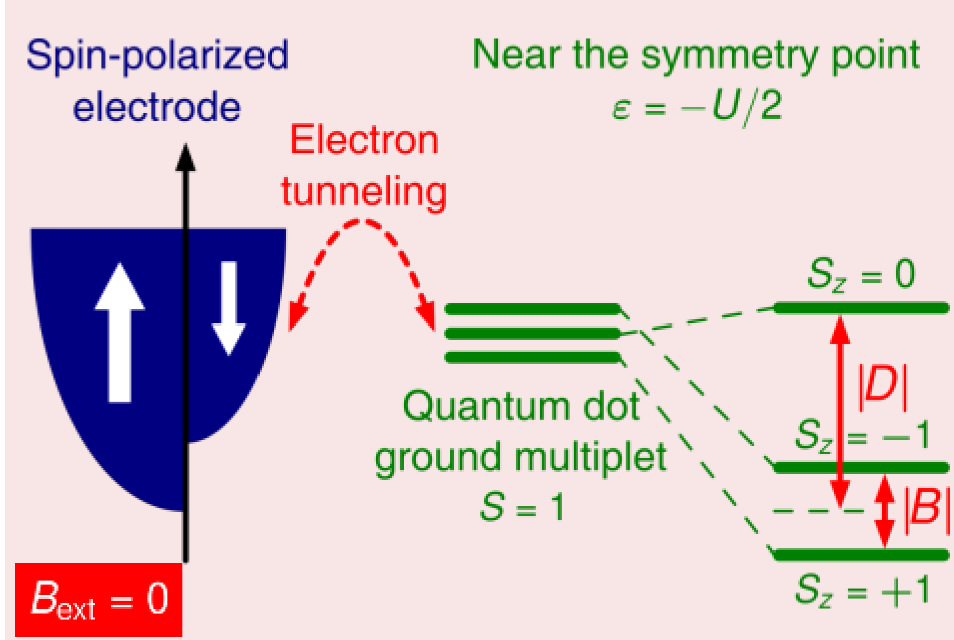
Atomic case



$$B \equiv B_{\text{ext}}$$

$$D = -\frac{3}{2}\frac{\lambda^2|\langle g|L_z|e\rangle|^2}{\Delta}$$

Spintronic case

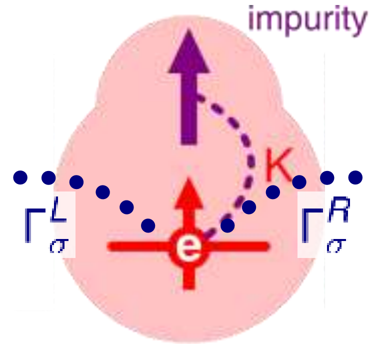


Spintronic exchange fields

- **DIPOLAR** B (known) Martinek *et al.*, PRL **91**, 127203 (03); PRB **72**, 121302 (05)
- **QUADRUPOLAR** D (**NEW**!)

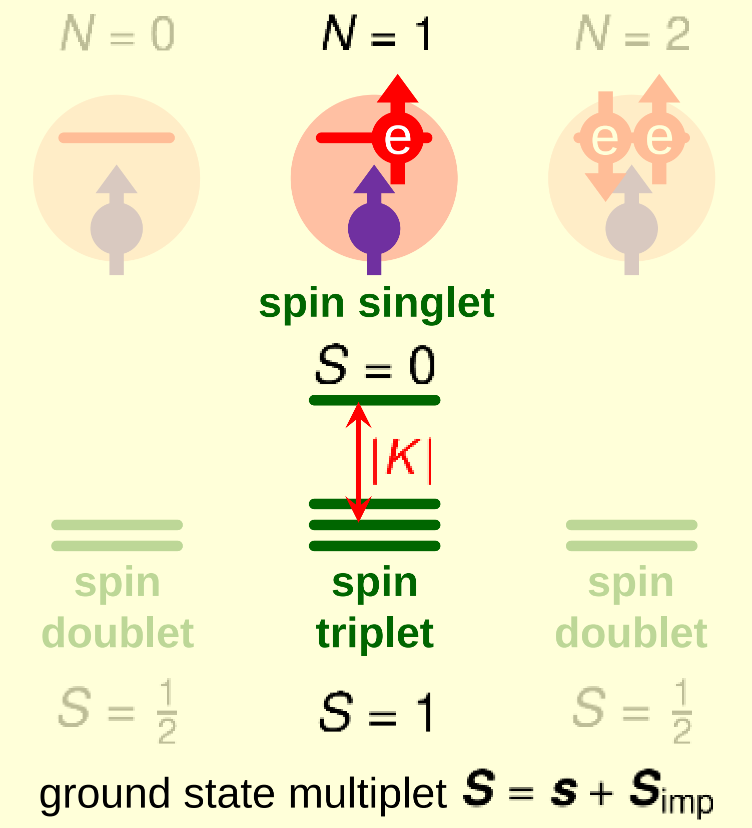
MODEL: high-spin quantum-dot spin valve

Starting point: spin-isotropic system!!!



ferromagnetic exchange coupling

$$\mathcal{H}_{\rm dot} = \varepsilon \sum_\sigma n_\sigma + U n_\uparrow n_\downarrow + K \boldsymbol{s} \cdot \boldsymbol{S}_{\rm imp}$$

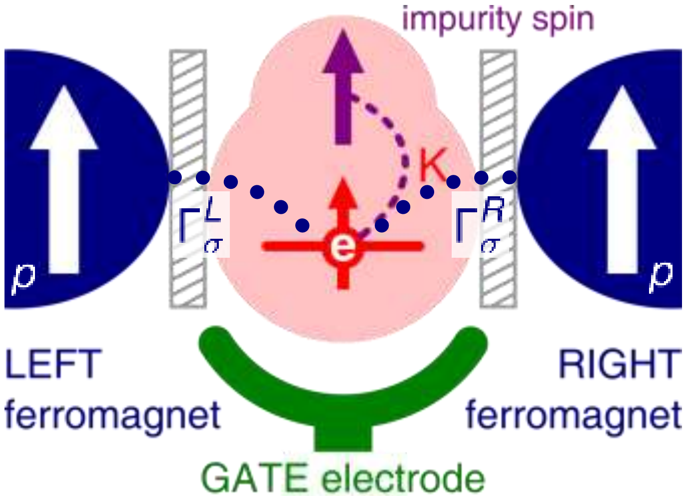


electron tunneling processes $\mathcal{H}_{\rm tun} = \sum_{qk\sigma} \sqrt{\frac{\Gamma^q_\sigma}{\pi \nu_\sigma}}\, d^\dagger_\sigma a^q_{\boldsymbol{k}\sigma} + {\rm H.c.}$

Focus on $N=1$, $S=1$ state: fluctuations into virtual $N=0,2$ lead to renormalization

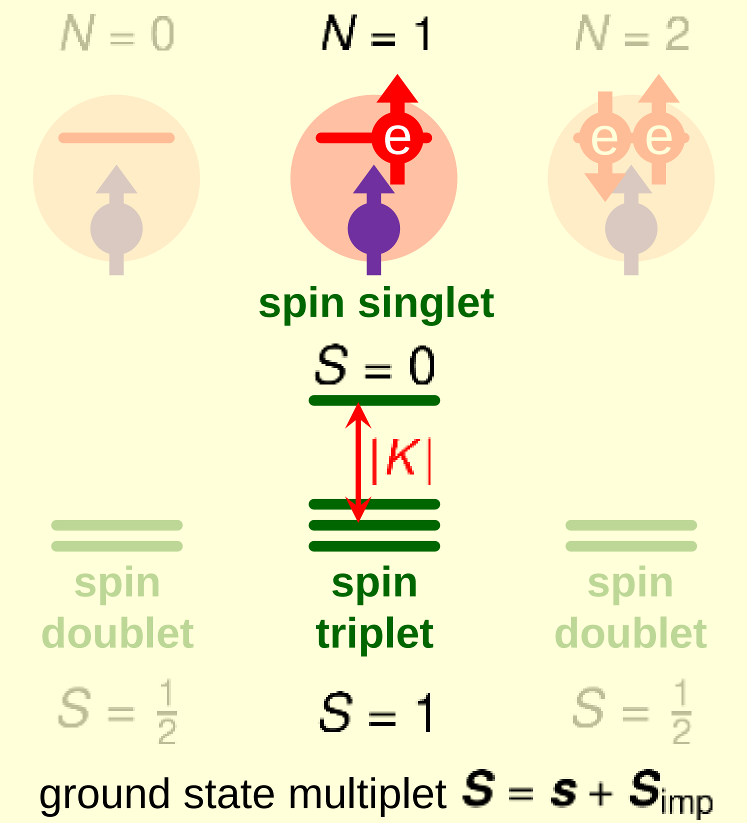
MODEL: high-spin quantum-dot spin valve

Starting point: spin-isotropic system!!!



ferromagnetic exchange coupling

$$\mathcal{H}_{\text{dot}} = \varepsilon \sum_\sigma n_\sigma + U n_\uparrow n_\downarrow + K \boldsymbol{s} \cdot \boldsymbol{S}_{\text{imp}}$$



➡ **electron tunneling processes** $\mathcal{H}_{\text{tun}} = \sum_{q\boldsymbol{k}\sigma} \sqrt{\frac{\Gamma_\sigma^q}{\pi \nu_\sigma}} d_\sigma^\dagger a_{\boldsymbol{k}\sigma}^q + \text{H.c.}$

➡ **IDEA:** electrons localized in a high-spin ($S \geqslant 1$) **spin-isotropic** quantum dot probe the broken spin symmetry in attached ferromagnets by **virtual charge fluctuations**

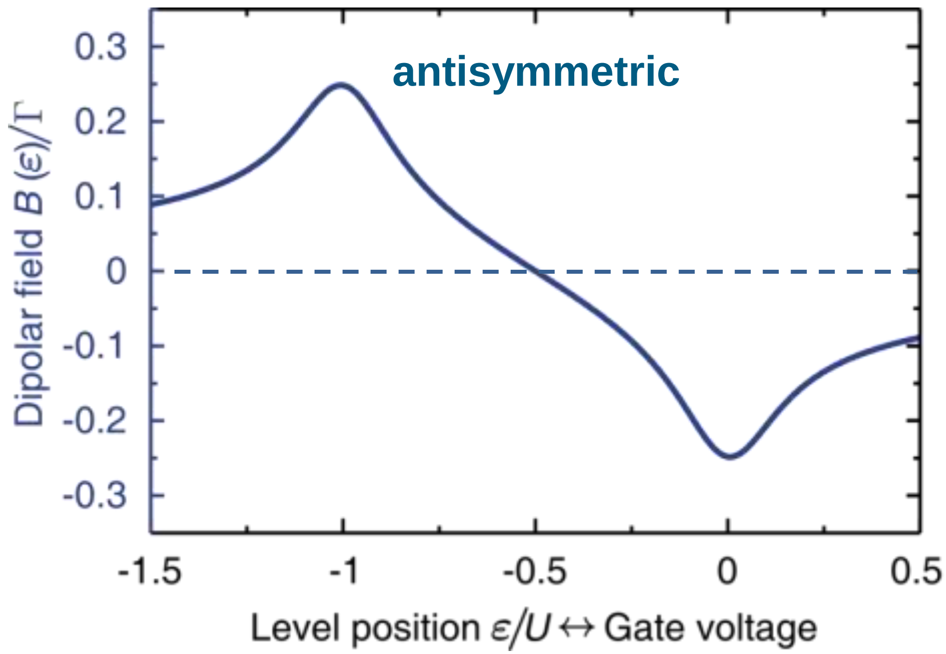
Dipolar vs. quadrupolar exchange field

$$\mathcal{H}_{\text{eff}} = BS_z + DQ_{zz}$$

Perturbation theory $\Gamma \ll T$

$B = B_0 - B_2$ → $B \propto p\Gamma$

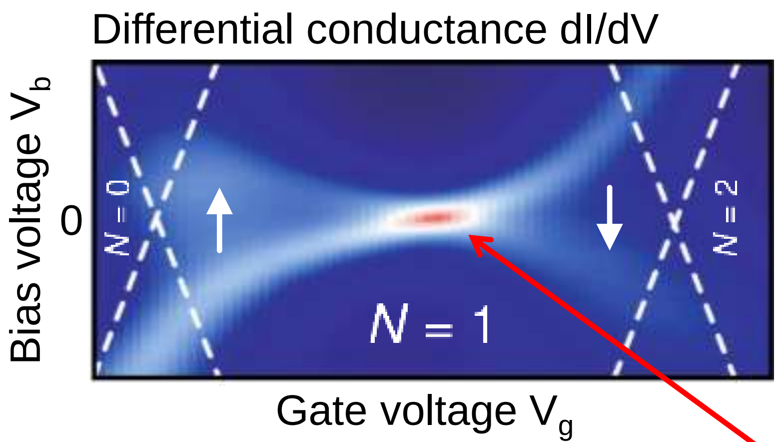
$$B_n = \sum_{q=L,R} \mathcal{P}\int \frac{d\omega}{2\pi} \frac{p_q \Gamma_q f(\omega)}{\omega - \varepsilon - nU/2}$$



Martinek et al., PRL **91**, 127203 (03); PRB **72**, 121302 (05); König et al., Lec. Notes Phys. **658**, 145 (05)

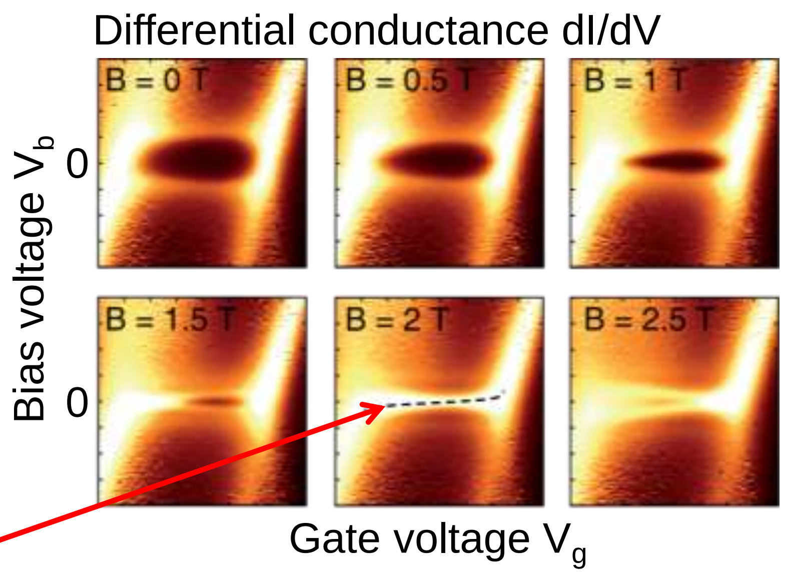
Dipolar vs. quadrupolar exchange field
Experimental evidences of dipolar field in CNT QDs

Gate-tunable



Hauptman *et al.*, Nature Phys. **4**, 373 (08)

Magnetic field-tunable $B_{ext} \neq 0$



restored Kondo effect

Gaass *et al.*, PRL **107**, 176808 (11)

Dipolar vs. quadrupolar exchange field

$$\mathcal{H}_{\text{eff}} = BS_z + DQ_{zz}$$

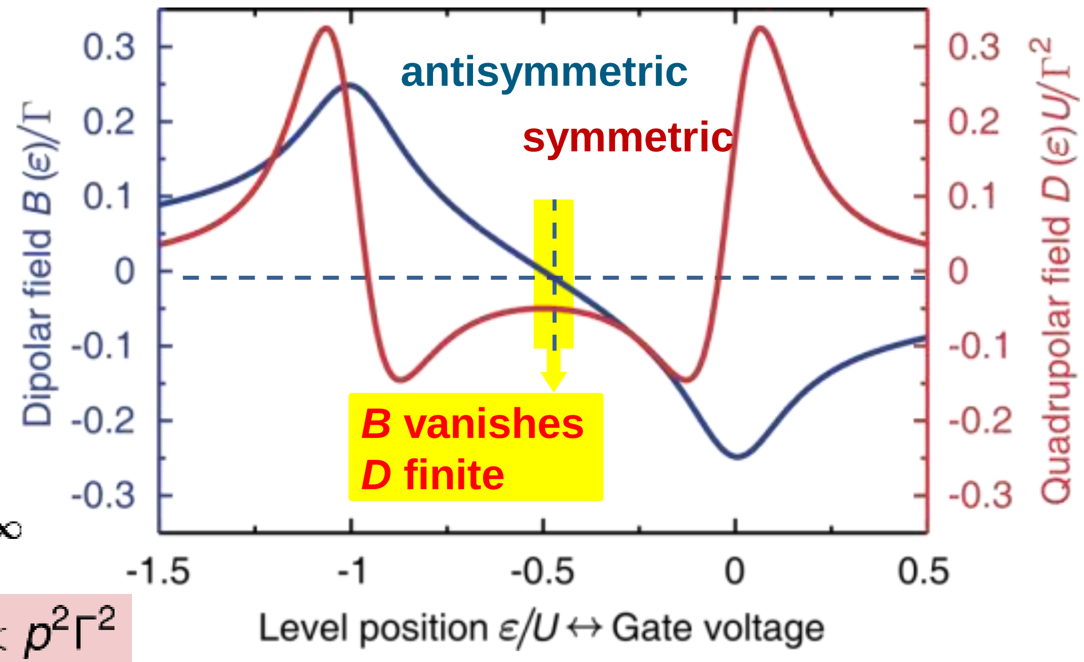
Perturbation theory $\Gamma \ll T$

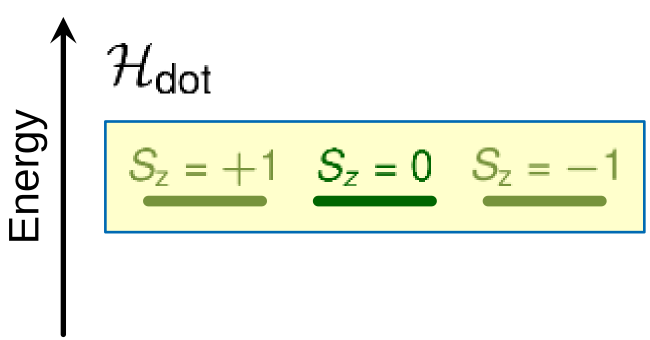
$$B = B_0 - B_2 \;\rightarrow\; B \propto p\Gamma$$

$$B_n = \sum_{q=L,R} \mathcal{P} \int \frac{d\omega}{2\pi} \frac{p_q \Gamma_q f(\omega)}{\omega - \varepsilon - nU/2}$$

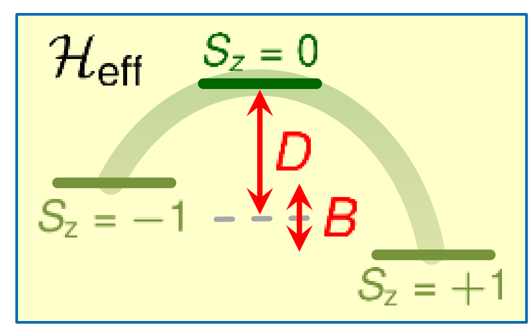
deep in Coulomb blockade regime & $K \to \infty$

$$D \approx -B_0 \frac{dB_0}{d\varepsilon} - B_2 \frac{dB_2}{d(-\varepsilon)} \;\rightarrow\; D \propto p^2\Gamma^2$$





attach ferromagnets



MM, Hell & Wegewijs, Nature Phys. **9**, 801 (13)

Quadrupolar exchange field

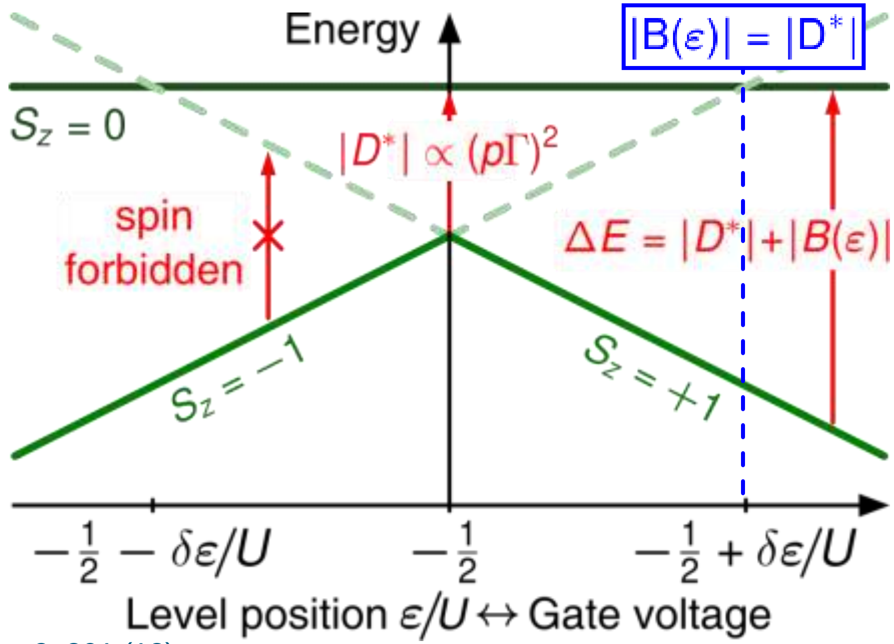
Strong coupling regime $\Gamma \gg T$

- **all-electric superparamagnet** has the characteristic properties of a **spintronic exchange field**: **electric tunability** and **scaling with tunnel-coupling (Γ) and spin-polarization (p)**

Dipolar exchange field — **Quadrupolar** exchange field

$$B \propto p\Gamma \quad \text{vs.} \quad D \propto p^2\Gamma^2$$

- **IDEA:** tune gate voltage to eliminate dipolar exchange field



$$T \ll U \ll W$$

$$D^* \equiv -\frac{1}{\pi^2}\frac{(p\Gamma)^2}{U}\ln\frac{2W}{U}$$

$$B(\varepsilon) \approx -\frac{2}{\pi}p\Gamma\left(\frac{\varepsilon}{U} + \frac{1}{2}\right)$$

$$\delta\varepsilon = \frac{\pi}{2}\frac{|D^*|U}{p\Gamma}$$

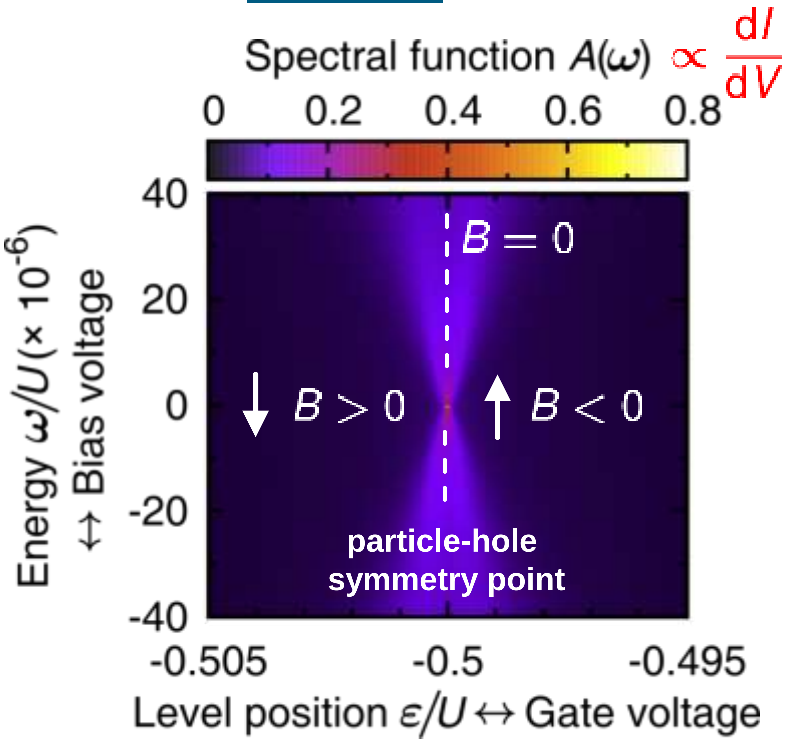
MM, Hell & Wegewijs, Nature Phys. **9**, 801 (13)

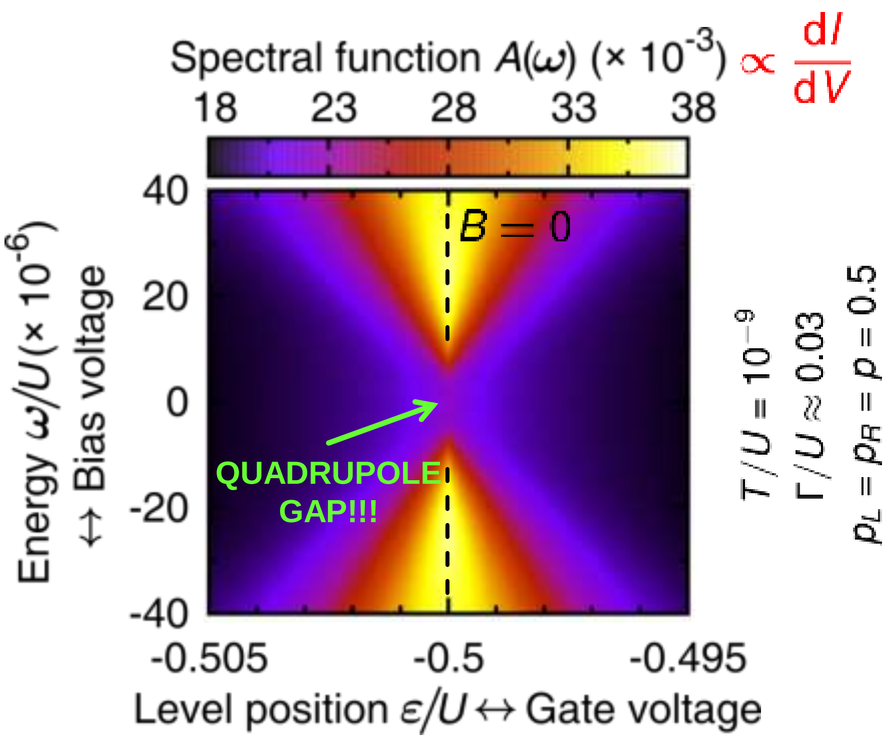
Low- vs. high-spin quantum dots

$$A(\omega) = 2\pi \sum_\sigma \Gamma_\sigma \frac{a_\sigma(-\omega) + a_\sigma(\omega)}{2}$$

$S = \frac{1}{2}$ → $K = 0$

$S = 1$ → finite exchange K

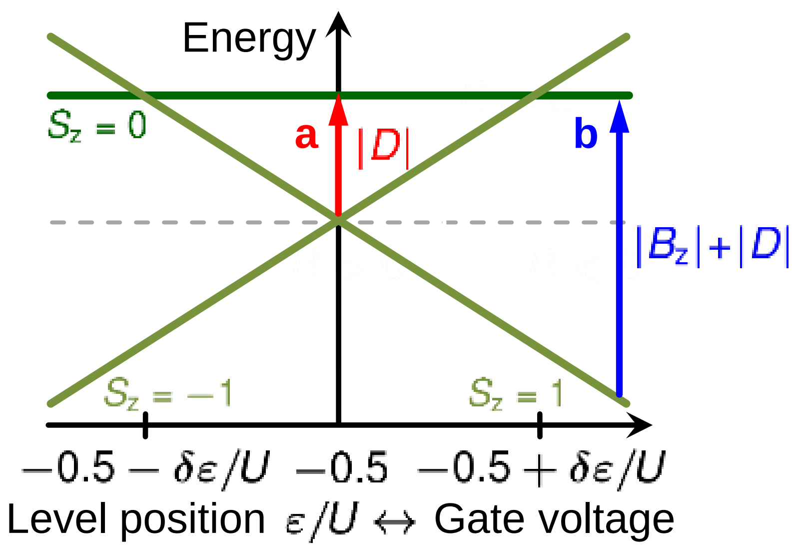


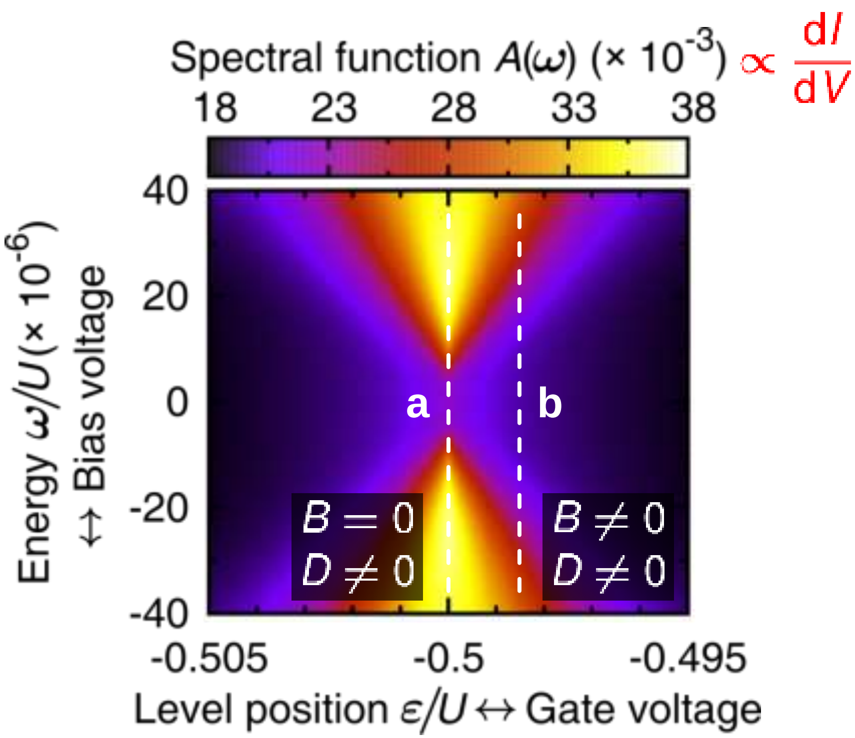


Numerical renormalization grioup (NRG):

K. Wilson, RMP **47**, 773 (75); R. Bulla et al., RMP **80**, 395 (08); Weichselbaum & von Delft, PRL **99**, 076402 (07); Toth et al., PRB **78**, 245109 (08); Budapest fDM-NRG code – O. Legeza et al., arXiv:0809.3143 (08)

Spectroscopic signatures of quadrupolar field

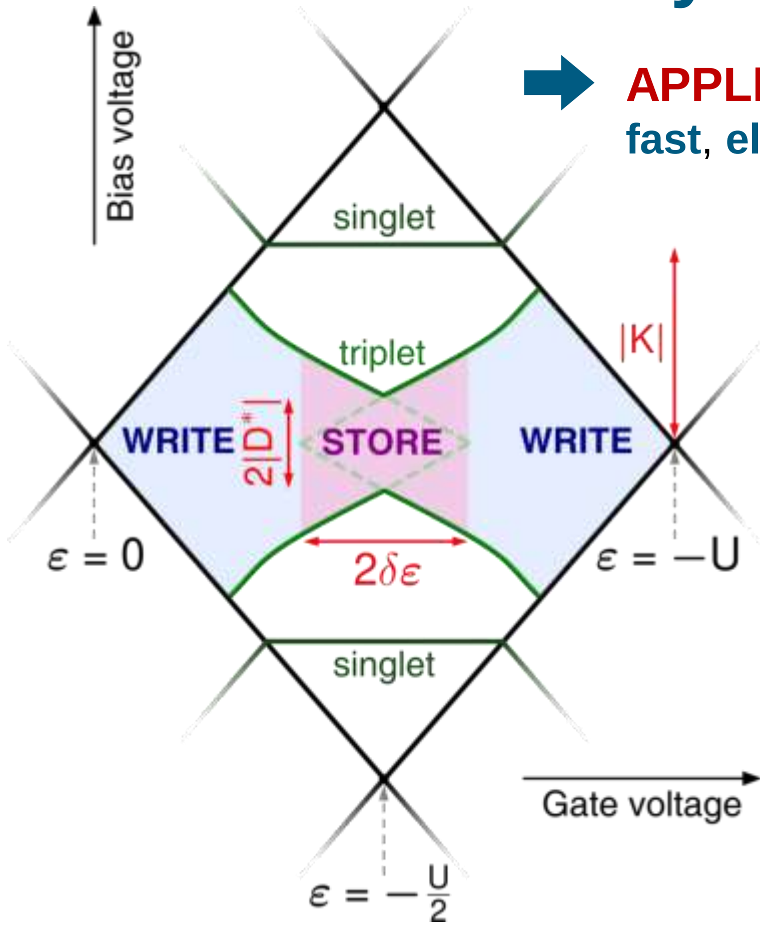




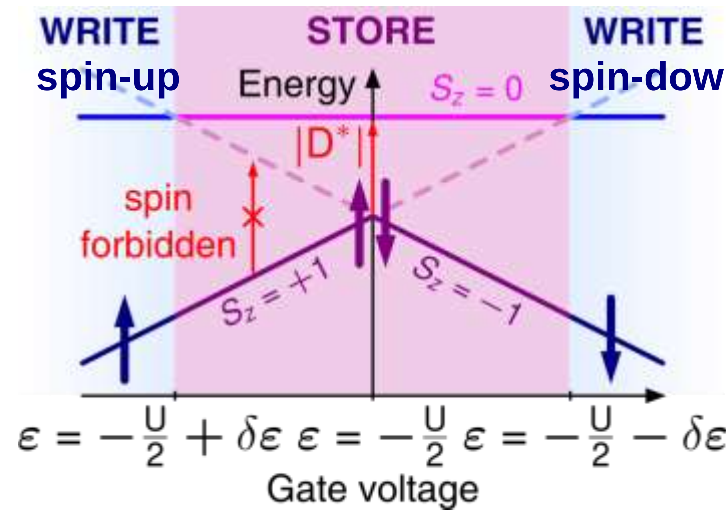
➡ inelastic cotunneling signatures as for **SMM** or **magnetic adatom**, but **NO intrinsic anisotropy**

Nature Phys. **9**, 801 (13);
Experiments with *intrinsic* anisotropy signatures: Hirjibehedin *et al.*, Science **312**, 1021 (06); Jo *et al.*, Nano Lett. **6**, 2014 (06); Zyazin *et al.*, Nano Lett. **10**, 3307 (10); Kahle *et al.*, Nano Lett. **12**, 518 (12)

On-demand bistability: writing and storing spin



➡ **APPLICATION:** quadrupolar field as a new means for **fast**, **electrical control** of nanomagnetic **memory cells**



$$D^* \approx -\frac{1}{\pi^2}\frac{(\rho\Gamma)^2}{U}\ln\frac{2W}{U} \qquad \delta\varepsilon = \frac{\pi}{2}\frac{|D^*|U}{\rho\Gamma}$$

➡ the spin can be switched by **electrically tuning** from the neutral **'STORE'** regime to **'WRITE spin-up'** or **'WRITE spin-down'** regime, returning then again to the **'STORE'** regime where $|B| \ll |D^*|$

➡ robust against perturbations on energy scales $< |D^*|$

MM, Hell & Wegewijs, Nature Phys. **9**, 801 (13)

Conclusions and outlook

- nanoscopic systems with uniaxial magnetic anisotropy = **magnetic bistability**
  - building blocks for a memory cell
  - spin-polarized currents for manipulation of the magnetic state of the system

- proximity-induced **spin-anisotropy** in high-spin **spin-isotropic system**

  KEY IDEA: spin-anisotropy = spintronic quantity

- tunnel-induced exchange fields:
  - **DIPOLAR** ~ *pseudo* magnetic field ~ spin torque
  - **QUADRUPOLAR** ~ spin-anisotropy tensor *a la* molecular magnetism

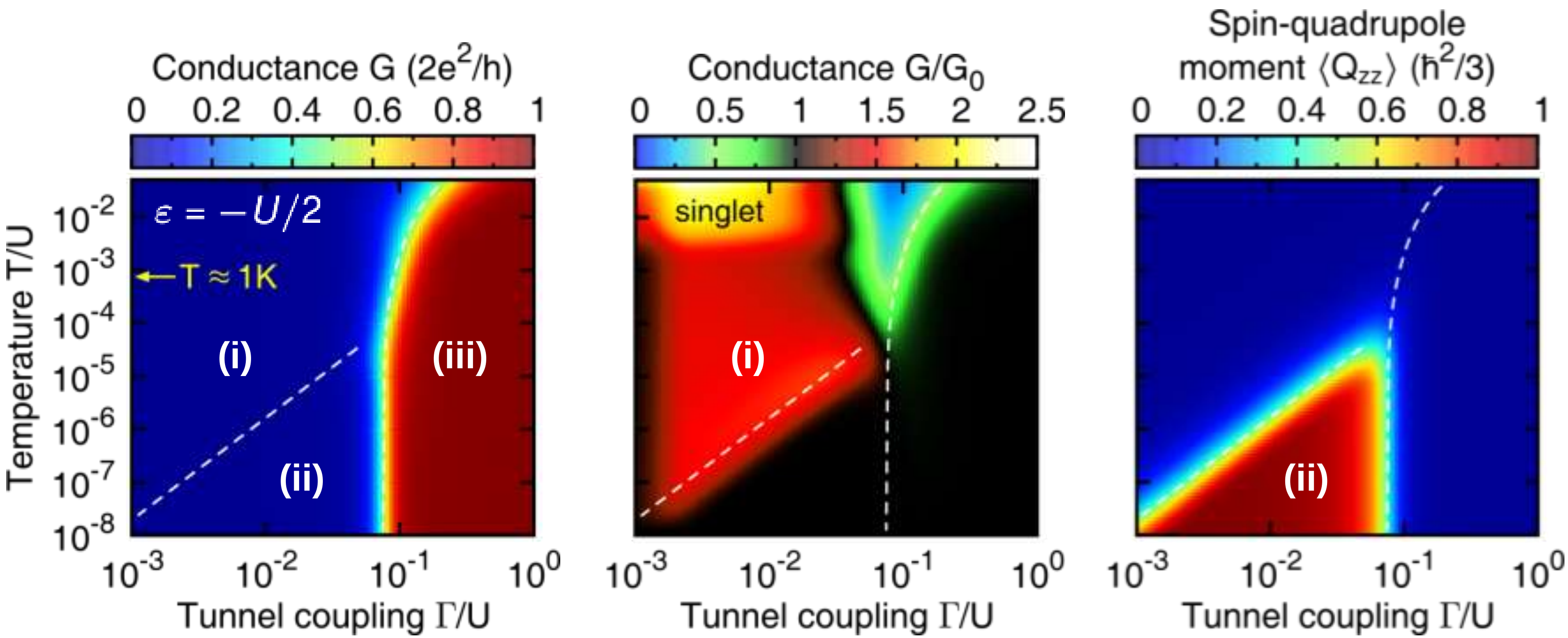
- scaling of cotunneling gap with tunnel coupling Γ and spin polarization p

- full **electric tunability** of all terms in magnetic Hamiltonian

For details see: Misiorny, Hell & Wegewijs, Nature Phys. **9**, 801 (13)
Misiorny & Barnaś, PRL **111**, 046603 (13)
Misiorny, Weymann & Barnaś, PRB **86**, 245415 (12)
Hell, Das & Wegewijs, PRB 88, 115435 (13)

Thank you for your attention

Competition between spintronic anisotropy and Kondo effect



- **(i)** **paramagnetic regime** — $T \gtrsim |D^*(\Gamma)|$ and $T_K(\Gamma, D^*(\Gamma))$
- **(ii)** **superparamagnetic regime** — $|D^*(\Gamma)| \gtrsim T$ and $T_K(\Gamma, D^*(\Gamma))$
- **(iii)** **Kondo screened regime** — $T_K(\Gamma, D^*(\Gamma)) \gtrsim |D^*(\Gamma)|$