Universal mechanism for electron paramagnetic resonance of individual adatoms

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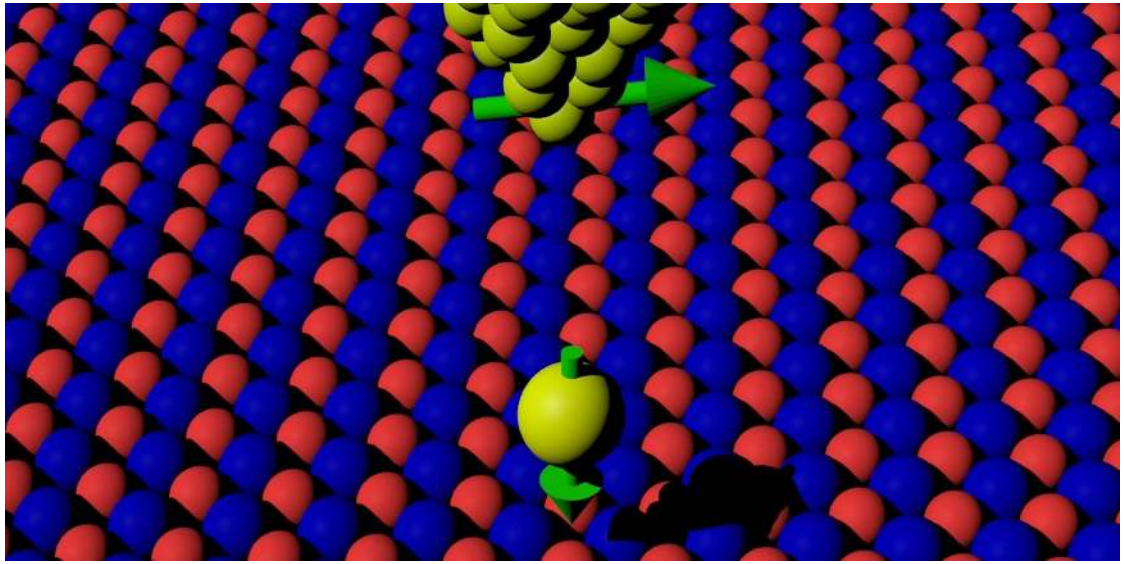
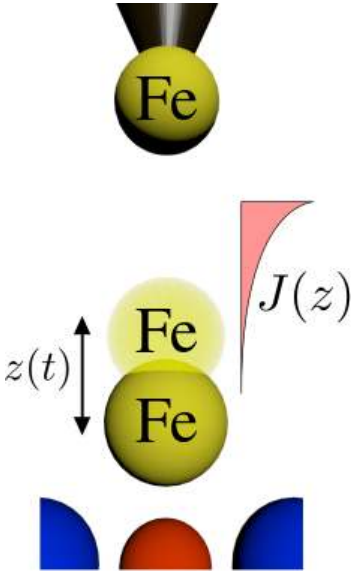




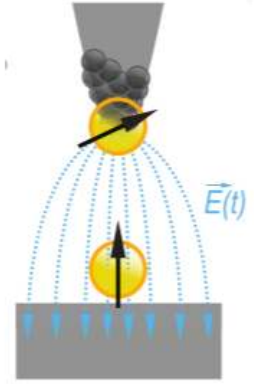
arXiv:1611.01110

APS March Meeting New Orleans 2017

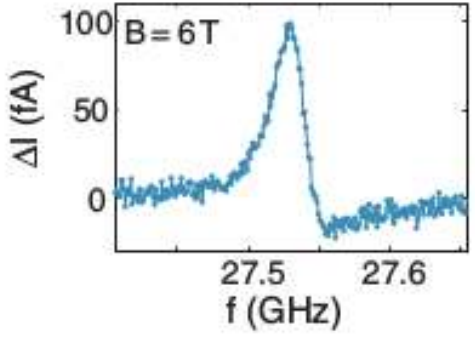
Experiment ESR-STM, resonance in a single Fe atom

Fe in MgO, S. Baumman et al, Science 2015

Detect spin excitations with an STM by resonance with an electrical signal



DC Current VS RF frequency



$$P_0 - P_1 = \tanh\left(\frac{\hbar\omega_0}{2k_BT}\right)\left(1 - \frac{\Omega_0^2 T_1 T_2}{1 + (\omega - \omega_0)^2 T_2^2 + \Omega_0^2 T_1 T_2}\right)$$

Ultimate energy resolution

Outperforms IETS by three orders of magnitude

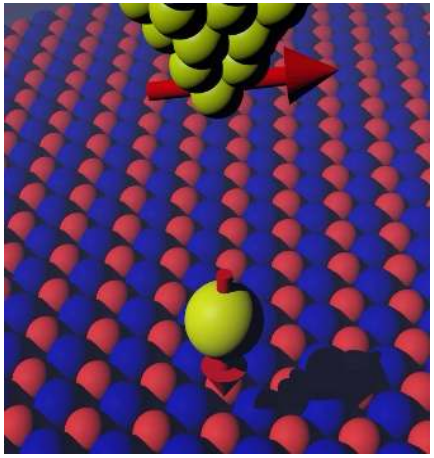
Pushes ESR down to the atomic limit

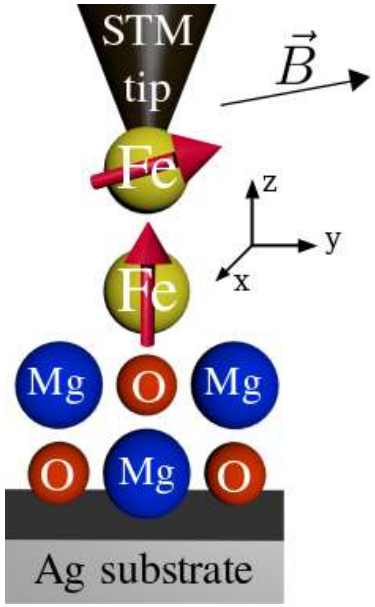
New spectroscopic technique

- T. Choi et al, Nat. Nano 2017
- F. Natterer et al, Nature 2017

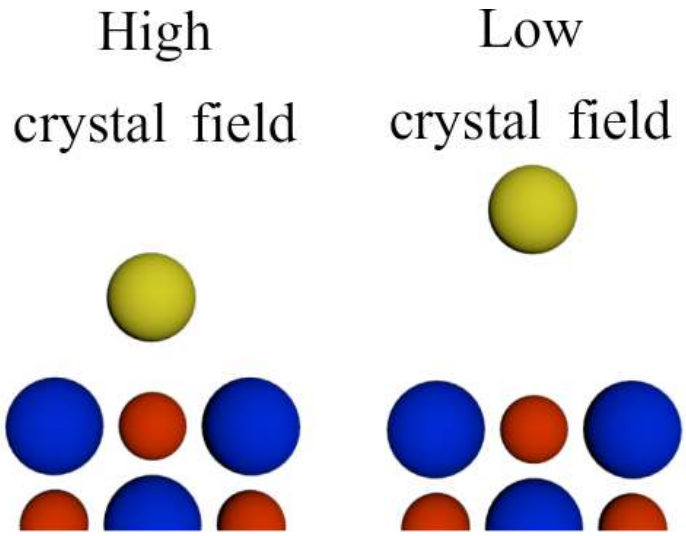
How to drive a spin excitation with a RF electrical signal?

Mechanism proposed in the experimental paper





Modulation of the crystal field

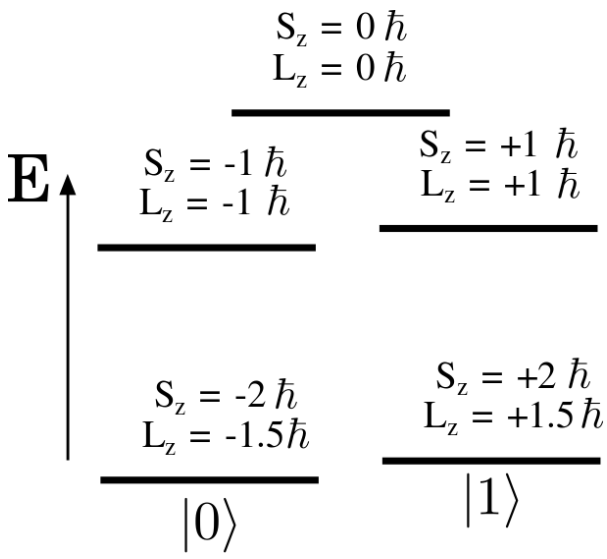


Spin Hamiltonian $\mathcal{H}(z) = DL_z^2 + F(L_+^4 + L_-^4) + \Lambda \vec{L} \cdot \vec{S} + \mu_B \vec{B} \cdot \left(\vec{L} + 2\vec{S}\right)$

Electrically modulated crystal field + spin orbit coupling → Resonance with spin excitations

ESR-STM, Fe Hamiltonian

Fe at MgO, spin model

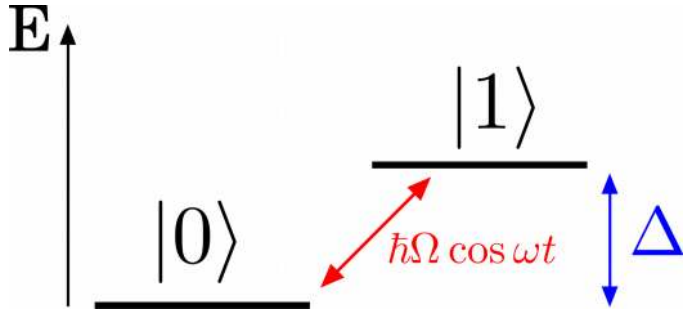


S. Baumman et at, PRL 2015

Time dependent two level Hamiltonian

$$\mathcal{H}_{\text{eff}} = \begin{pmatrix} \frac{\Delta}{2} & \hbar\Omega\cos\omega t \\ \hbar\Omega\cos\omega t & -\frac{\Delta}{2} \end{pmatrix}$$

Effective low energy model



Driven transitions between the two levels when $\hbar\omega = \Delta$

Ingredients for ESR with STM

Crystal field Rabi coupling, problems

Does not work for S=1/2 Needs very specific chemical environment

The question

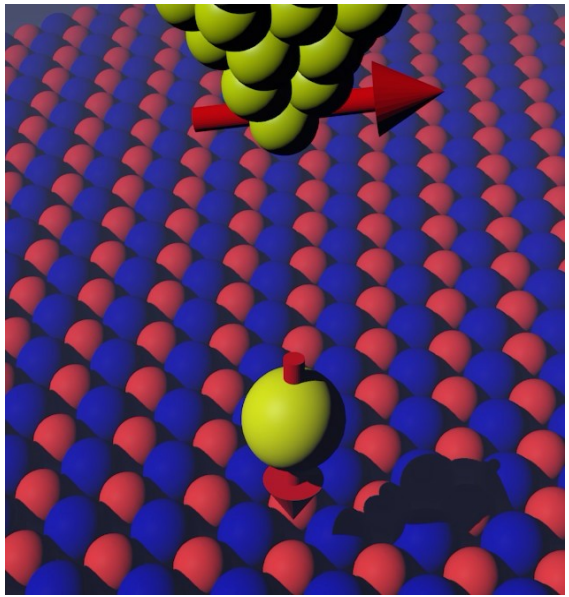
Is there a mechanism that allows to perform ESR for **any spin** in **any surface**?

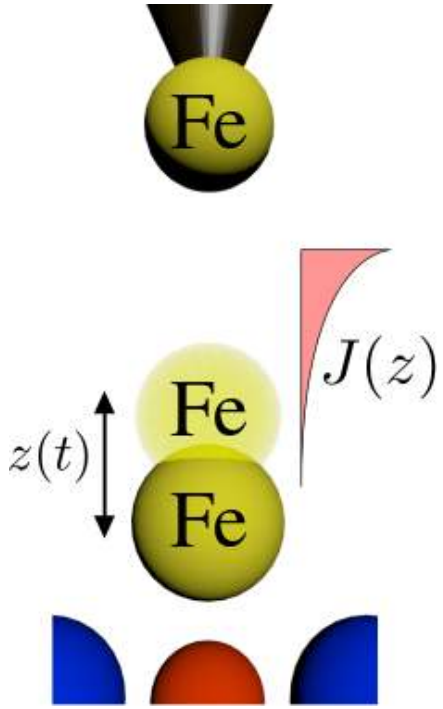
How to change a spin Hamiltonian with an electric field?

The prize

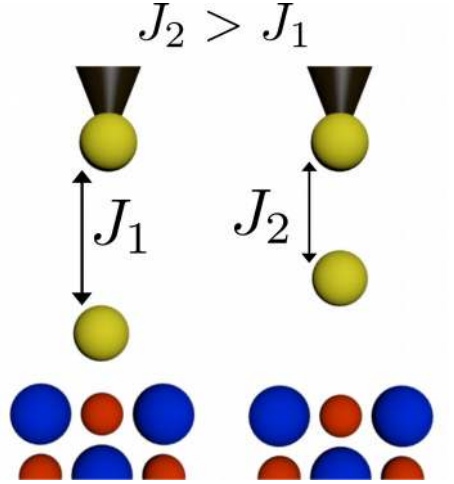
Atomic and ultimate energy resolution for any magnetic atom

Universal excitation mechanism, exchange coupling





Tip-atom **exchange** change with the electrical signal



Electrical modulation of exchange coupling

Electrically modulated position + exchange coupling → Resonance with spin excitations

Hamiltonian for Fe@MgO

Total Hamiltonian for Fe at MgO

Method: DFT + Wannier + Configuration interaction

$$\mathcal{H} = H_{\mathrm{CF}} + H_{\mathrm{SOC}} + H_{\mathrm{Coulomb}} + H_{\mathrm{Zeeman}} + H_{\mathrm{Dip}} + H_{\mathrm{J}}$$

Crystal field

Atomic Coulomb interaction

Dipolar coupling between tip and Fe

Spin-orbit coupling

External magnetic field

Exchange coupling between tip and Fe

Three possible mechanisms for spin driving with the electric signal

Rabi mechanism in a time dependent Hamiltonian

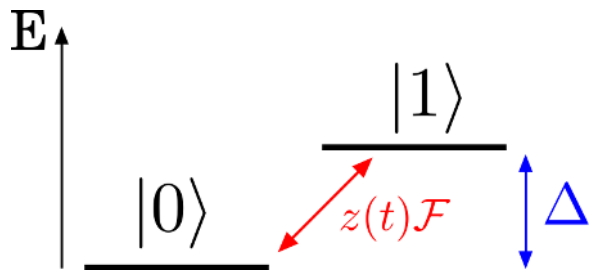
Derivation of the Rabi Hamiltonian

Time dependent Hamiltonian $\mathcal{H} \approx \mathcal{H}_0 + z(t)\left.\frac{\partial \mathcal{H}}{\partial z}\right|_{z=0}$

Effective Hamiltonian $\mathcal{H}_{\text{eff}} = \begin{pmatrix} \frac{\Delta}{2} & \hbar\Omega\cos\omega t \\ \hbar\Omega\cos\omega t & -\frac{\Delta}{2} \end{pmatrix}$

Rabi Force $\mathcal{F} = \langle 0|\frac{\partial \mathcal{H}}{\partial z}|1\rangle$

Two level system



Efficiency of the modulated spin mixing

Rabi frequency ⟶ $\Omega = \frac{\mathcal{F}}{\hbar} z$ ⟵ **Rabi force**

⟵ **Displacement of Fe**

Rabi forces of the three mechanisms

Rabi frequency

$$\Omega = \frac{\mathcal{F}}{\hbar} z$$

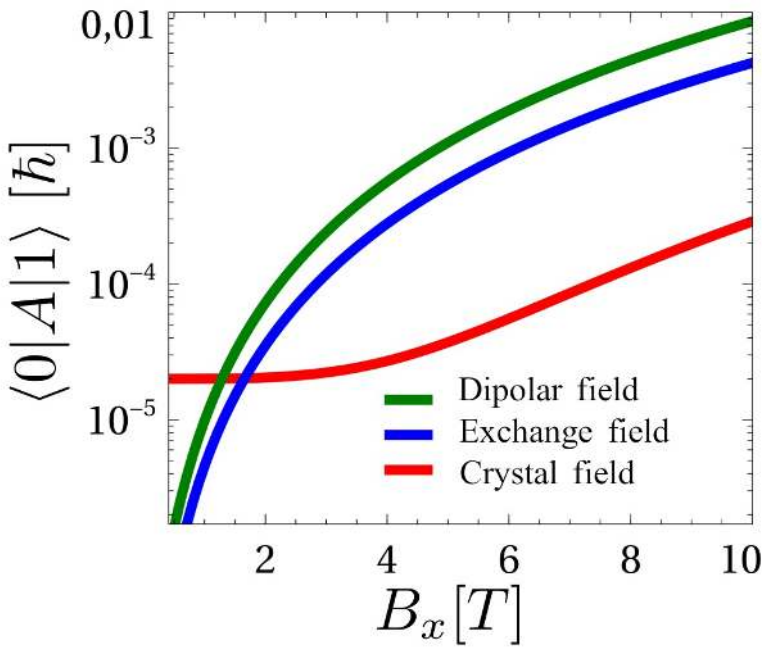
Rabi forces

$$\mathcal{F}_J = \frac{\partial J(z)}{\partial z} \langle \vec{S}_T \rangle \cdot \langle 0 | \vec{S} | 1 \rangle$$

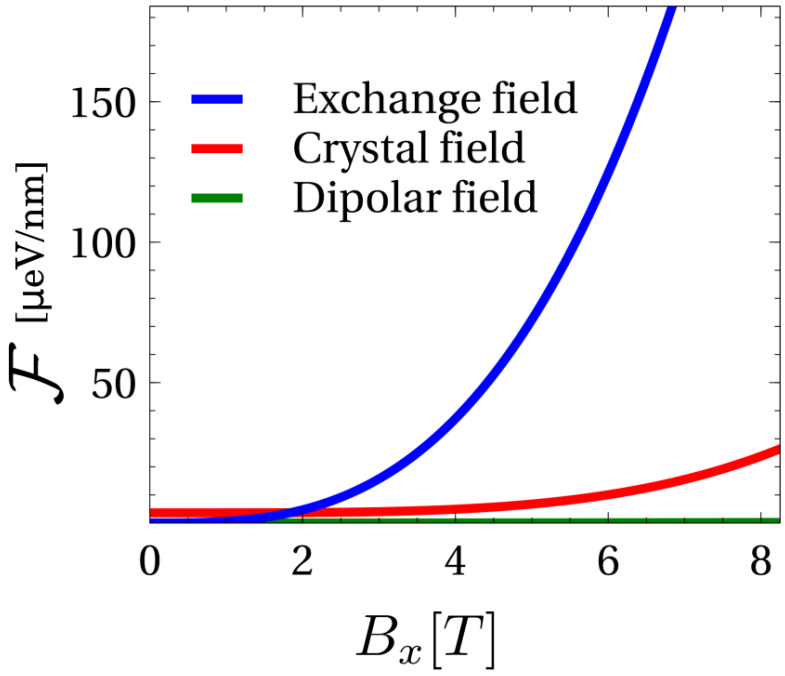
$$\mathcal{F}_{\text{dip}} = \mu_B \frac{\partial B_x^{\text{Tip}}}{\partial z} \langle 0 | L_x + 2S_x | 1 \rangle$$

$$\mathcal{F}_{CF} = \frac{\partial F_W}{\partial z} \langle 0 | l_x^4 + l_y^4 | 1 \rangle$$

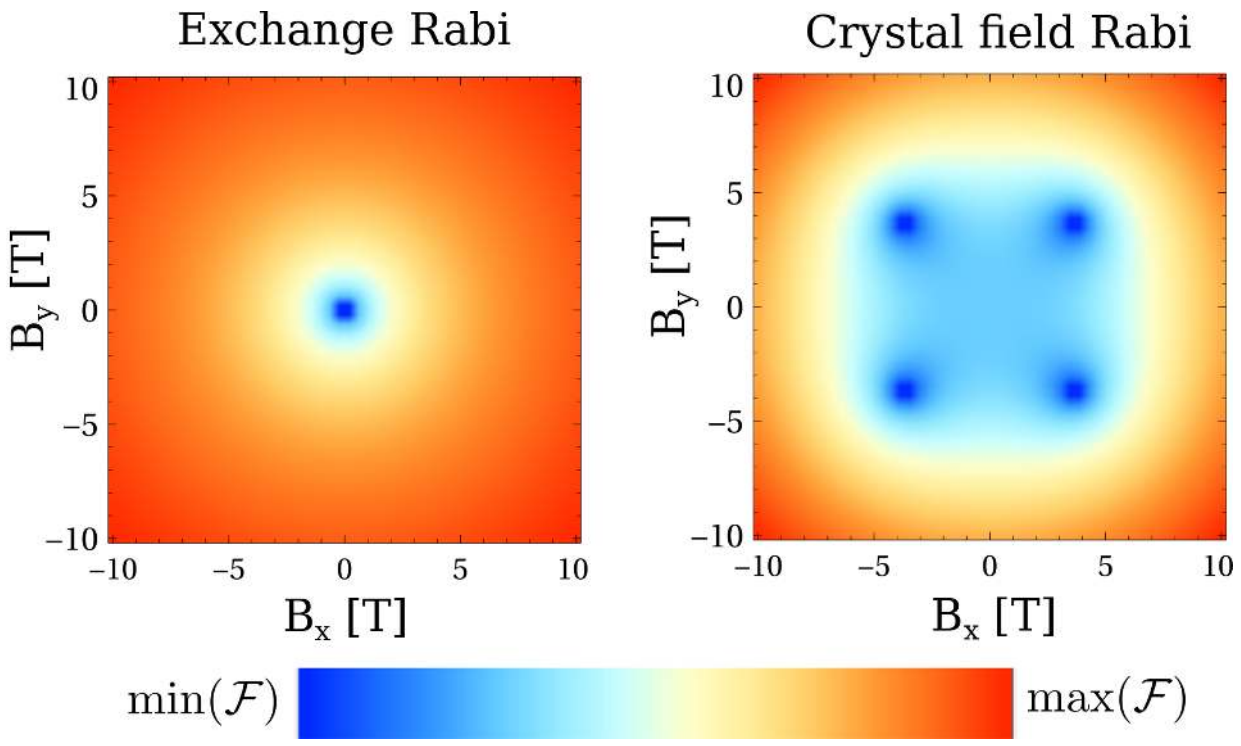
Matrix elements

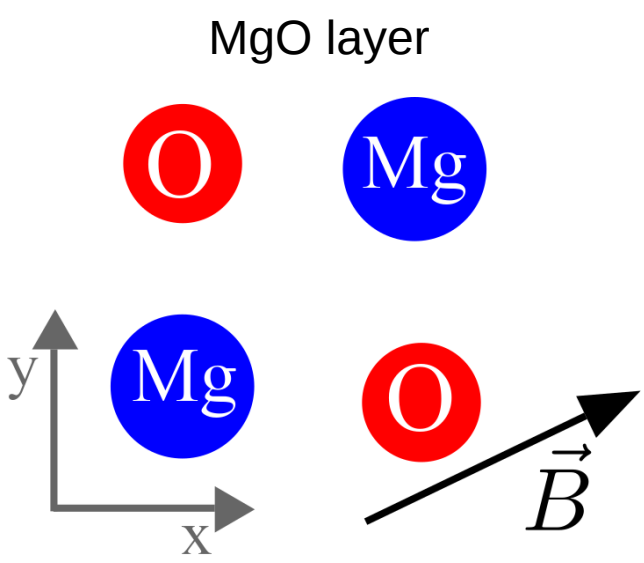


Rabi forces



How to test it? Symmetry of the Rabi efficiency





Exchange mechanism is equally efficient for any alignment between magnetic field and MgO

Take home

Modulation of the exchange coupling between tip and atom provides a universal mechanism for electron paramagnetic resonance with STM

Allows to apply ESR-STM to light atoms and S=1/2

Acknowledgement

Marie Curie ITN

Spinograph
Spintronics in Graphene

Thank you!!!

Estimate of the atomic motion

Harmonic approximation

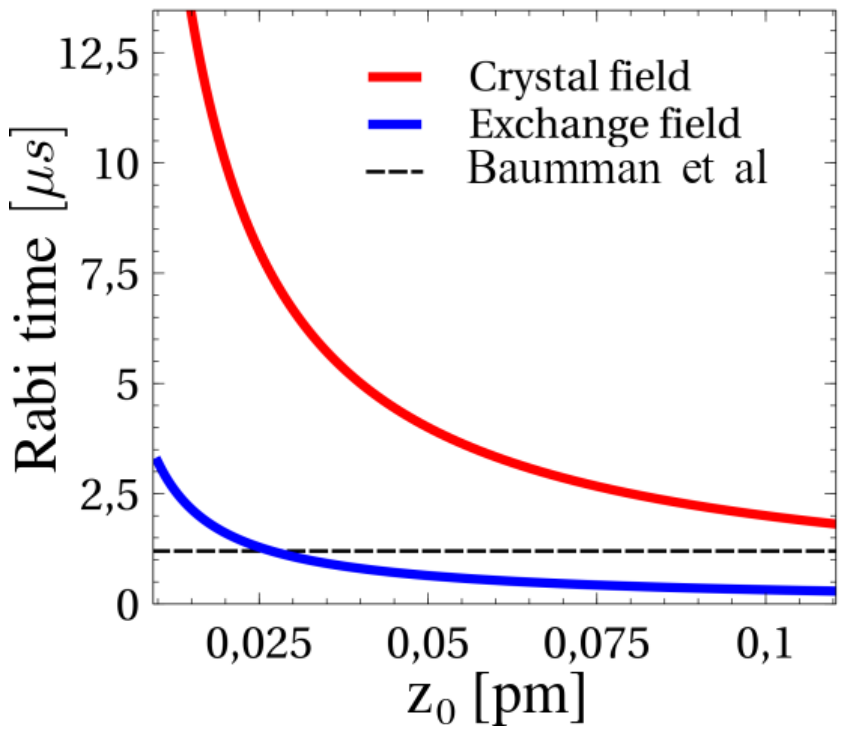
$$z(t) = \frac{q_{\mathrm{atom}}}{k}\frac{V_{\mathrm{RF}}(t)}{d}$$

DFT elastic constant

$$k \approx 600\ \mathrm{eV/nm^2}$$

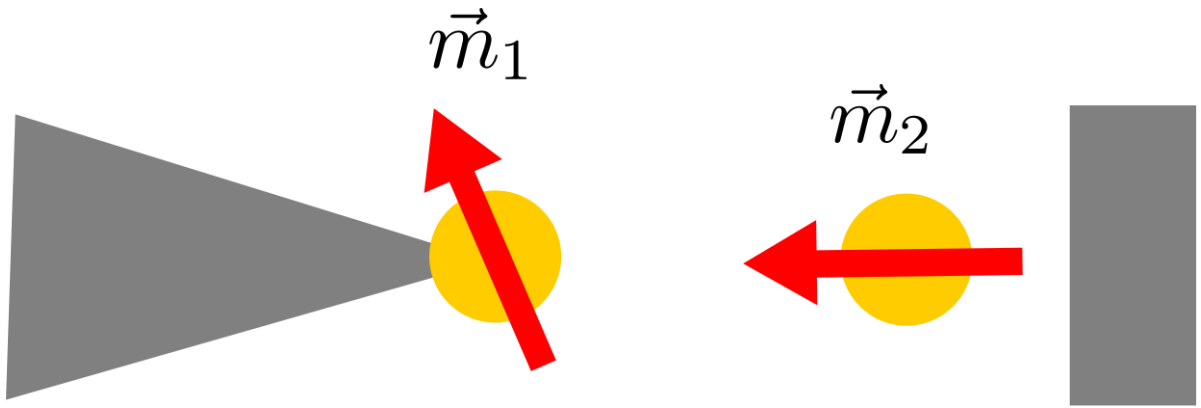
$$z = 0.044\ \mathrm{pm}$$

Fitting to experiment



Detection of the signal

Conventional magneto-resistive effect



$$\delta \frac{dI}{dV} \propto \langle \vec{m}_1 \rangle \cdot \delta \langle \vec{m}_2 \rangle$$

$$P_0 - P_1 = \tanh\left(\frac{\hbar\omega_0}{2k_BT}\right)\left(1 - \frac{\Omega_0^2 T_1 T_2}{1 + (\omega - \omega_0)^2 T_2^2 + \Omega_0^2 T_1 T_2}\right)$$