Many-Body Effects in the Magnetization Tunneling of an Ensemble of Mesoscopic Spins

highly diluted Ho^{3+} ions in $LiYF_4$

Romain GIRAUD*,

Bernard BARBARA, Wolfgang WERNSDORFER
Laboratoire de Magnétisme Louis Néel, Grenoble, France

Alexandra TKACHUK
Vavilov State Optical Institute, 199034 St. Petersburg, Russia

Dominique MAILLY
Laboratoire de Photonique et de Nanostructures, CNRS, Marcoussis, France

* ***Now @ Laboratoire de Spectrométrie Physique,***
Equipe mixte CEA-CNRS 'Nanophysique et Semiconducteurs', Grenoble , France

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Outline

- ***Introduction***

- ***The quantum dynamics of a single Ho^{3+} ion***
  - ✓ **Origin of the mesoscopic behavior**
  - ✓ **Strong hyperfine coupling limit**
  - ✓ **Hysteresis loops @ low field sweep rates**

- ***The quantum dynamics of an ensemble of Ho^{3+} ions***
  - ✓ **Cross-spin relaxations (co-tunneling & biased tunneling)**
  - ✓ **Anisotropic interactions**
  - ✓ **Hysteresis loops @ fast field sweep rates; Ac-susceptibility**

- ***Conclusion***

About the large scale …

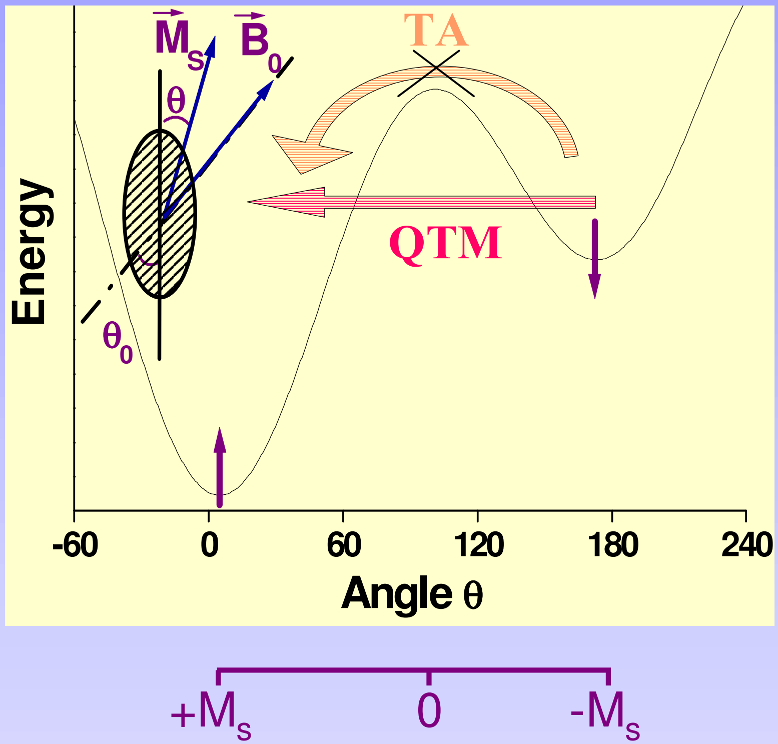
- **Mesoscopic spins (S~10)** **periodic 3D-array** ***or* diluted**

- **Inhomogeneous magnetic behavior**
  - *Pining of a narrow domain wall*
  - *Spin glass*

  ?

Problematic

$E(\theta) = K\sin^2\theta - M_S B_0 \cos(\theta_0 - \theta)$



- ***Thermal activation***

  ↳ below T_B , $\tau_{TA} \geq \tau_{exp}$

- ***Slow quantum fluctuations***

  ↳ Mesoscopic spin (**S~10**)

$$H = H_1 + H_2$$

$[H_1, S_Z] = 0$

level separations

axial anisotropy

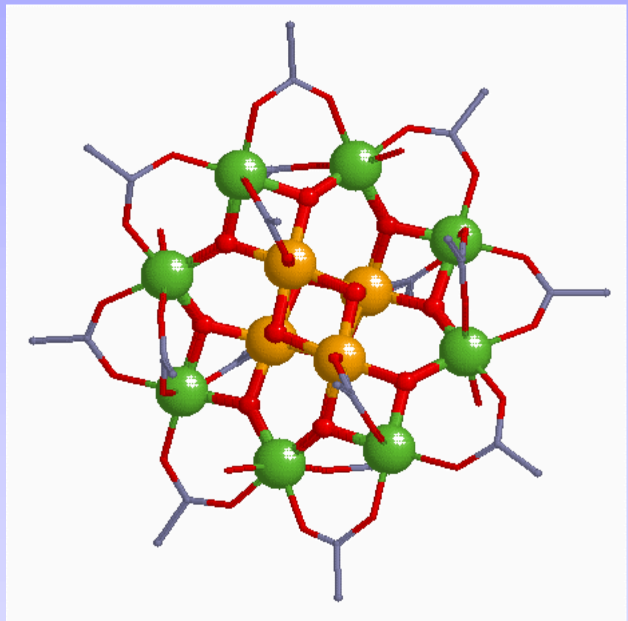
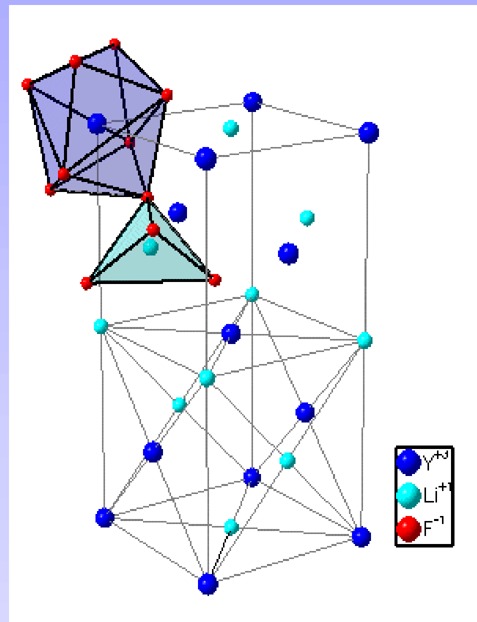
$[H_2, S_Z] \neq 0 \rightarrow i\hbar\partial\psi/\partial t \neq 0$

level repulsions

time evolution (QTM)

axial symmetry breaking

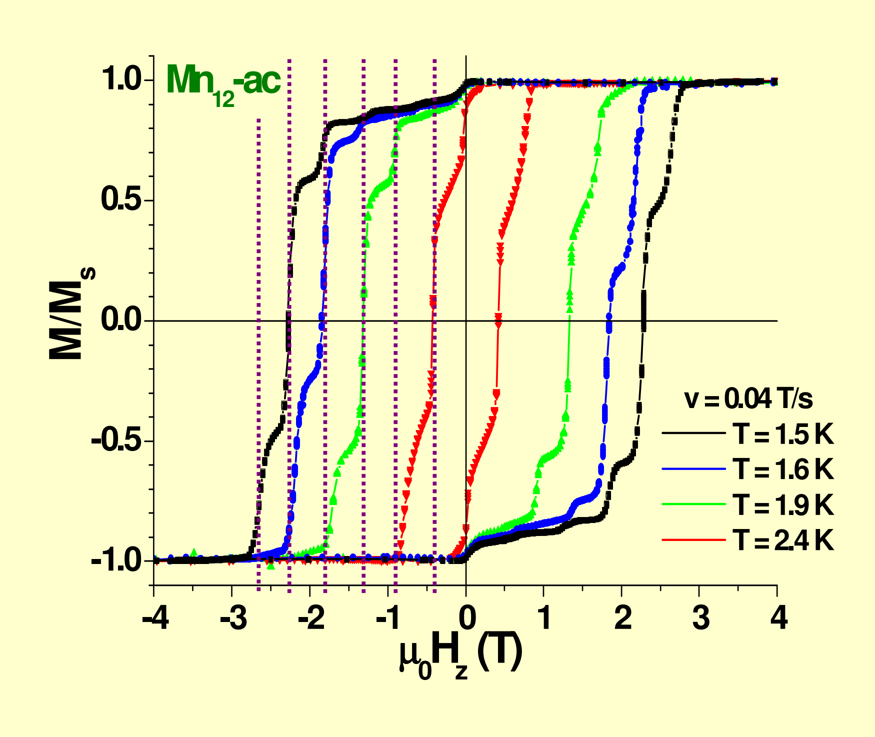
Mesoscopic spins

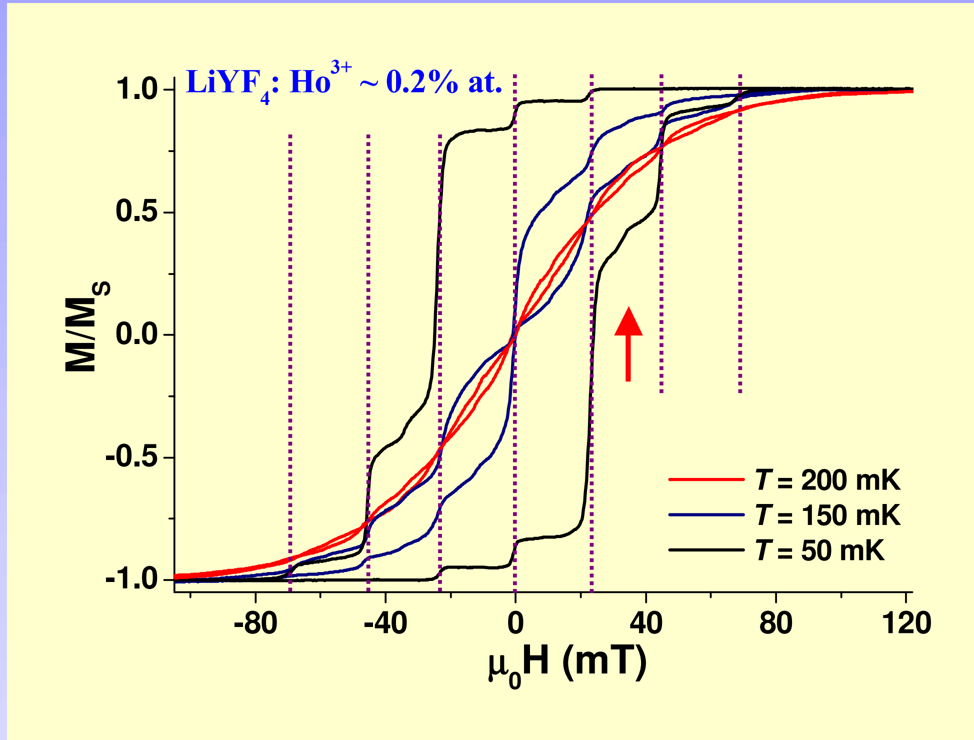
Single molecule magnets	***Diluted rare-earth ions***
Mn_{12}-ac	**$LiY_{1-x}Ho_xF_4$**
	
S = 10	**J = 8**

Mesoscopic spins

Single molecule magnets

Diluted rare-earth ions





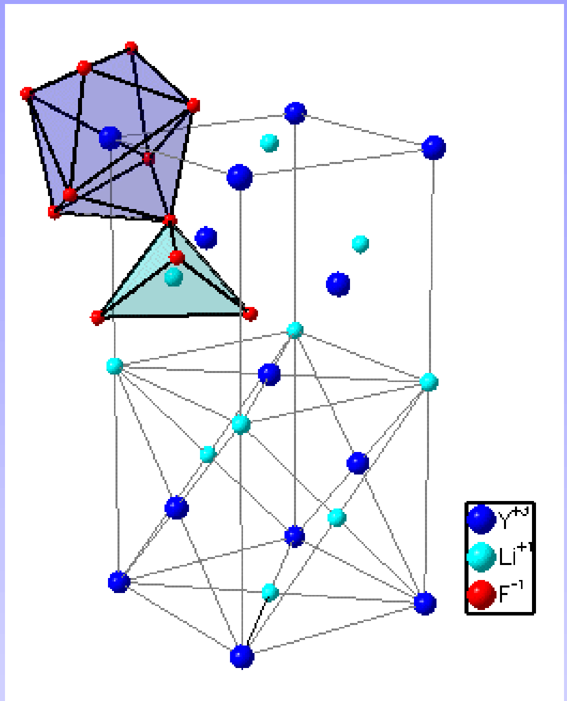
$\mu_0 H_n \approx n \times 0.4$ T (*n integer*)

L. Thomas et al., Nature 383, 145 (1996)

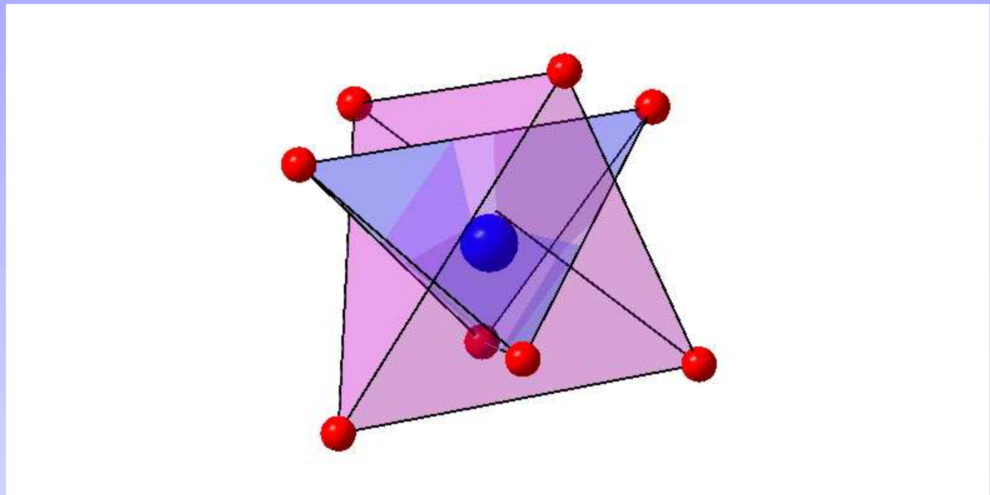
$\mu_0 H_n \approx n \times 23$ mT (*n integer*)

R. Giraud et al., PRL 87, 057203 (2001)

$LiY_{1-x}Ho_xF_4$ *a model system*



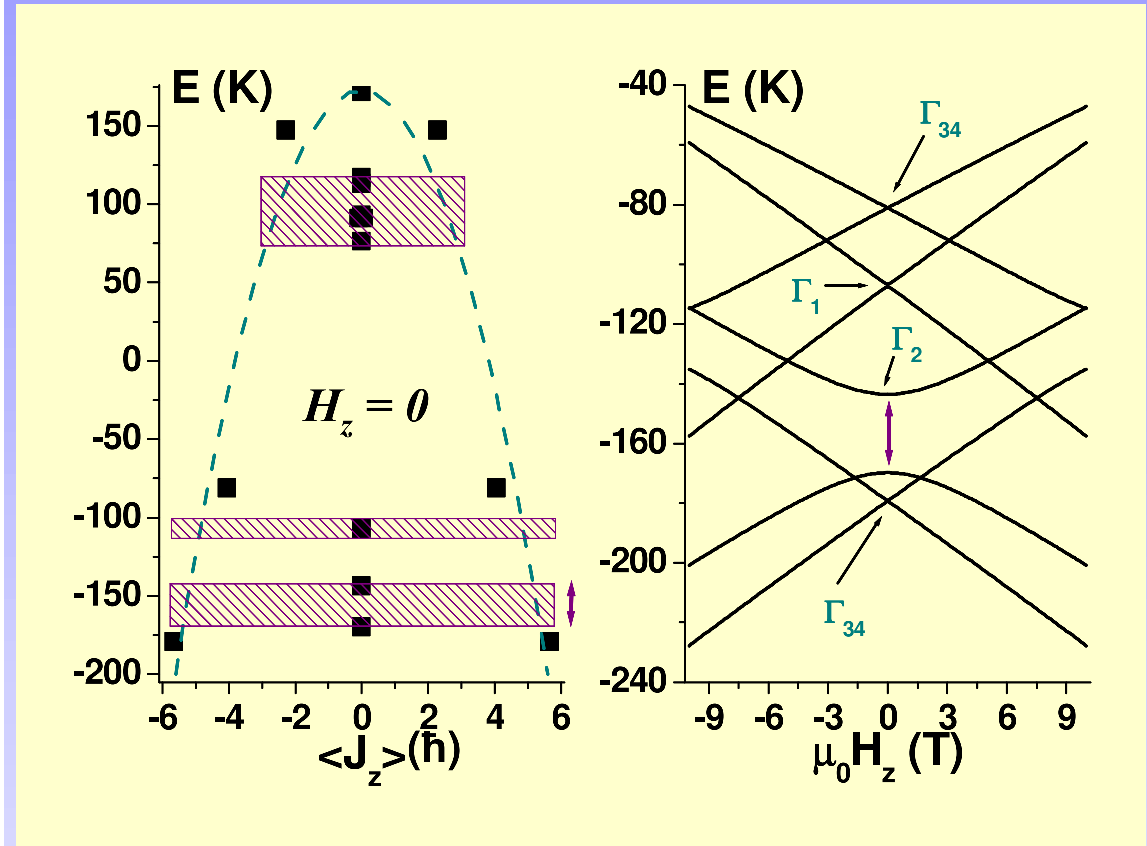
- Free ion **J=8** ⇨ **2J+1 = 17** **|J,M>**
- Crystal field point symmetry **S_4** **IR: $\Gamma_1, \Gamma_2, \Gamma_3, \Gamma_4$**



$$H_{CF} = B_{20}O_{20} + B_{40}O_{40} + B_{60}O_{60} + B_{44}O_{44} + B_{64}O_{64}$$

level separations **level mixings**

Electronic Zeeman diagram



- ***Thermal activation***

  $T_1(T) \Rightarrow T_B$

  Orbach $T_1 \propto e^{\delta E/kT}$

  $\delta E \sim 10K$

- ***Slow quantum fluctuations***

  Ising-like ground doublet

  ↓

  $S=½$ with **$g_{eff} \sim 13$**

  Stability of this doublet…?

  ↳ **Holmium *nuclear spin***

Strong hyperfine coupling limit

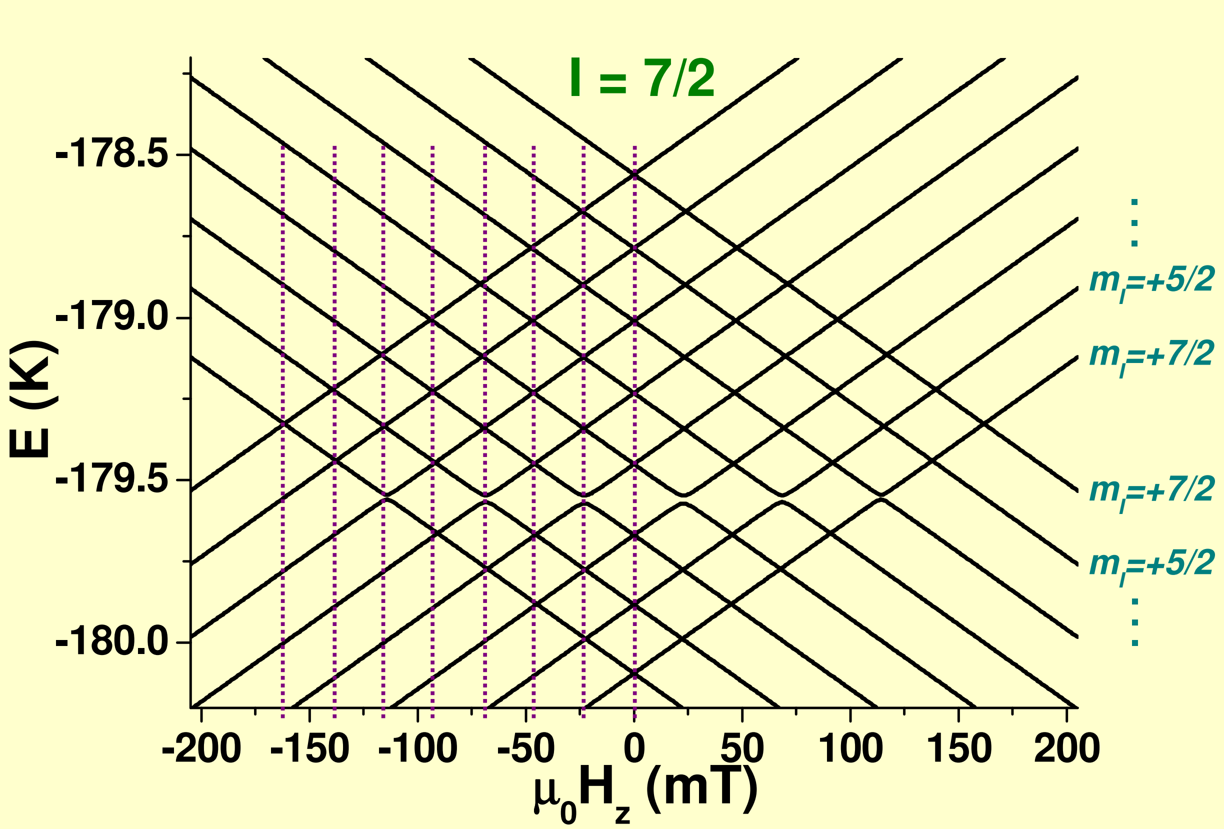
$H = H_{CC} + A_J \vec{J} \cdot \vec{I}$

$A_J [J_z I_z + ½(J_+ I_- + J_- I_+)]$

17 * 8 = 136

Hyperfine structure of the doublet (16) →

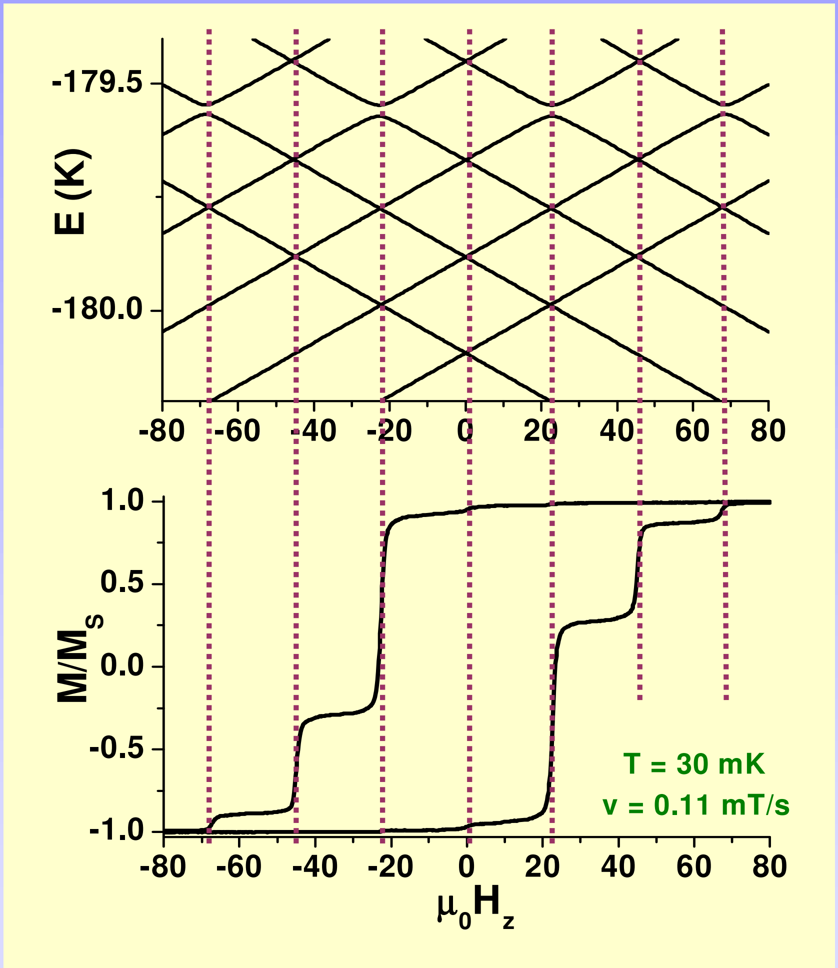
δE~205 mK



<u>Resonance fields</u> : $\mu_0 H_n \approx n \times 23$ mT (-7≤n≤+7)

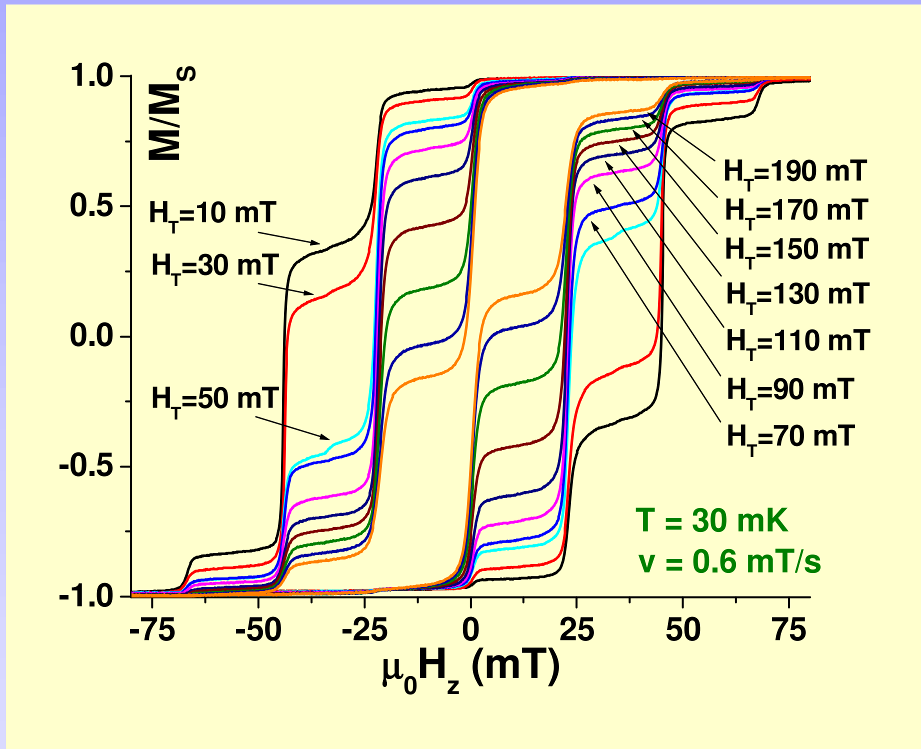
The quantum dynamics of a single Ho^{3+} ion

@ slow field sweep rates (v~1mT/s)

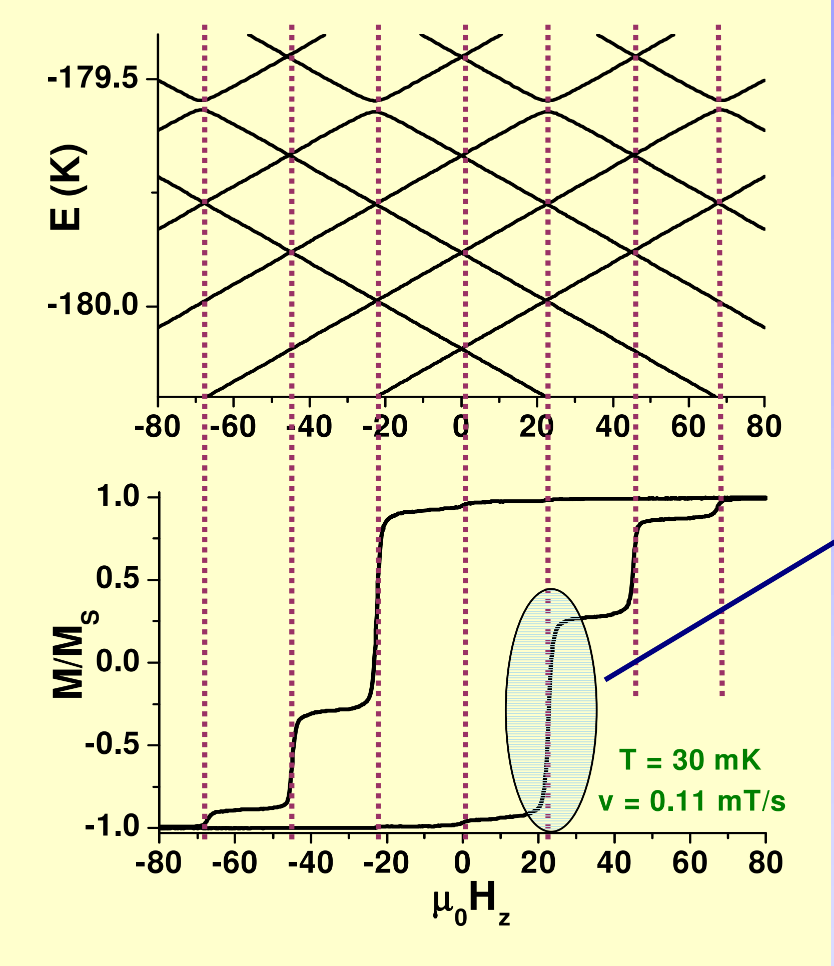


Additional symmetry breaking

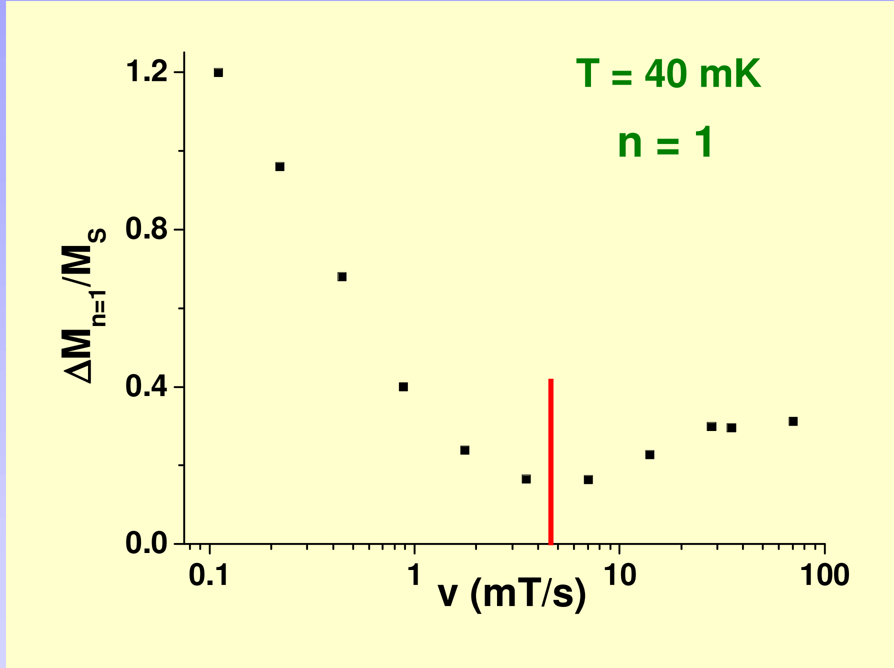
due to a **transverse field H_T**



From the quantum dynamics of a single Ho^{3+} ion…
…to many-body effects



Field sweep rate dependence…



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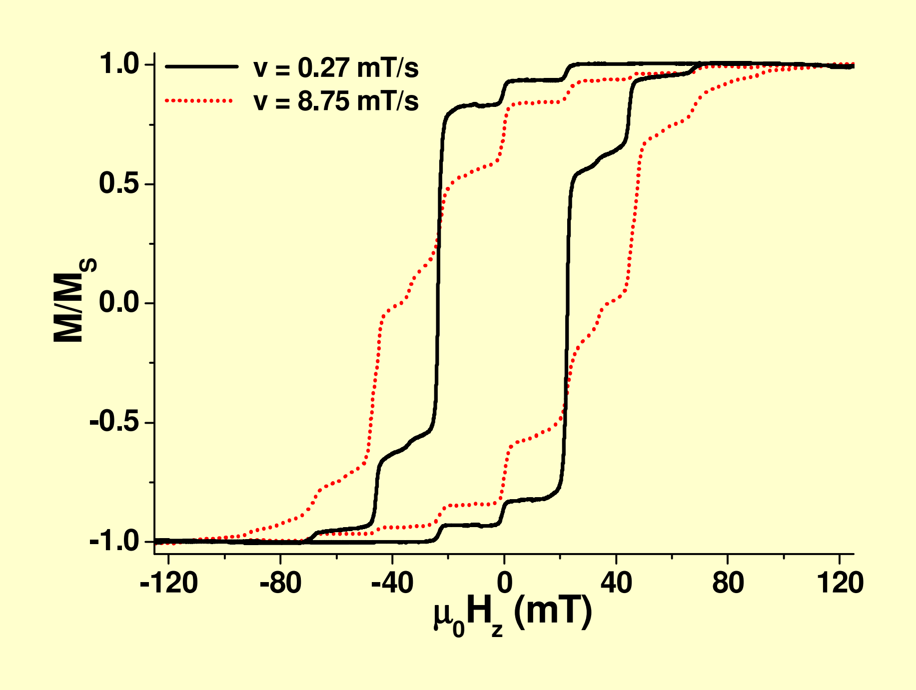
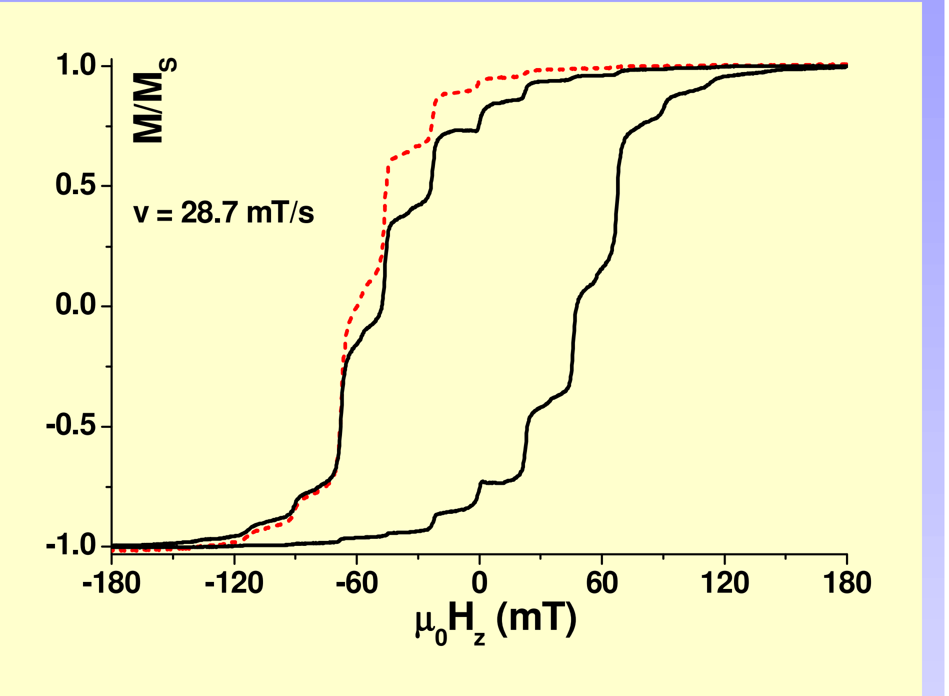
Crossover to a strongly out-of-equilibrium spin system

Phonon bottleneck

Raise of the internal temperature *when increasing the sweep rate*



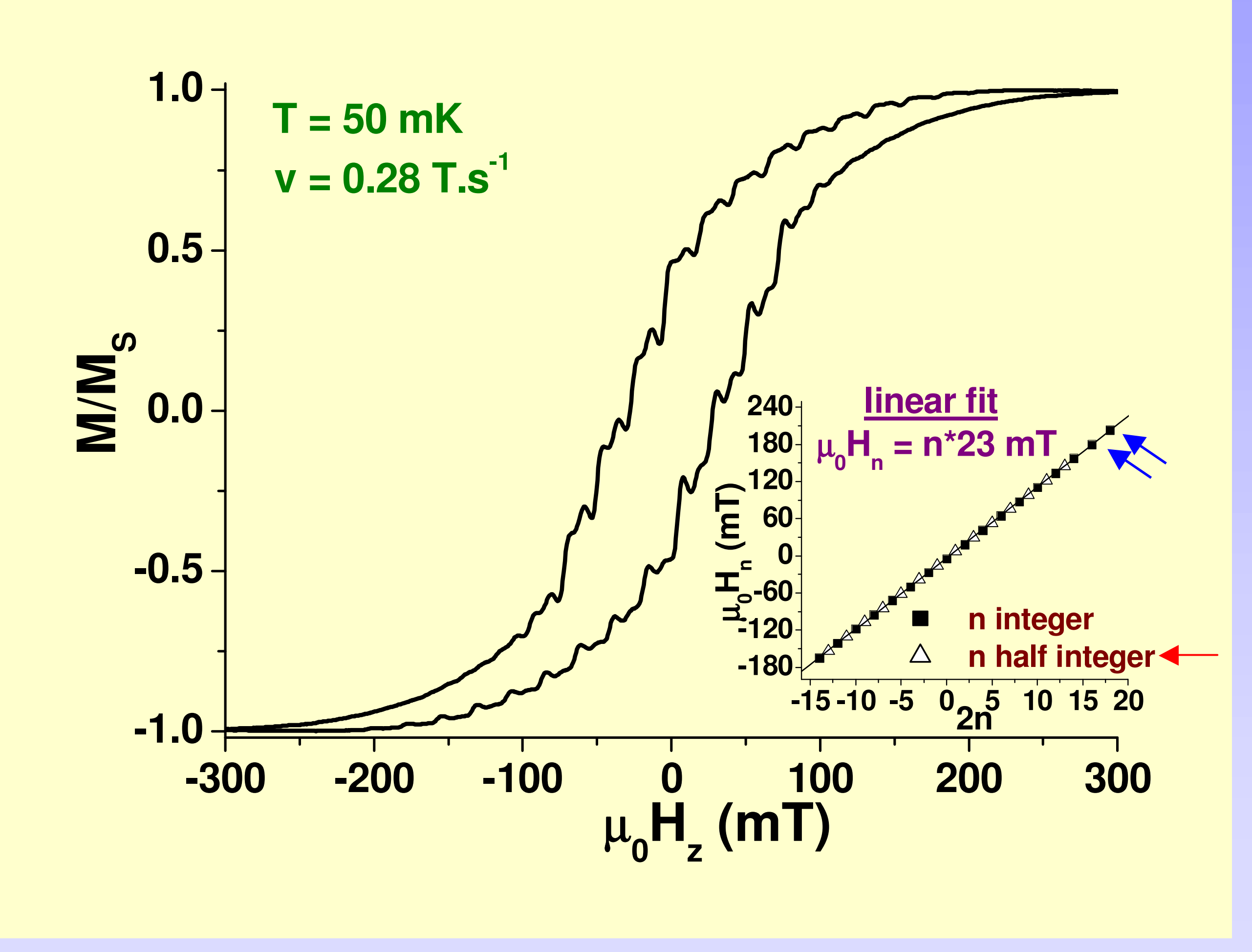


The field sweep drives the spin system

far from its internal equilibrium

The quantum dynamics of an ensemble of Ho^{3+} ions

@ fast field sweep rates (v~1T/s)



Additional magnetization steps

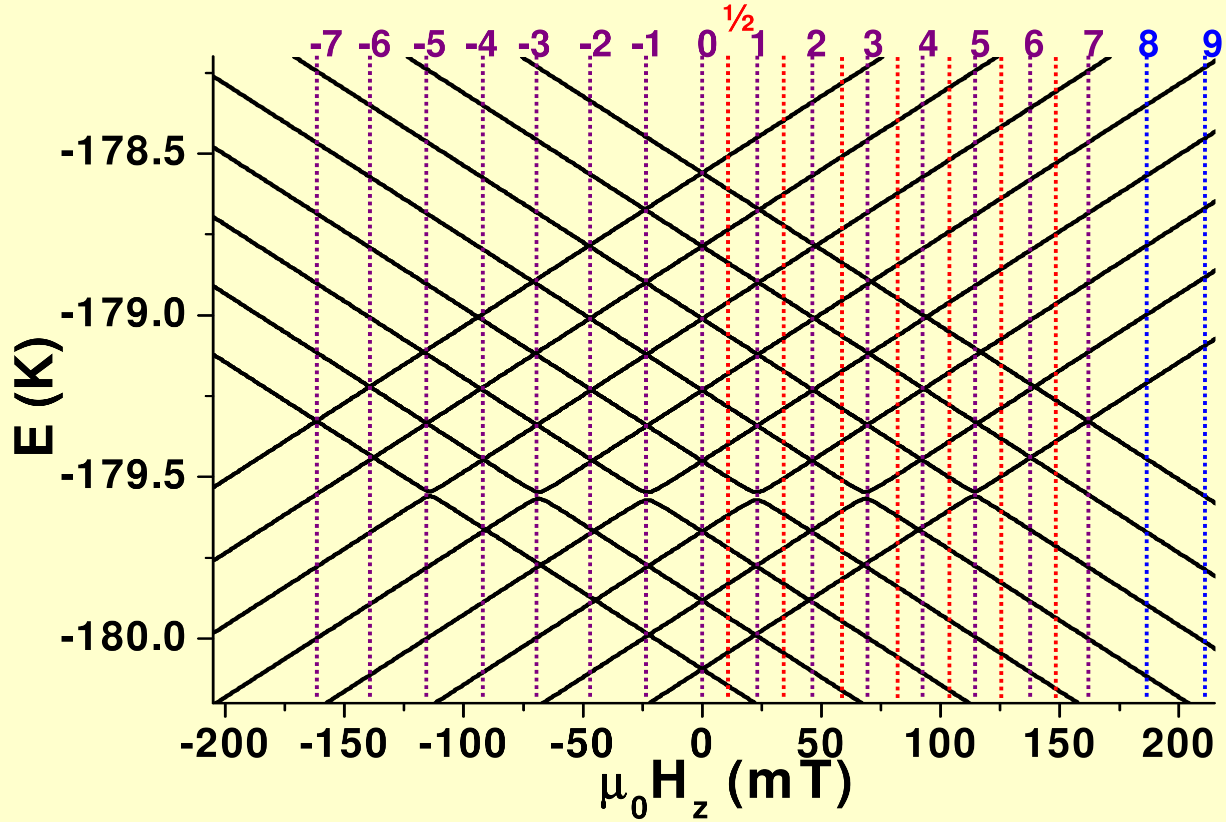
No explanation in terms of level crossings within the *single-ion representation*

n = 8, 9

-13 ≤ 2n ≤ +13

(n half integer)

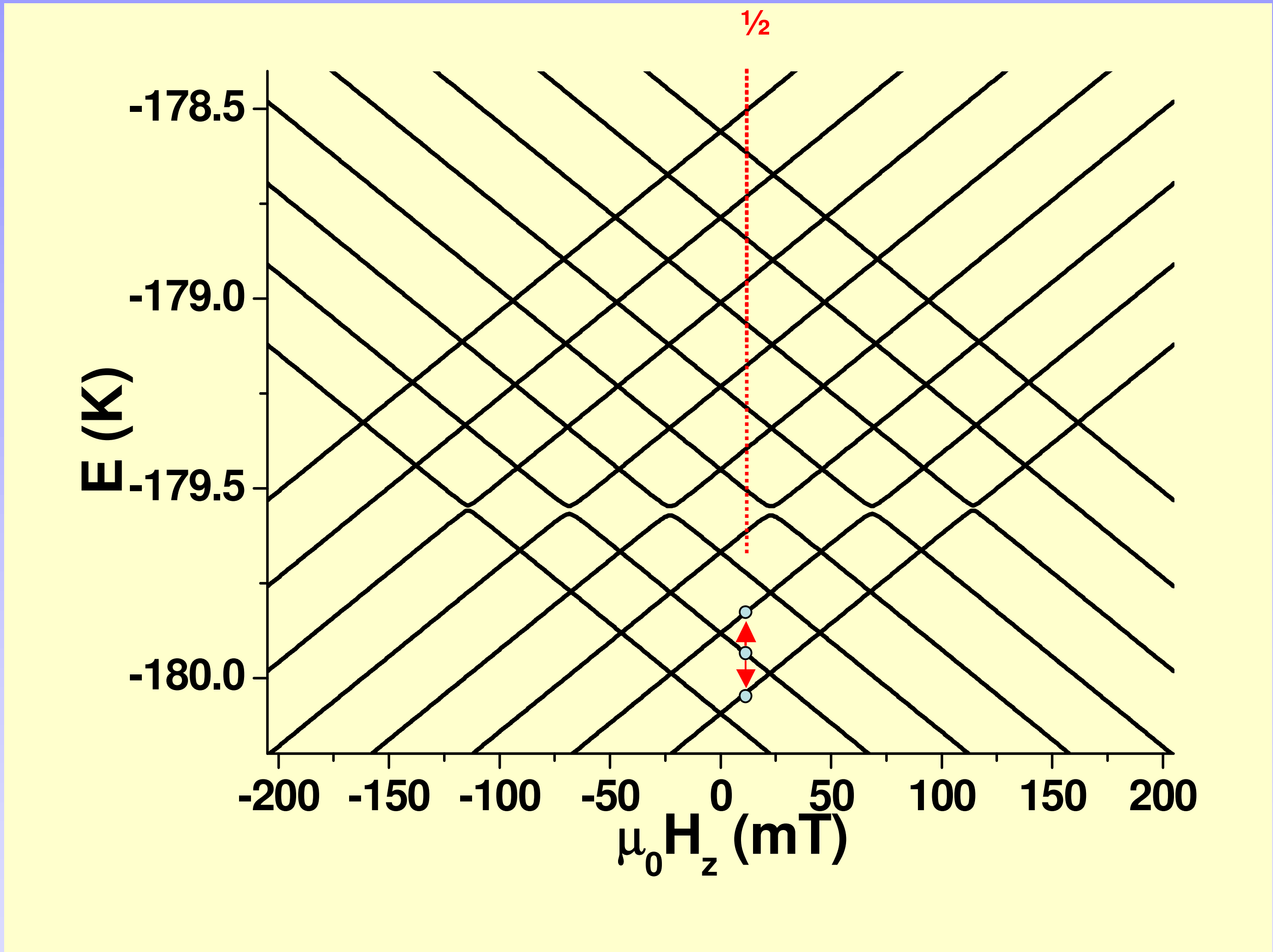
Spin-spin cross relaxations

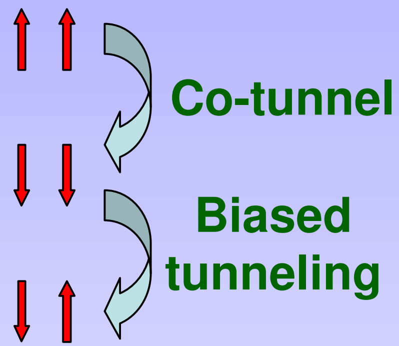


No level crossings ***but***… **equally-spaced levels**

Some multi-spins transitions allow for energy conservation @ resonant fields

Spin-spin cross relaxations : co-tunneling & biased tunneling





The two-ions representation

$$H = H^{(1)} + H^{(2)} + H^{12}$$

136 * 136 = <u>18496</u>

Doublet Γ_{34}

J = 8 ⇒ S = ½

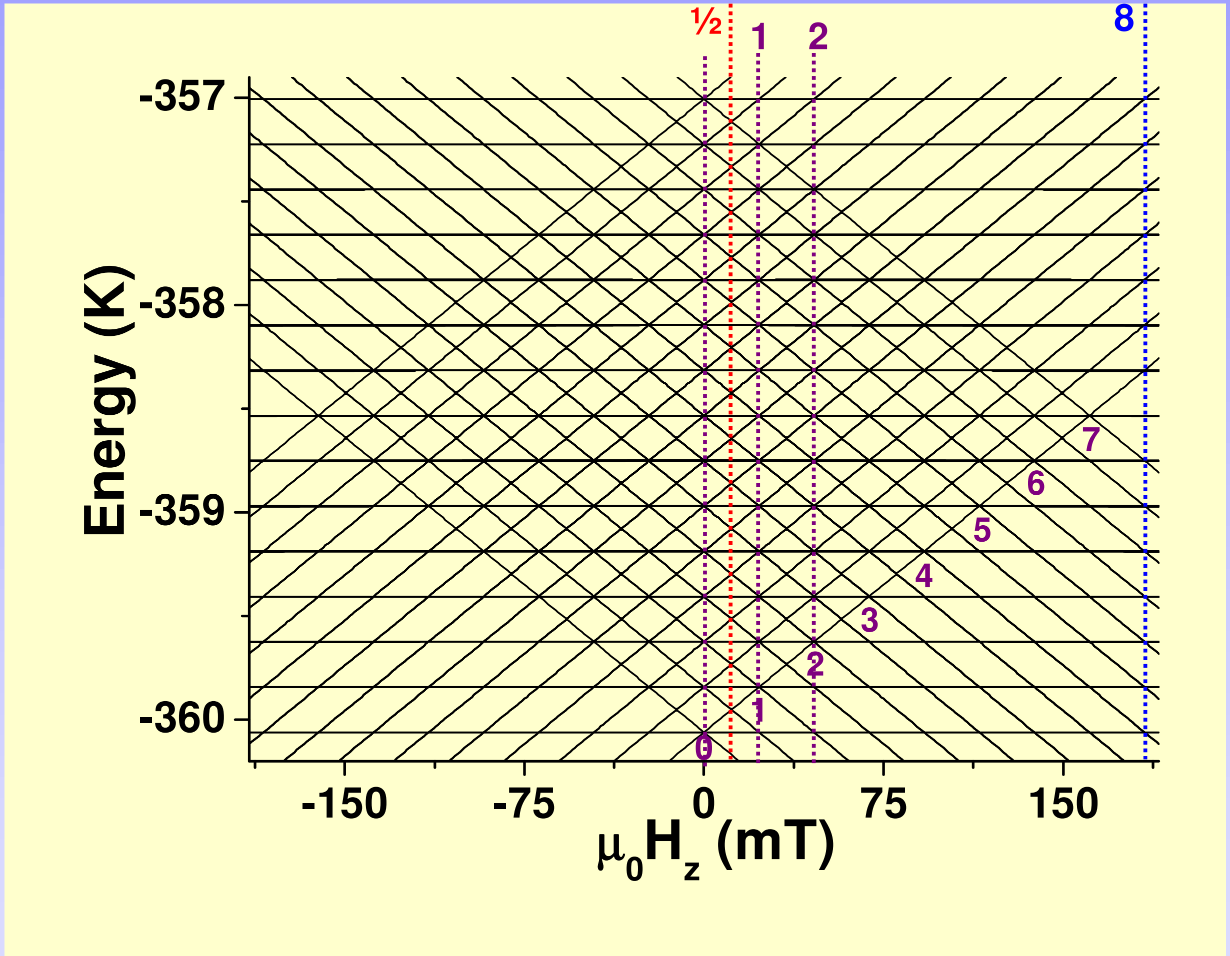
16 * 16 = <u>256</u>

$H^{12} = 0$

Doublet Γ_{34}

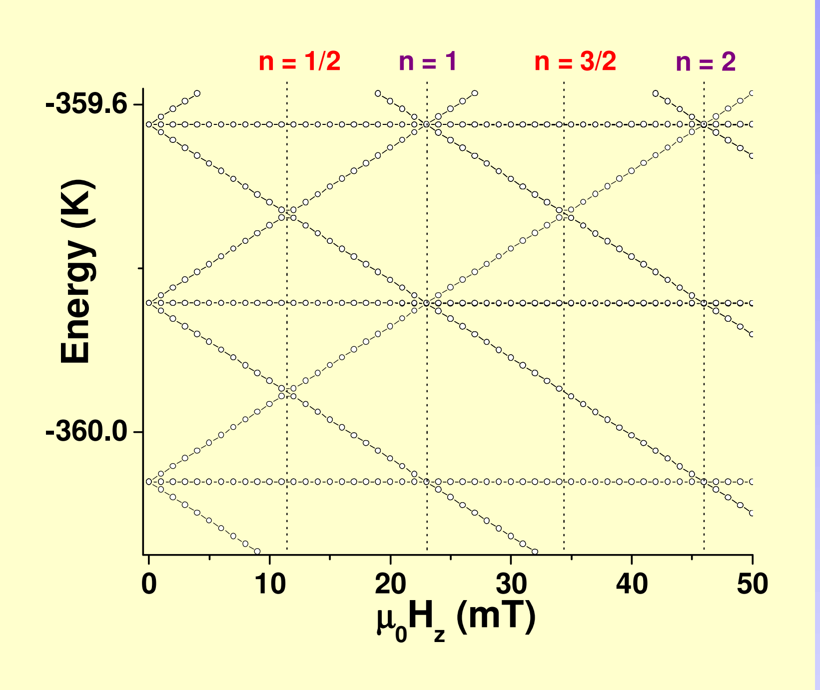
Singlet Γ_{2}

24 * 24 = <u>576</u>

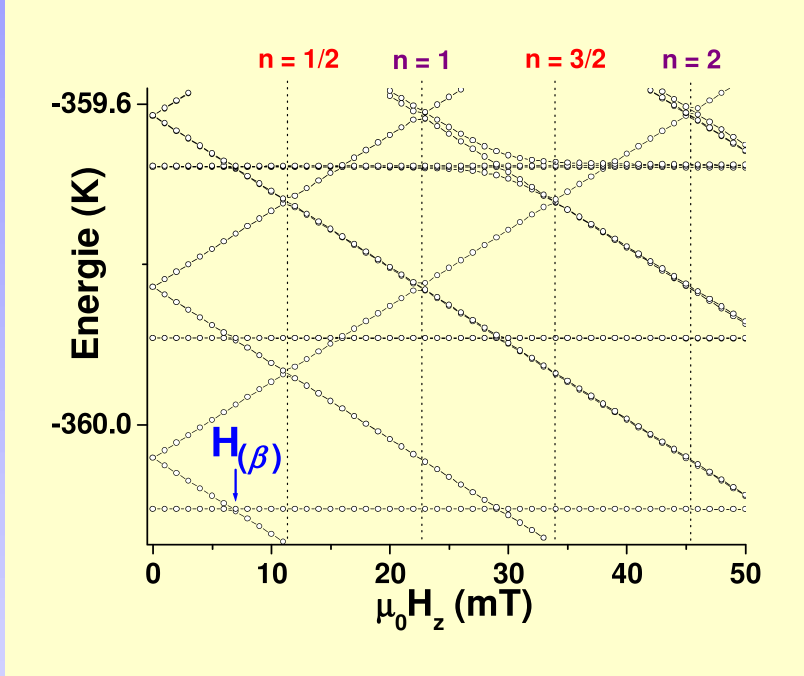


The two-ions representation

Without any coupling



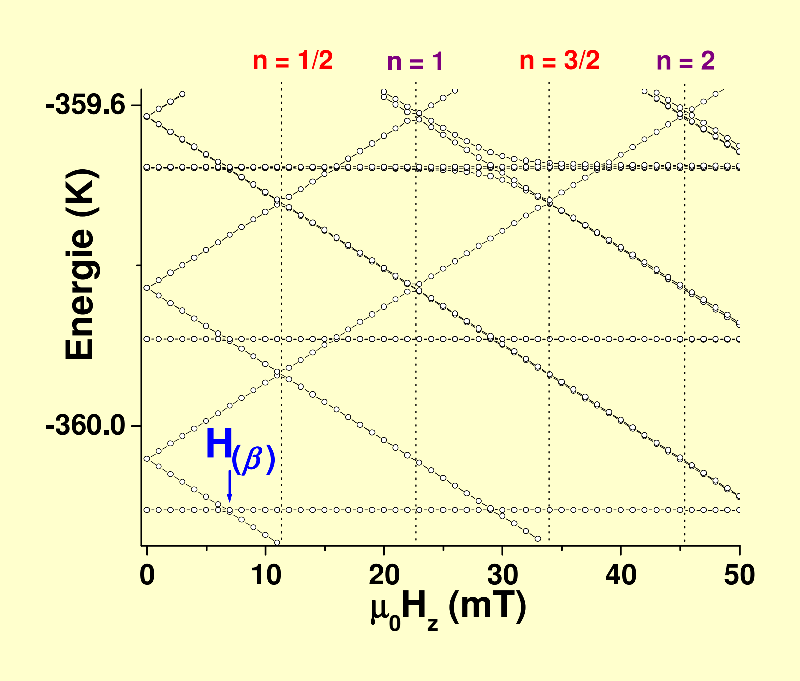
With an *isotropic* interaction



- **Co-tunneling crossing fields are independent of the coupling strength *β***
  (transitions are allowed by dipolar/anisotropic exchange interactions)
- **Biased tunneling crossing fields (by a '<u>molecular field</u>')**
  (fine structure *if* exchange couplings > dipolar interactions)

The two-ions representation

With an *isotropic* interaction



Co-tunneling transitions are allowed by anisotropic interactions

Dipole-dipole interactions

$$H_{dip\text{-}dip} = \tfrac{1}{2}\Sigma\, [g_k g_l \mu_B^2 / r_{kl}^3$$

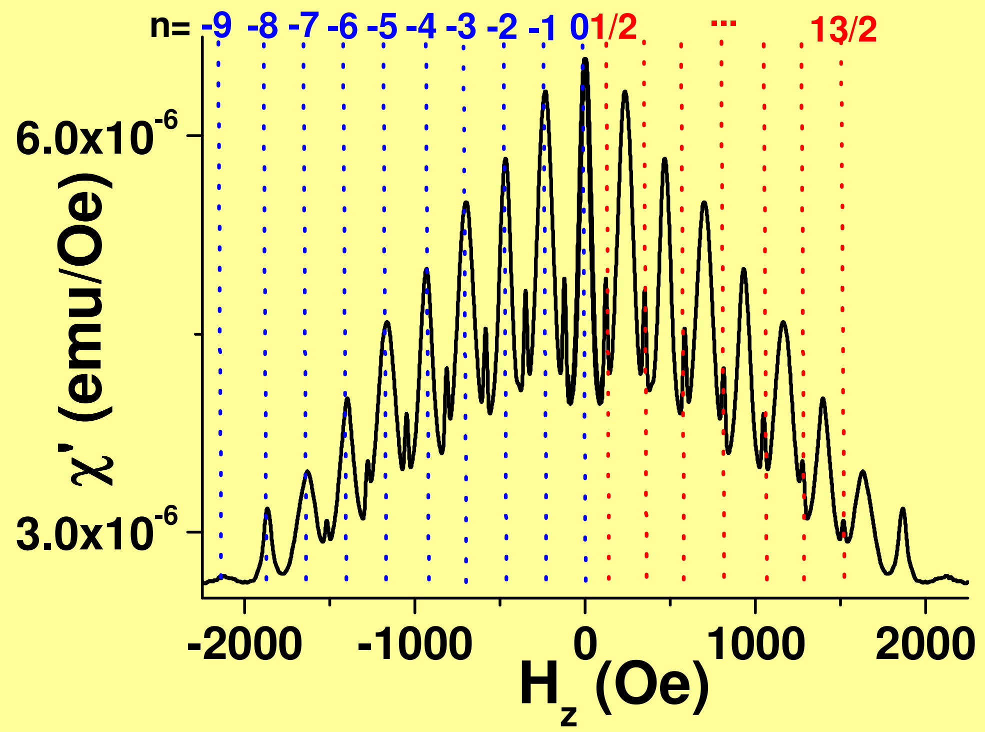
$$(1-3\cos^2\theta_{kl})[\mathbf{S_{zk}S_{zl}} - \tfrac{1}{4}(\mathbf{S_{+k}S_{-l}} + \mathbf{S_{-k}S_{+l}})]$$

$$-3/2\cos\theta_{kl}[e^{-i\varphi}\,(\mathbf{S_{+k}S_{zl}} + \mathbf{S_{zk}S_{+l}}) + e^{-i\varphi}\,(\mathbf{S_{-k}S_{zl}} + \mathbf{S_{zk}S_{-l}})]$$

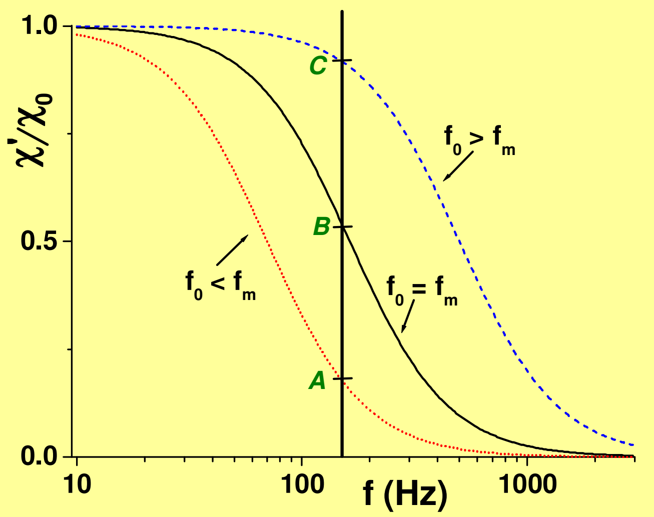
$$+\tfrac{3}{4}\sin^2\theta_{kl}(e^{-i2\varphi}\,\mathbf{S_{+k}S_{+l}} + e^{-i2\varphi}\,\mathbf{S_{-k}S_{-l}})]$$

Ac-susceptibility measurements

@ higher temperatures (T>1.8K)



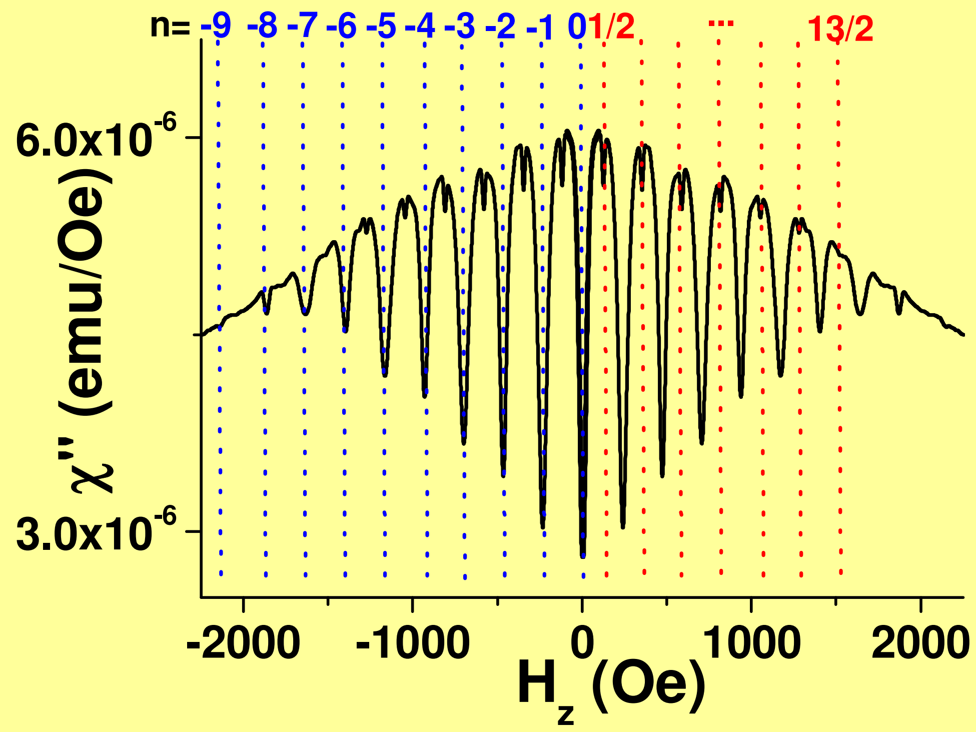
Debye model

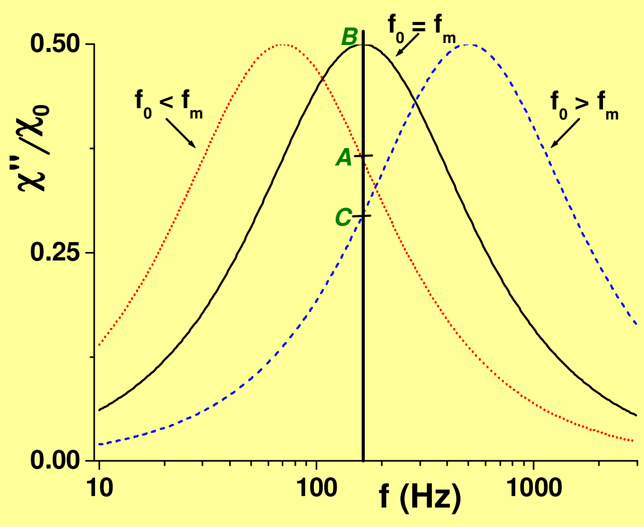


f_0 (H_z)

Ac-susceptibility measurements

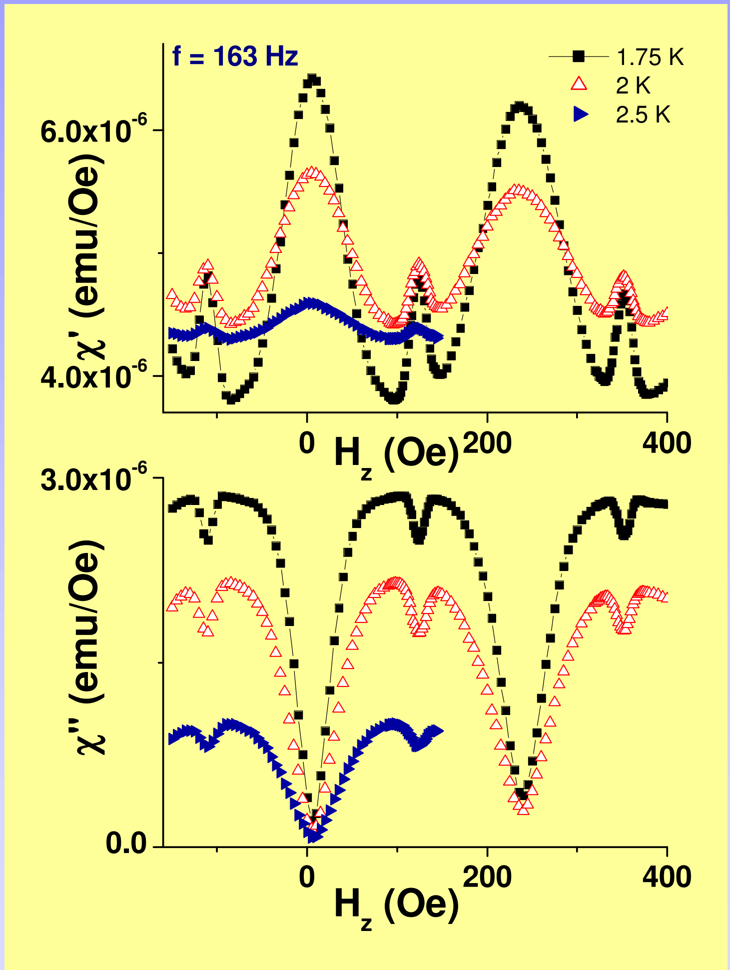
@ higher temperatures (T>1.8K)



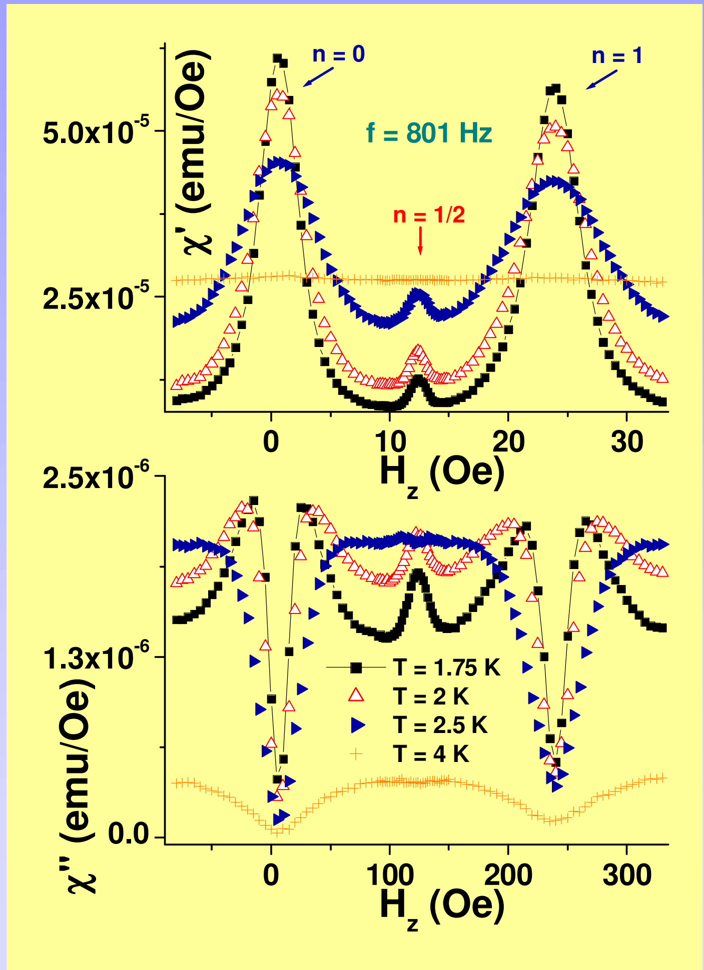


Ac-susceptibility measurements

@ f = 163 Hz



@ f = 801 Hz



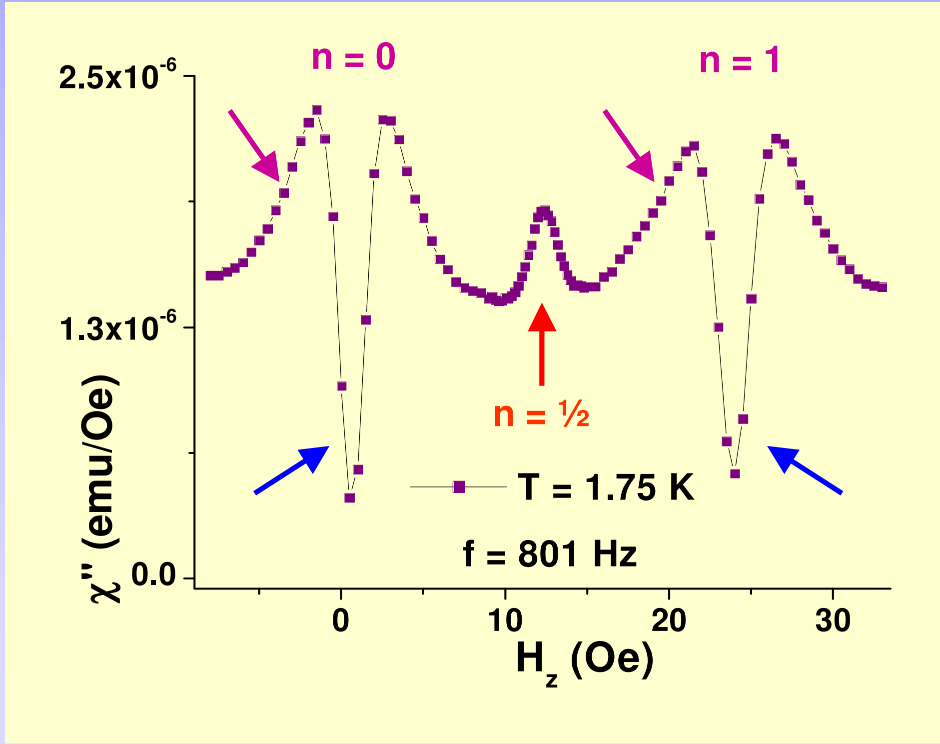
Ac-susceptibility measurements

1.75 K < T < 5 K

↳ Homogeneous phonon broadening > Inhomogeneous dipolar broadening

Allows to distinguish between the various tunneling mechanisms

- **Single-ion tunneling**
- **Two-ions co-tunneling**
- **Dipolar-biased tunneling**



Spin-spin cross relaxations : **a KEY ingredient …**

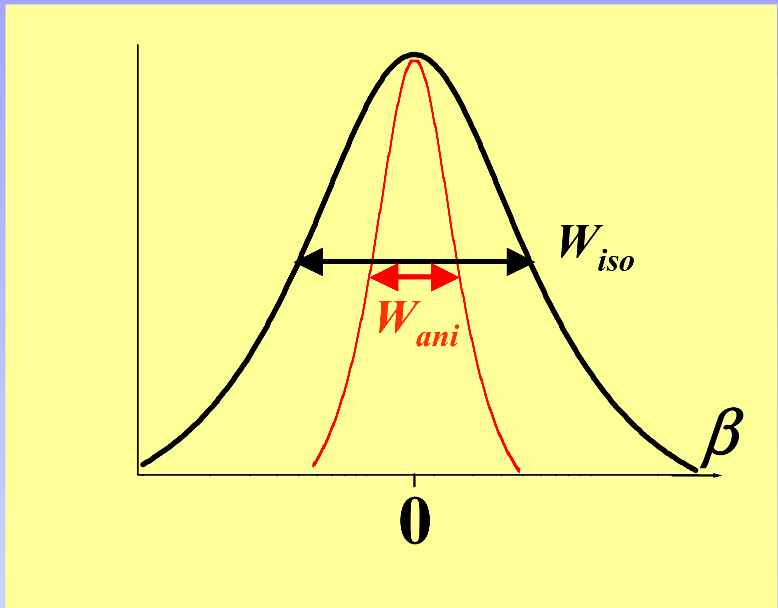
… to understand quantum fluctuations in an ensemble of interacting spins

- ***Weakly coupled mesoscopic spins :*** *Multi-spins dynamics*
  - ↳ *Spin bath theory* (***Prokof'ev & Stamp***)
  - ↳ *Co-tunneling dynamics* **@** ***fast field sweep rates***
- ***Spin glass***
  - ↳ *Role of weakly interacting spins…?*

… Strength & Nature of the interaction…

Spin-spin cross relaxations : a KEY parameter …

… to understand cross-spin relaxations…



$$R = \frac{W_{ani}}{W_{iso}}$$

Large R required to allow for quantum fluctuations at large scales…

Quantum spin glass

- *Dipolar spin glass*
- *Diluted magnetic semiconductors (hole-mediated exchange interactions)*

Summary

Highly diluted Ho^{3+} ions in $LiYF_4$ exhibit a mesoscopic behavior at low T

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The measure of an ensemble demonstrate *various* quantum relaxation mechanisms

- ***Single-ion behavior*** : ***strong hyperfine coupling***
- ***Many-body effects*** : ***co-tunneling*** **&** ***biased tunneling***

↳ ***Crucial role of anisotropic interactions for quantum fluctuations at a large scale***

Thank you!