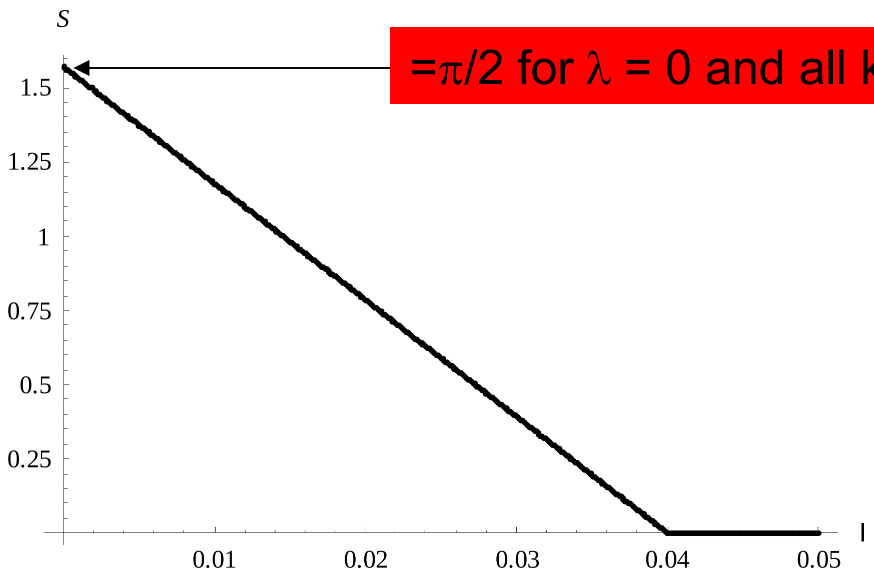
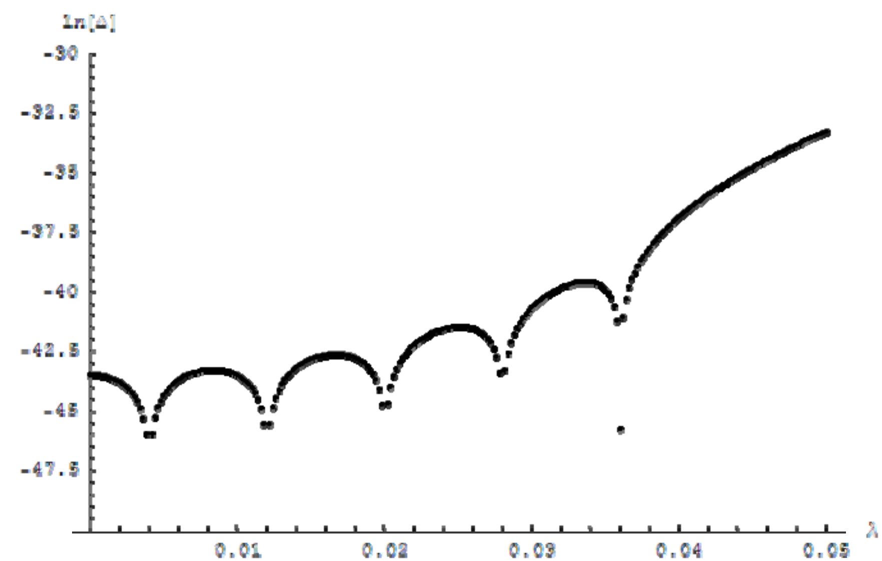
Oscillatory Tunnel Splitting In Mn12

This expression for Δ looks nice, but…

$$S_I = \text{[illegible]}$$





Numerical Integration reveals a linear dependence of S_I on λ, in agreement with the even quench spacing.

Phonon-Bottleneck-Driven Relaxation and Tunneling in Single-Molecule Magnets

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Thanks to: Myriam Sarachik, Eugene Chudnovsky, Dmitri Garanin

Funding: NSF, Alfred P. Sloan Foundation and Amherst College Dean of Faculty

Single-Molecule Magnets: An Overview

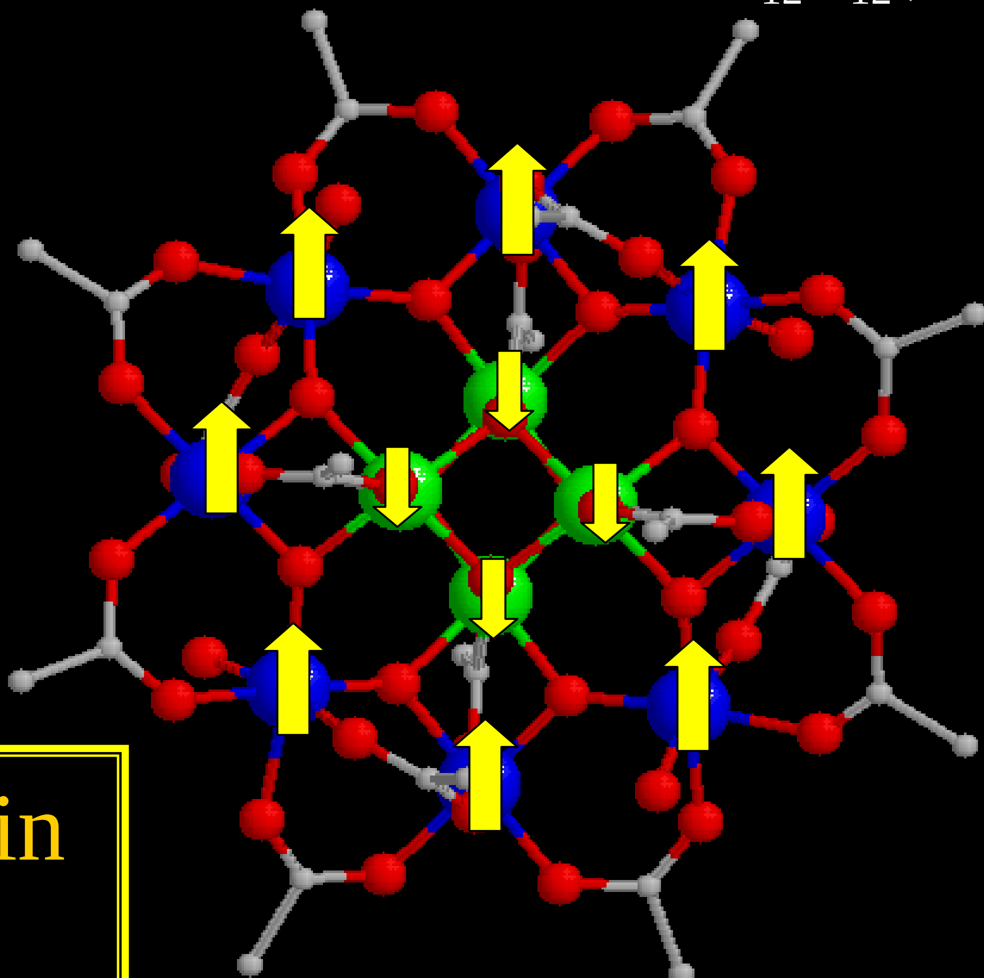
- Single-Molecule Magnets are Single-Molecule Magnets.
- Each individual molecule is bistable (has its own hysteresis) due to anisotropy effects.
- Exchange coupling within a molecule is strong, and therefore usually ignorable at low temperatures $\rightarrow$ a rigid high-spin object.
- The molecules grow into crystals, yet interact with each other weakly.

Mn_{12} Acetate

$Mn_{12}O_{12}(CH_3C0_2)_{16}(H_2O)_4$

Mn(III)
S=2

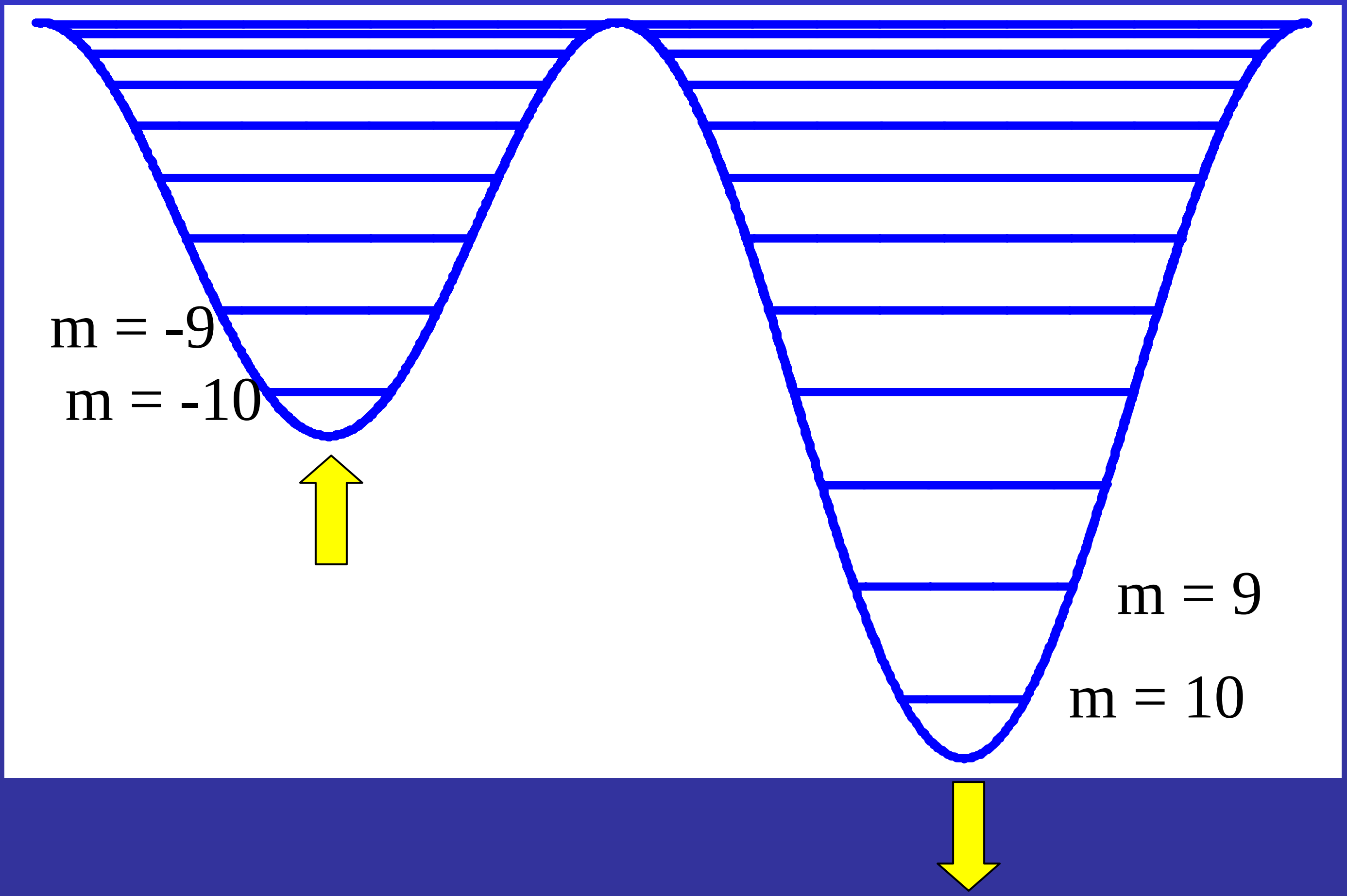
Mn(IV)
S=3/2



Total Spin
= 10

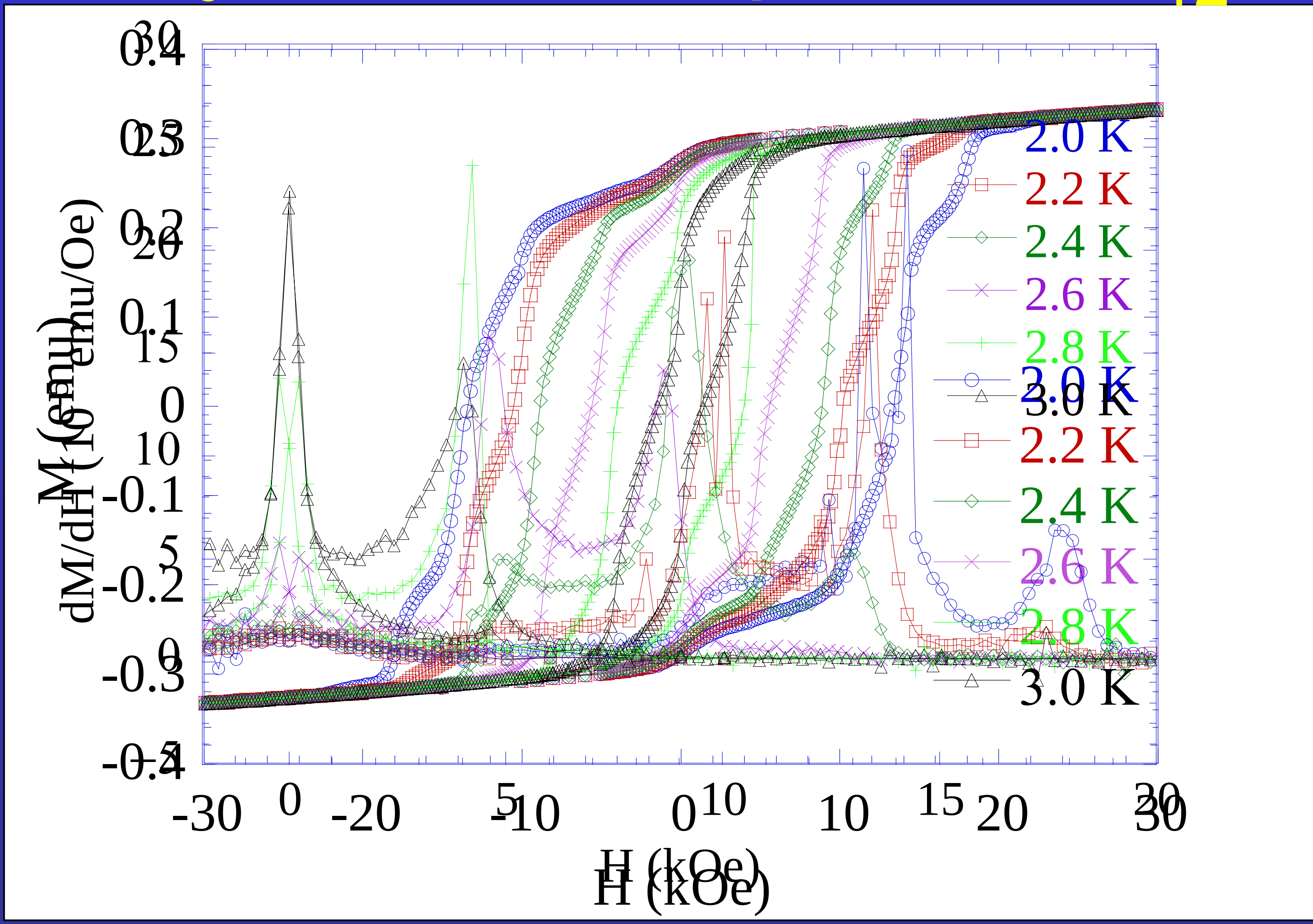
= oxygen
= carbon

Double-well Potential Model for Anisotropic Nanomagnet



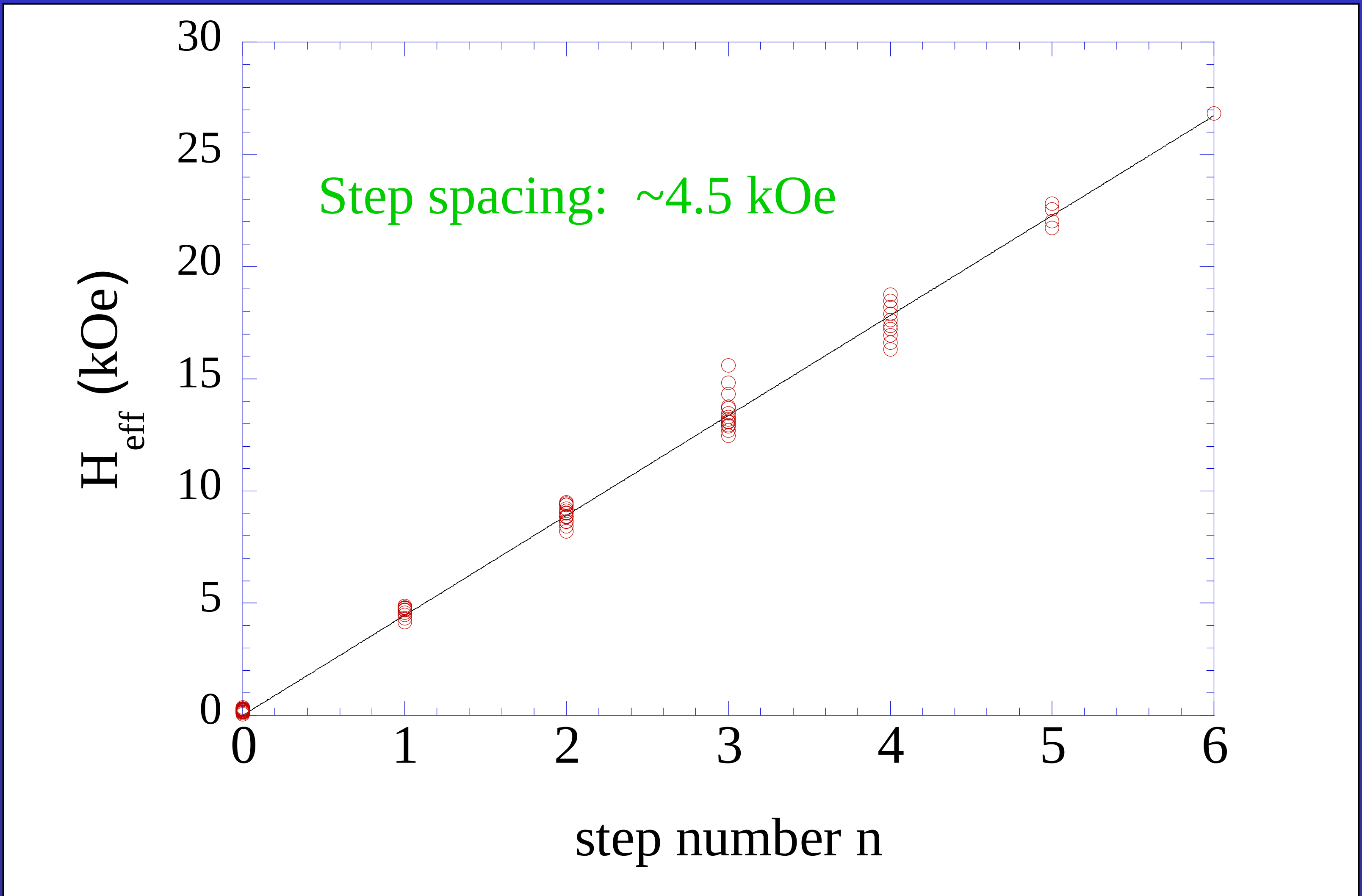
Magnetic field tilts potential. Energy levels correspond to different orientations of the magnet.

Hysteresis loops for Mn_{12}

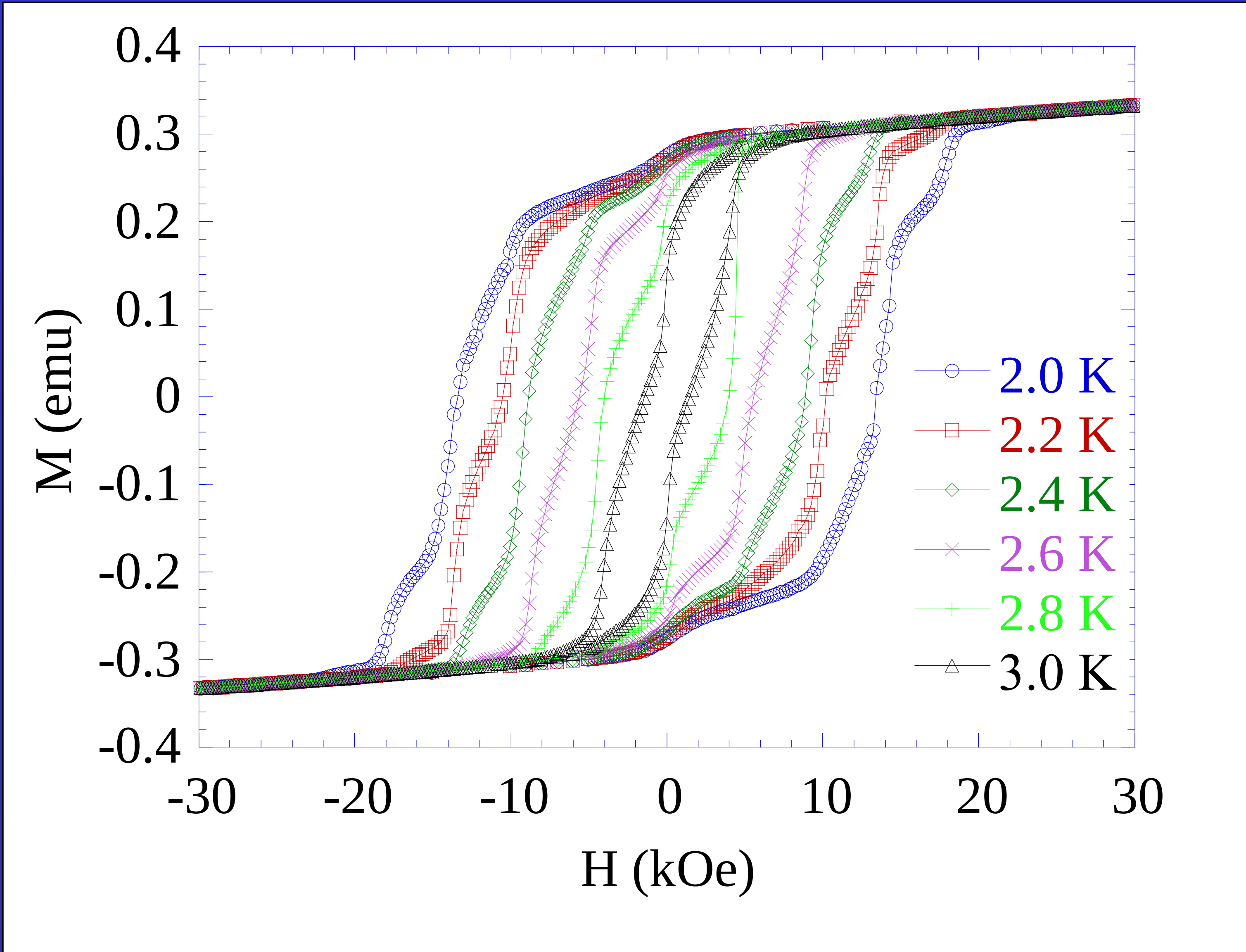


Friedman et al., PRL, 1996; Hernandez et al, EPL, 1996; Thomas et al., Nature, 1996; Hernandez et al., PRB, 1997.

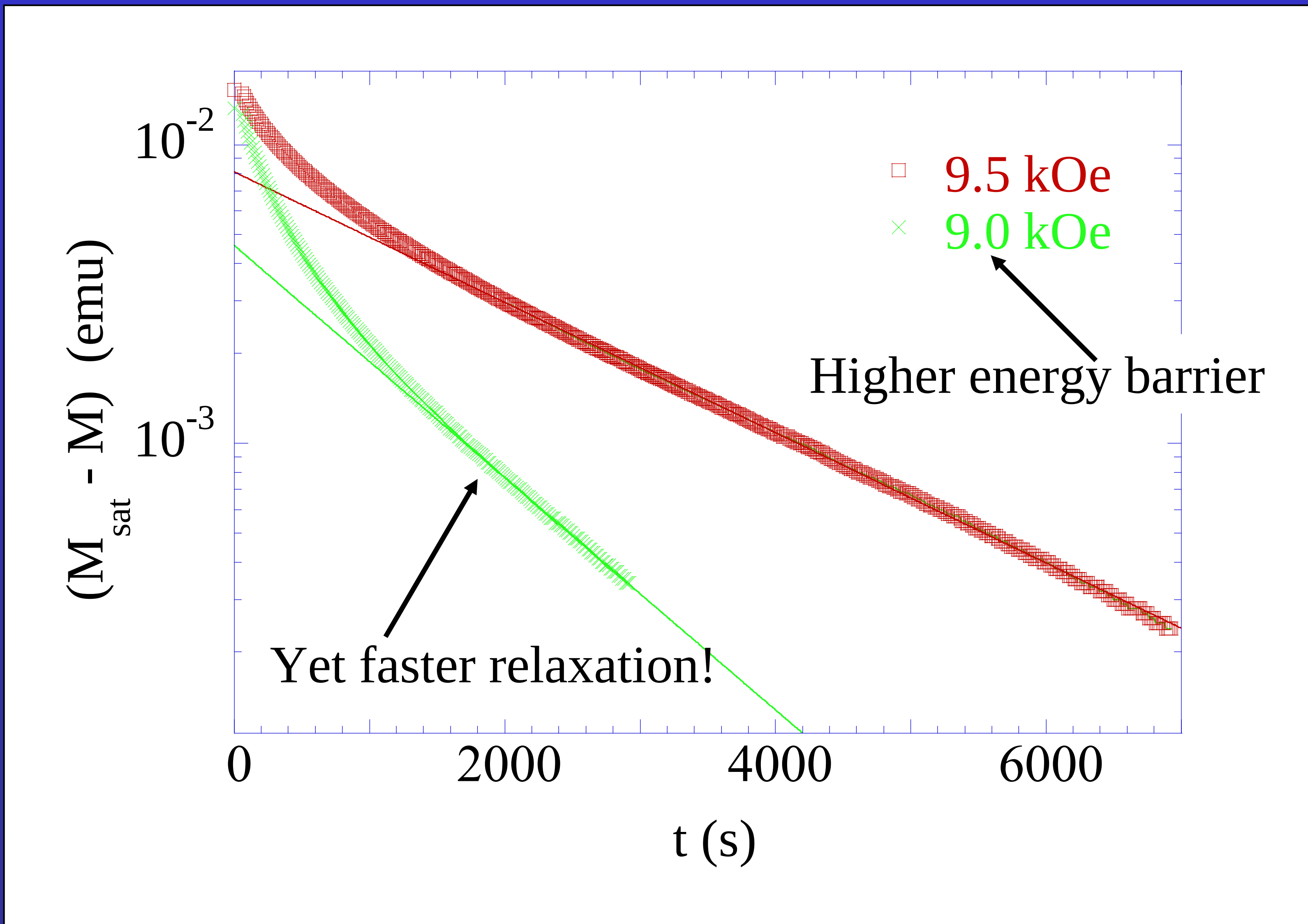
Uniform spacing between steps



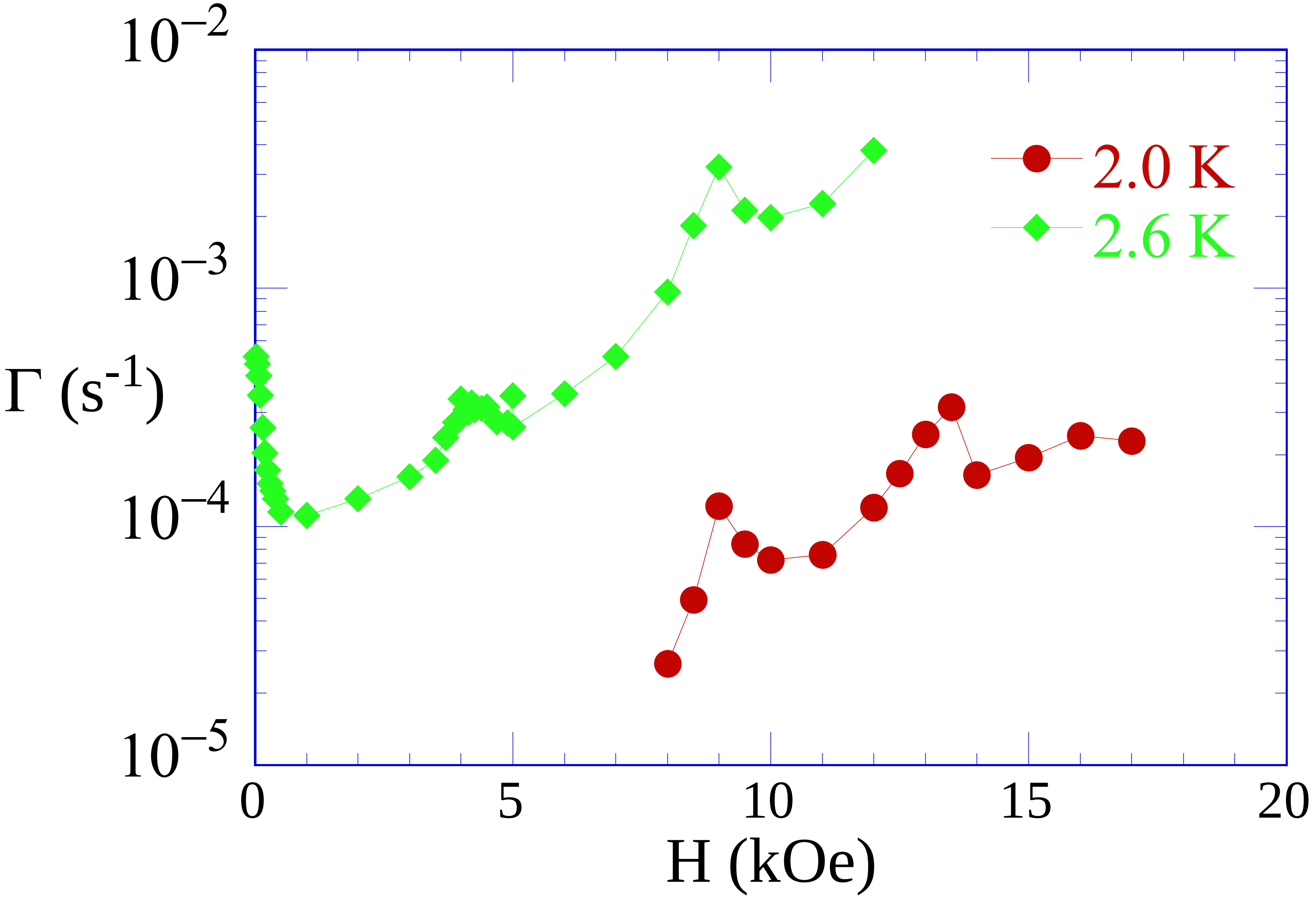
Hysteresis loops for Mn_{12}



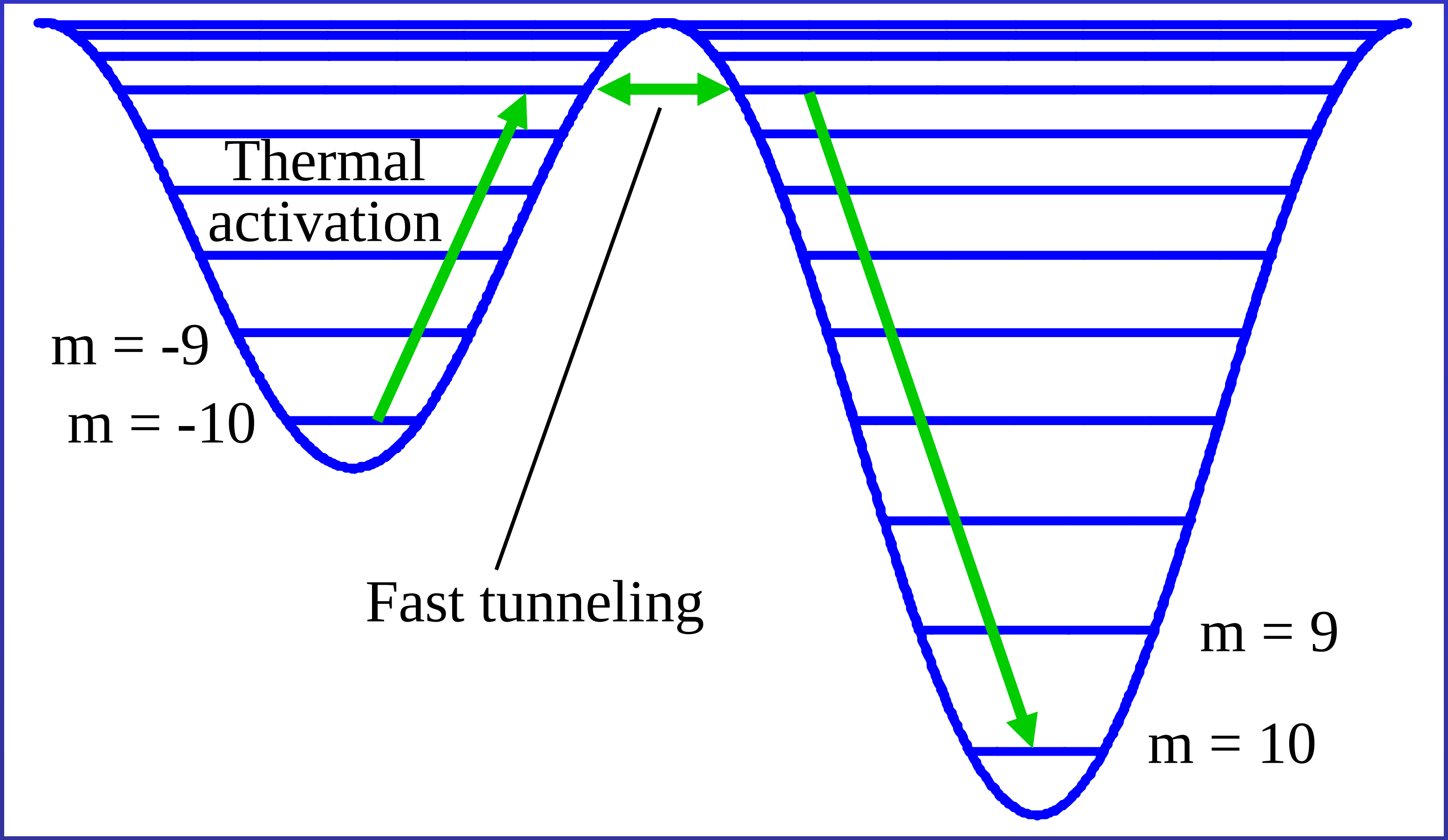
Enhanced Relaxation at Step Fields



Enhanced Relaxation at Step Fields



Thermally Assisted Resonant Tunneling



Tunneling occurs when levels in opposite wells align.

Spin Hamiltonian for Mn_{12}

$$\mathcal{H} = -DS_z^2 - g\mu_B \mathbf{S}\cdot\mathbf{H}$$

The field at which $|m\rangle$ (in the left well) crosses $|-m+n\rangle$ (in the right well):

$$H_{m,-m+n} = \frac{-Dn}{g\mu_B}$$

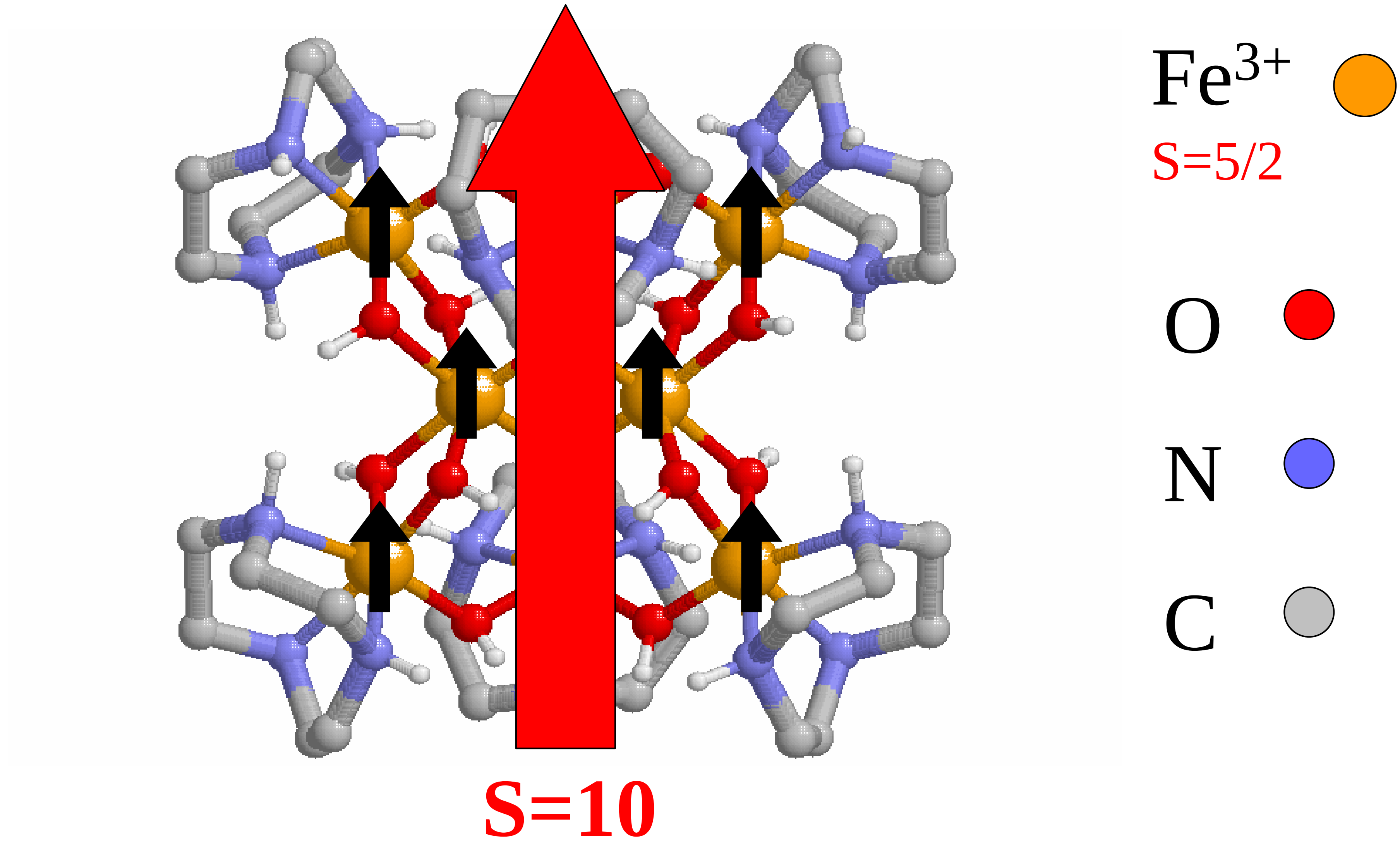
Steps occur at regular intervals of field, as observed.

Step occurs every 4.5 kOe $\Rightarrow$ **D/g = 0.31 K**

Compare with ESR data (e.g. Barra et al., PRB, 1997) :

D = 0.56 K, g = 1.93 **D/g = 0.29 K**

$[Fe_8O_2(OH)_{12}(tacn)_6]Br_8.9H_20$



Fe_8 Hamiltonian

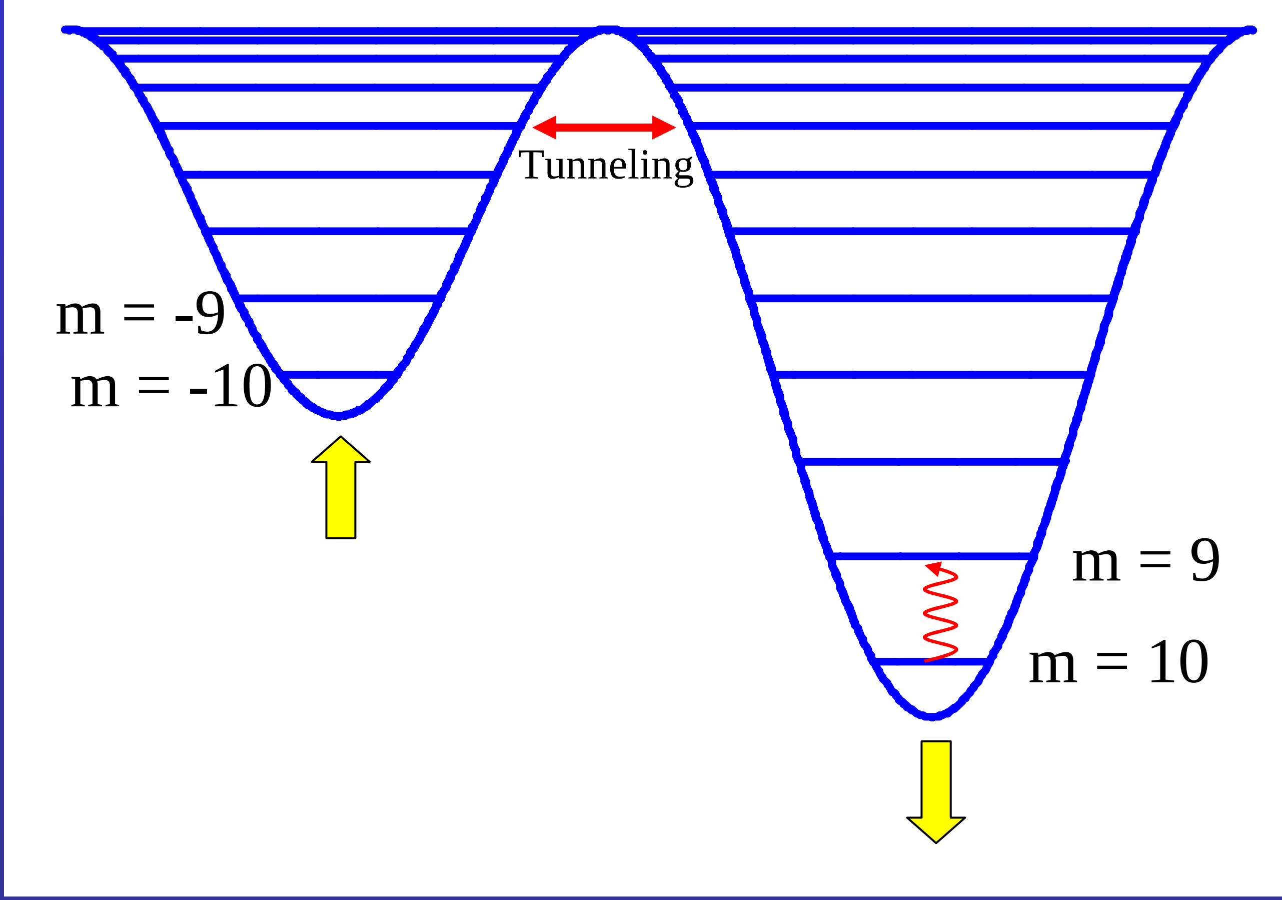
$$\mathcal{H} = -DS_z^2 + E(S_x^2 - S_y^2) + C(S_+^4 + S_-^4) - g\mu_B \vec{S} \cdot \vec{H}$$

$D = 0.292\,\mathrm{K}$

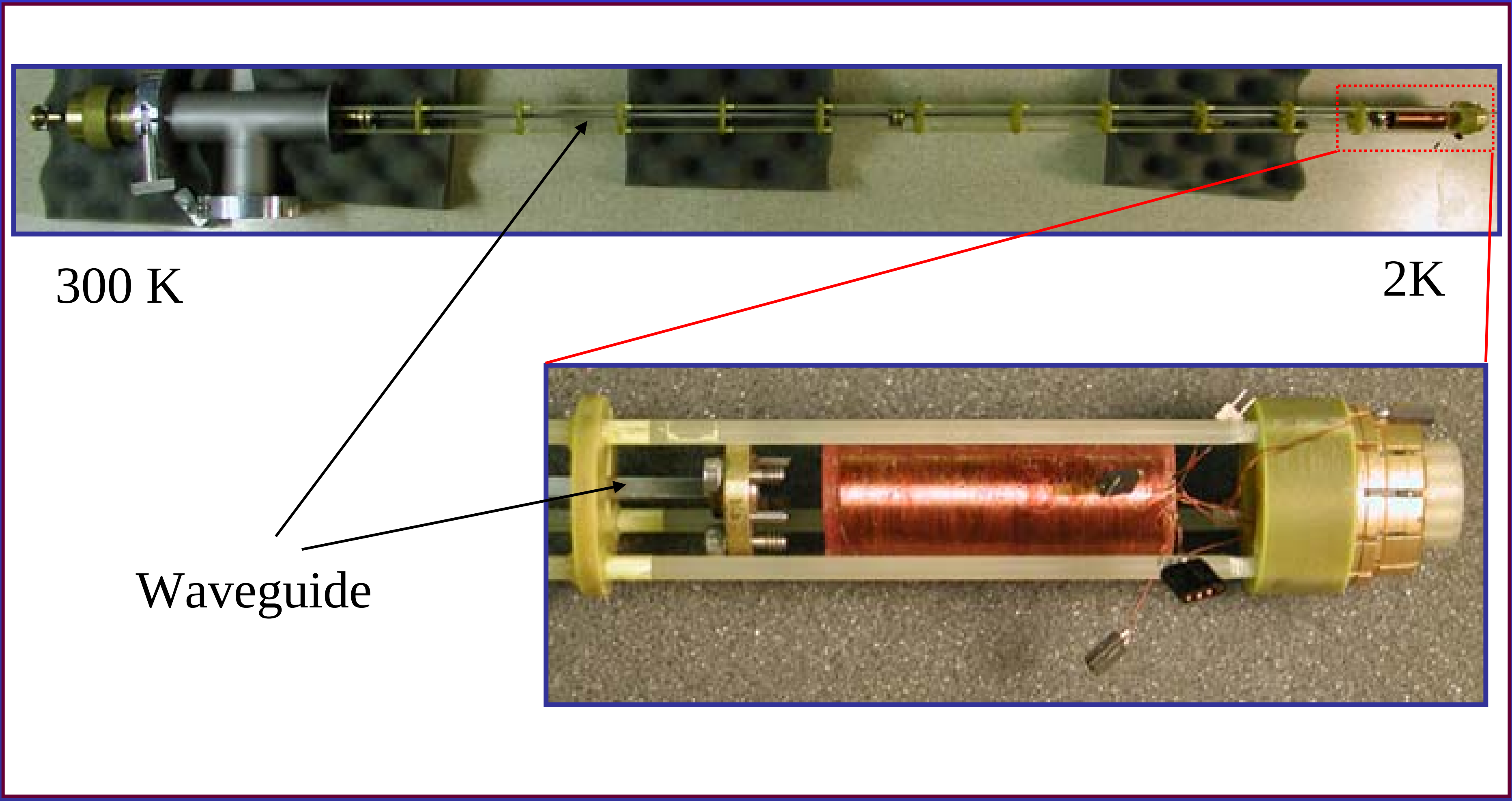
$E = 0.046\,\mathrm{K}$

$C = -2.9 \times 10^{-5}\,\mathrm{K}$

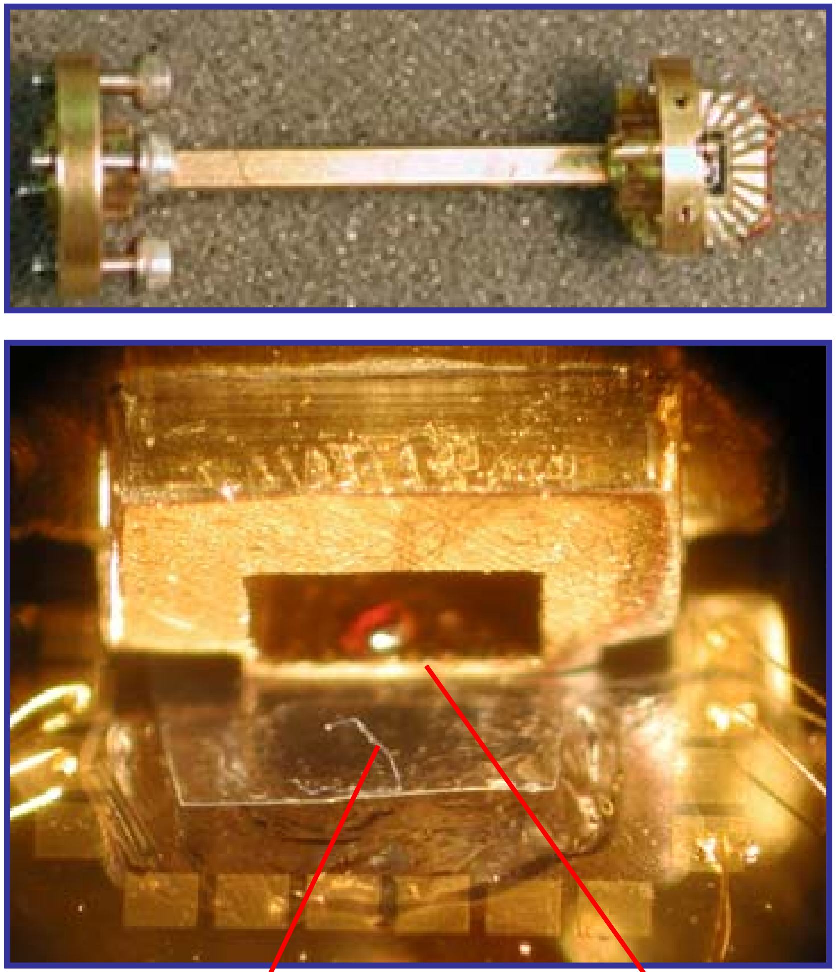
$g = 2$



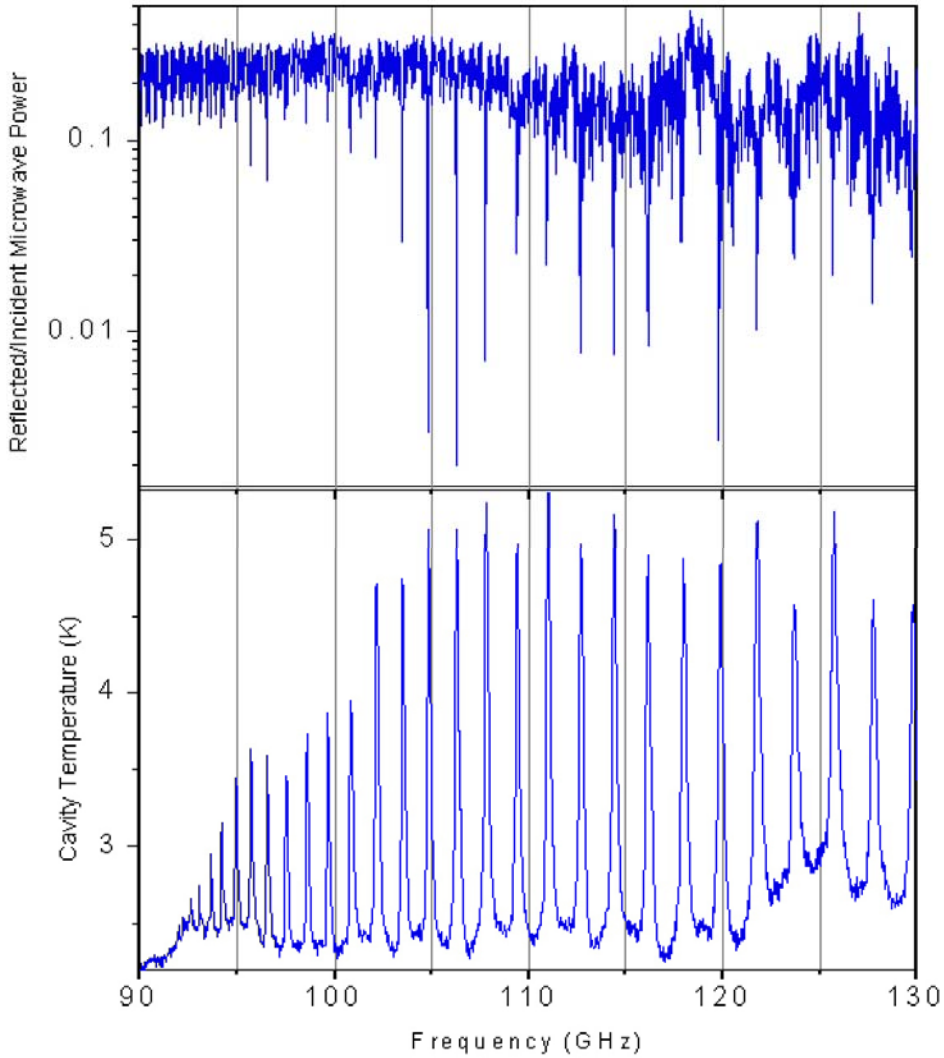
Low Temperature Millimeter Wave Probe



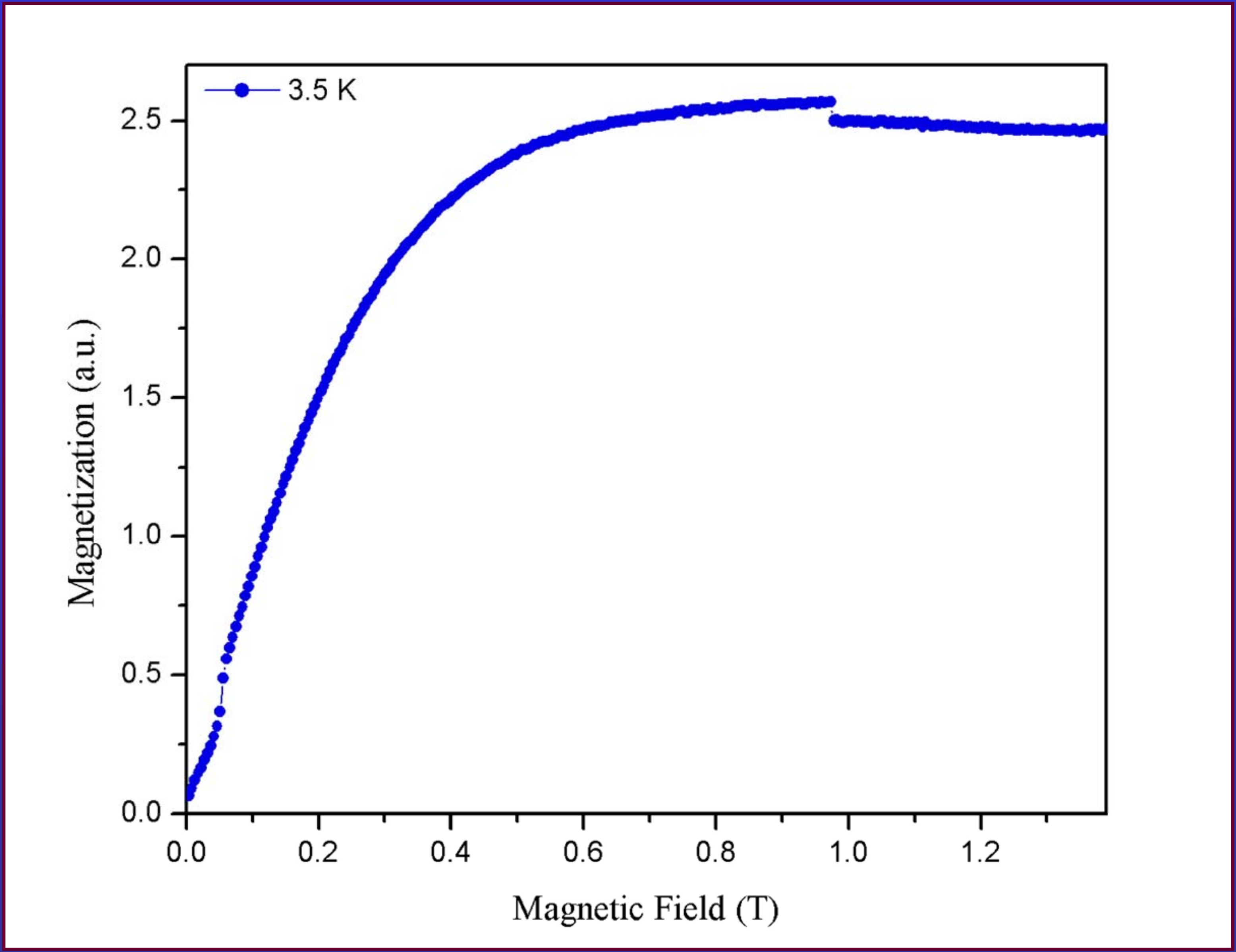
Experimental Cavity and its Resonances



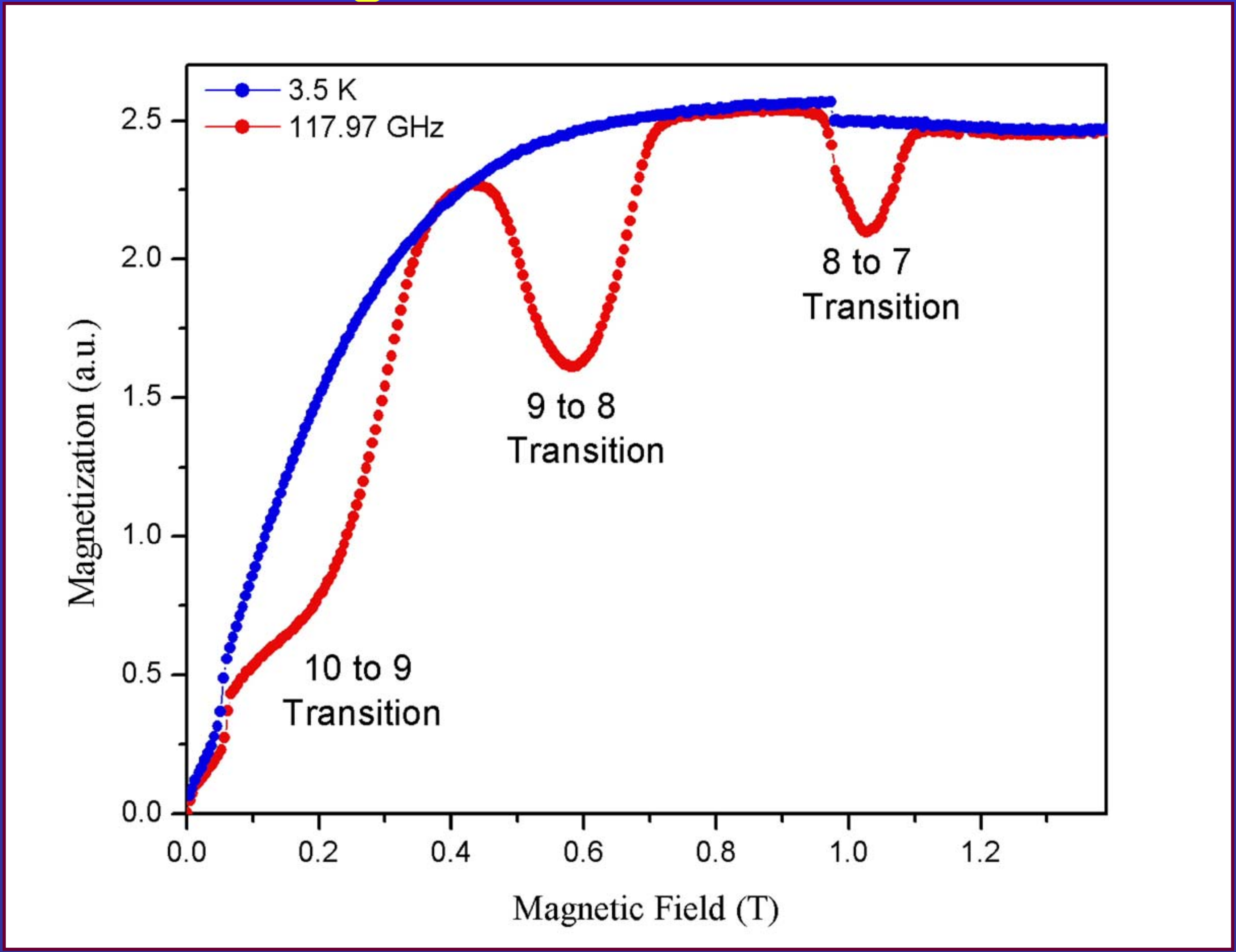
Hall Bar Detector Fe_8 Crystal



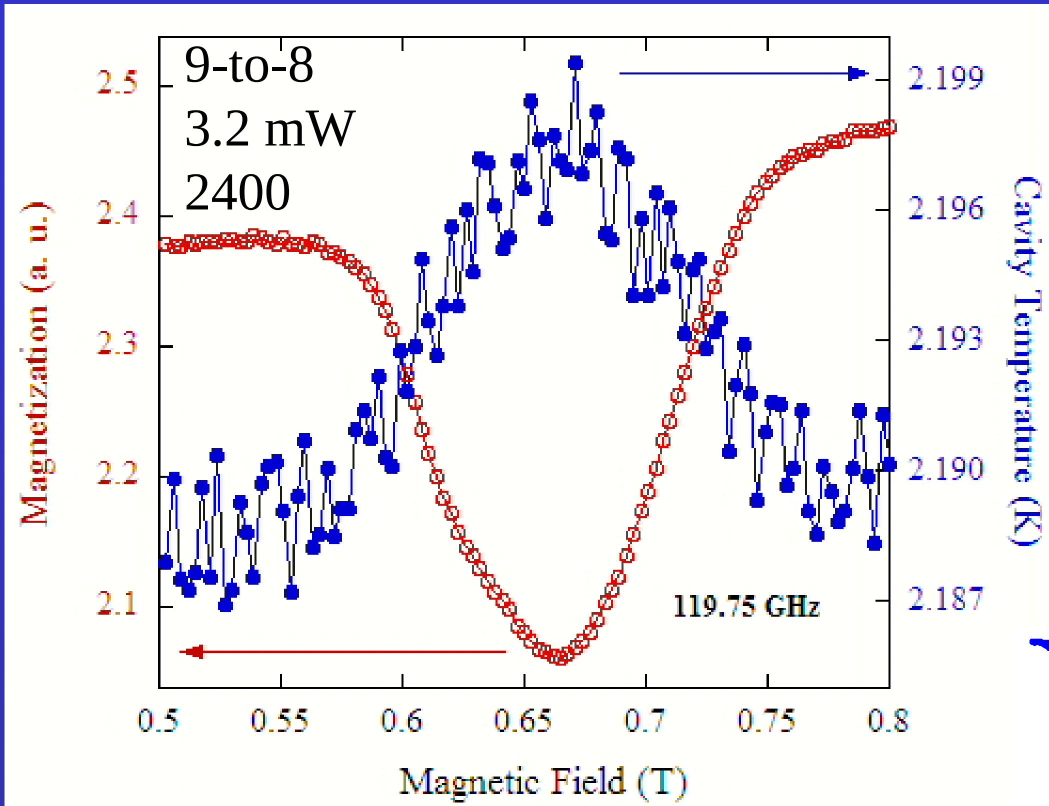
Equilibrium Magnetization of Fe_8



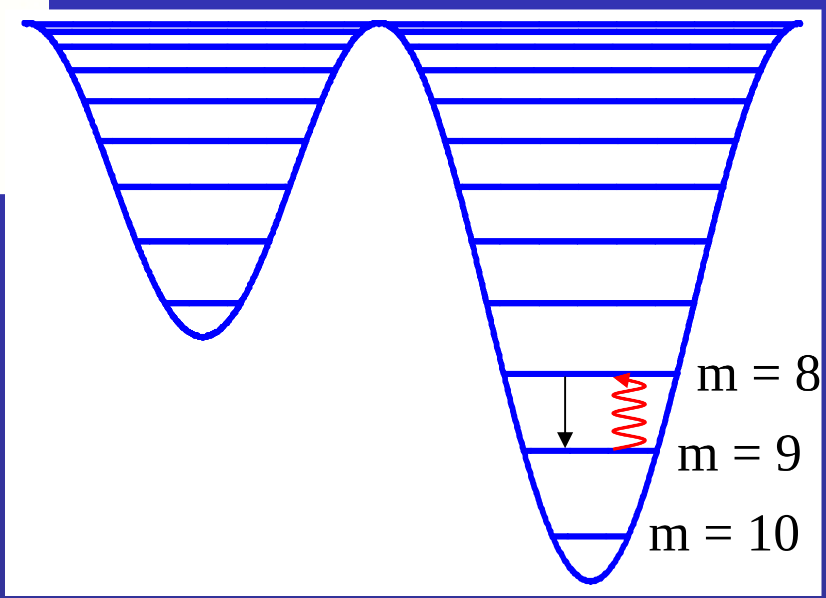
Equilibrium Magnetization of Fe_8 with and without High Power Radiation



Radiation Induced Effects: Heating



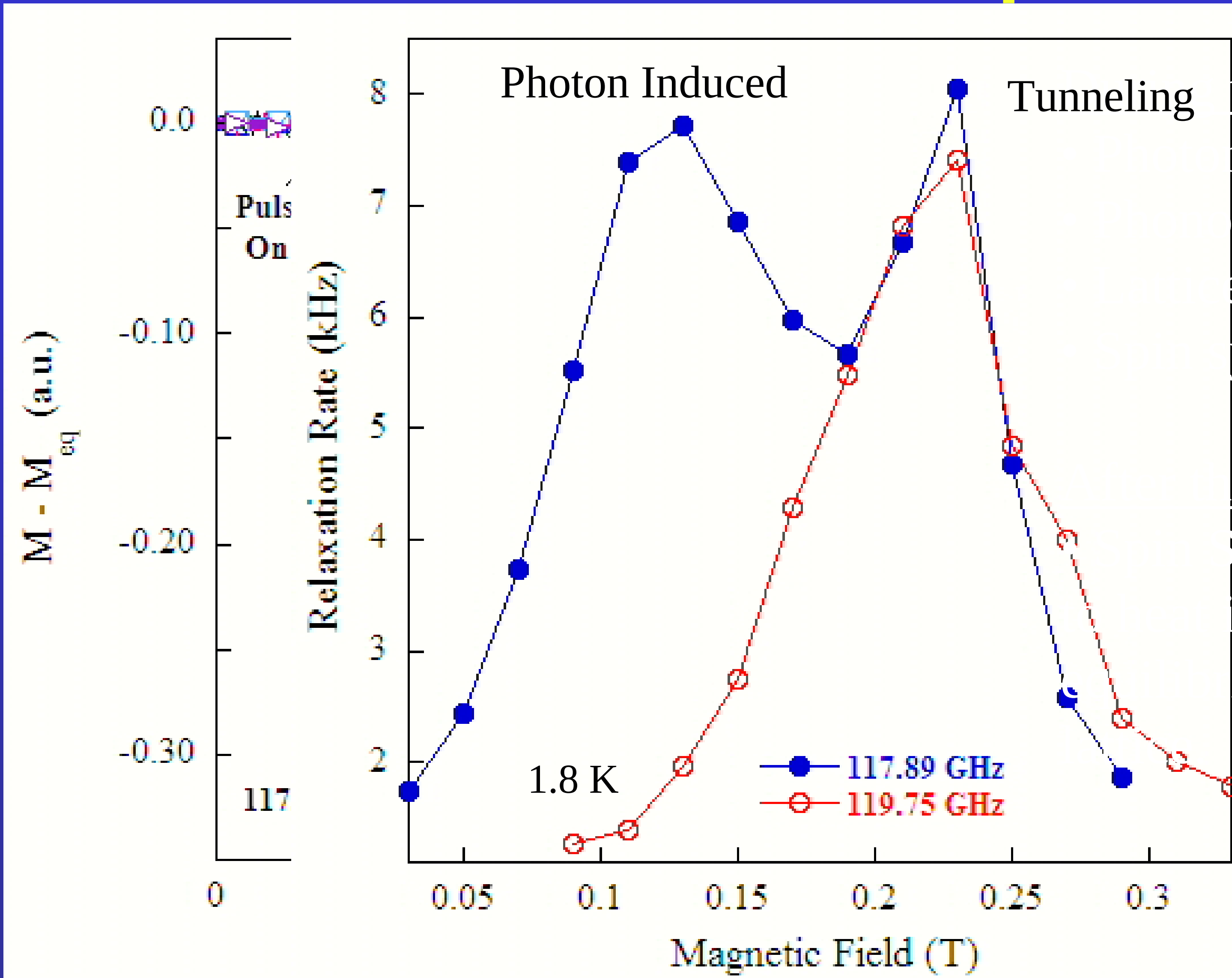
Heating



- Photon Absorbed
- Phonon Emitted
- Lattice Heats
- Spin System Heats
- Population of 9 increases

Positive Feedback

Pulsed Radiation Experiments



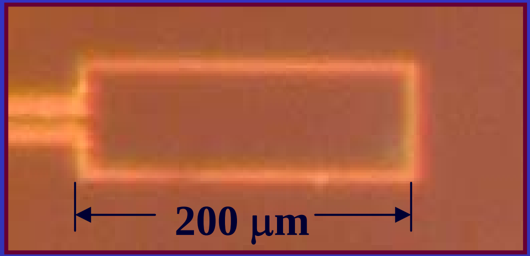
the Pulse
n absorbed.
n emitted.
e heats.
ystem heats.

e Pulse
ystem Continues
n establishing
ium with the

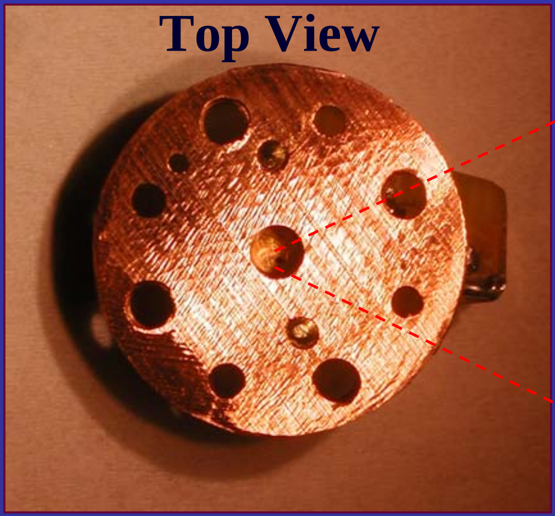
- M. Bal et al., Europhys. Lett **71**, 110 (2005) and J. Appl. Phys., **99**, 08D102 (2006).
- See also K. Petukhov et al., Cond-mat/0502175 (2005).

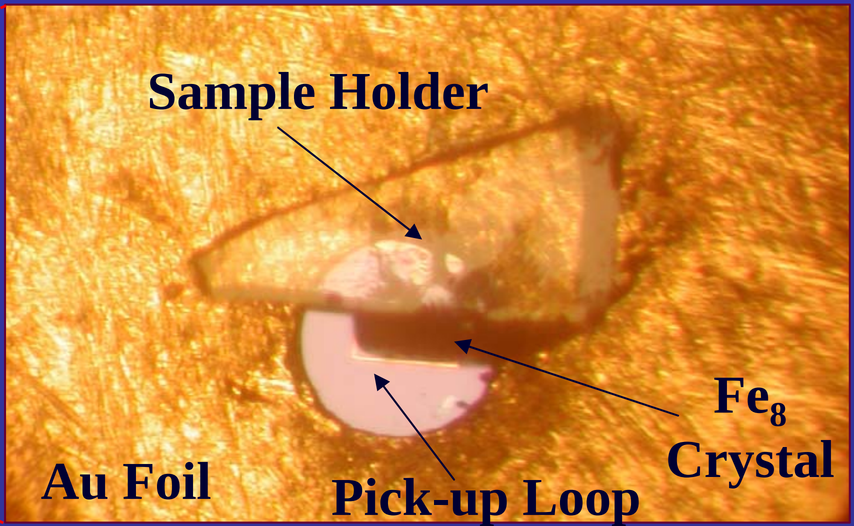
Measuring Fast and Small Magnetic Signals

- **Hall bar detectors used in our previous studies are slow.**
- **Inductive Pick-up Loop coupled to SQUID voltmeter as a fast detector.**

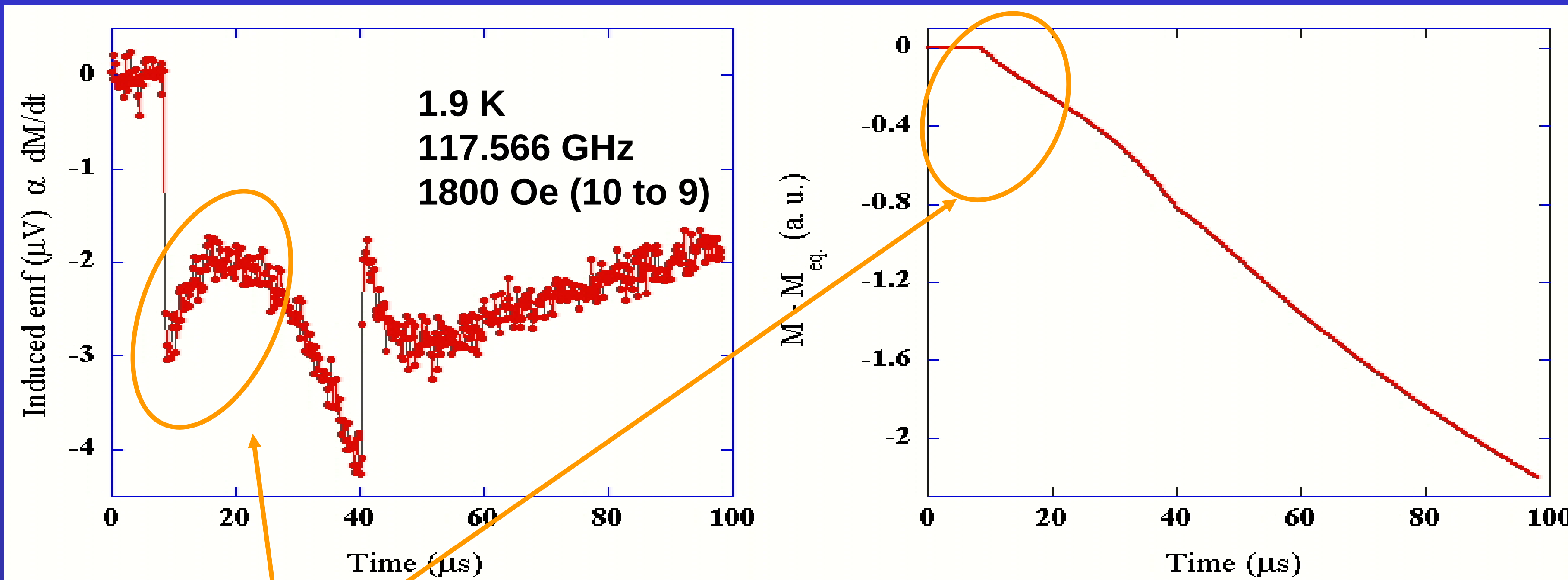


- **High-Q Cylindrical cavity (Q ~ 6500)**





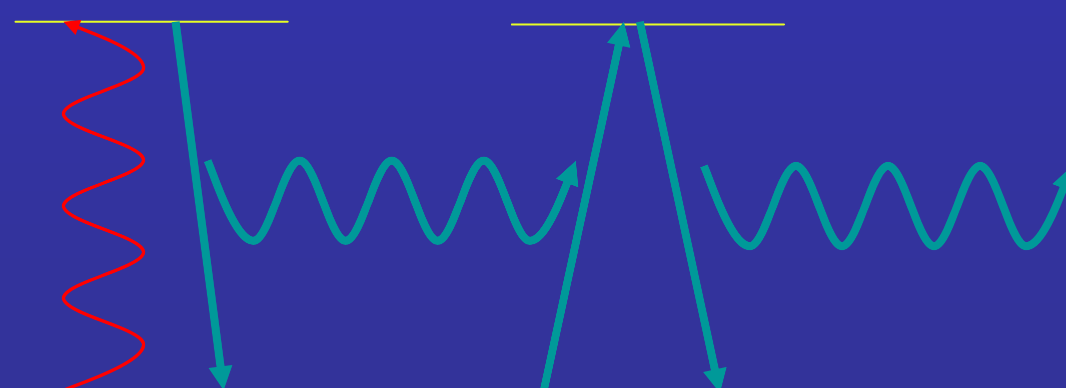
Radiation Induced Magnetization Changes at Short Time Scales



Phonon Bottleneck

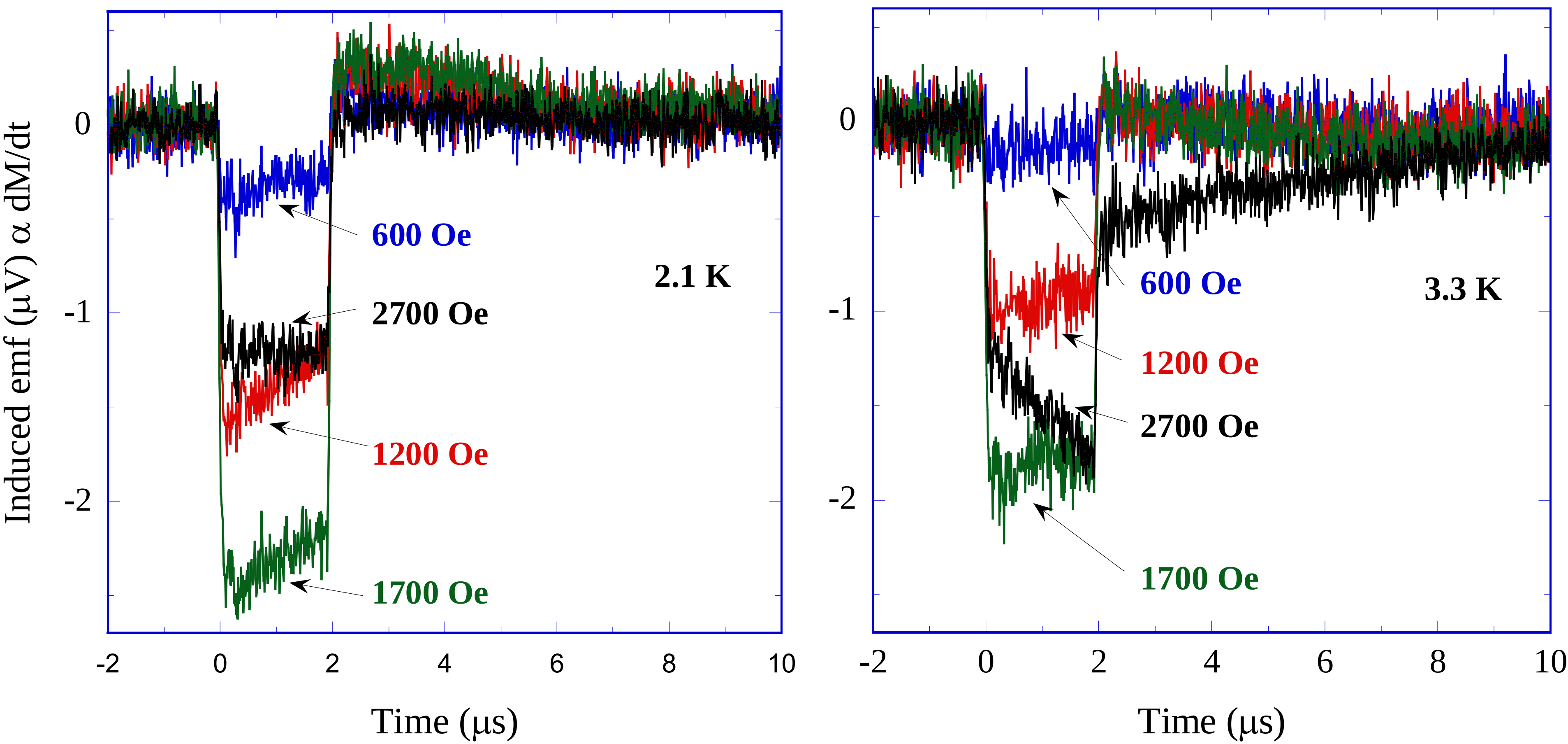
~5 µs

- What is this Decay?
- Phonons are Repeatedly Emitted and Reabsorbed
- Increased Excited-State Population

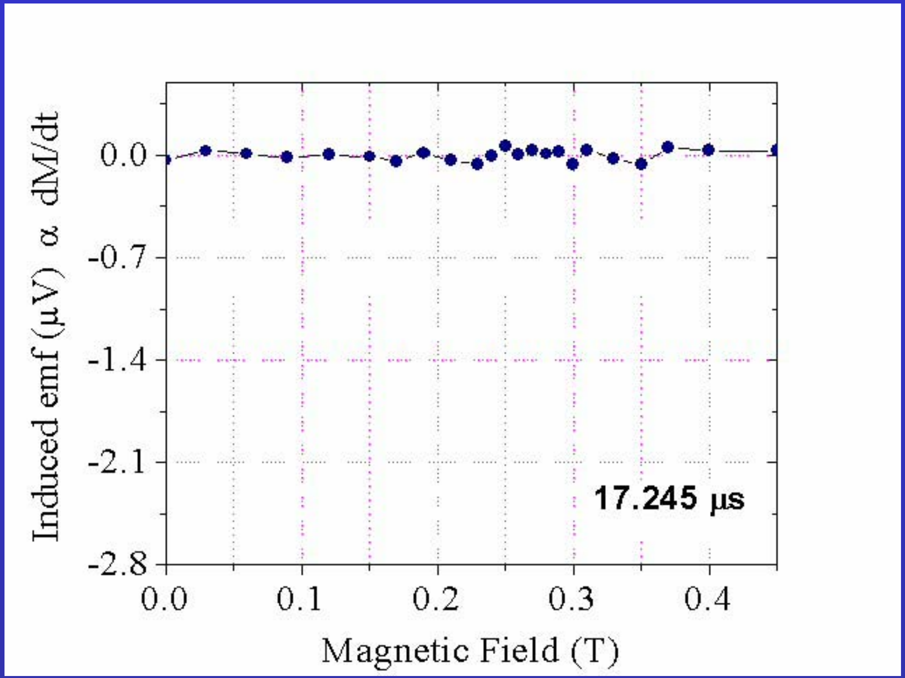


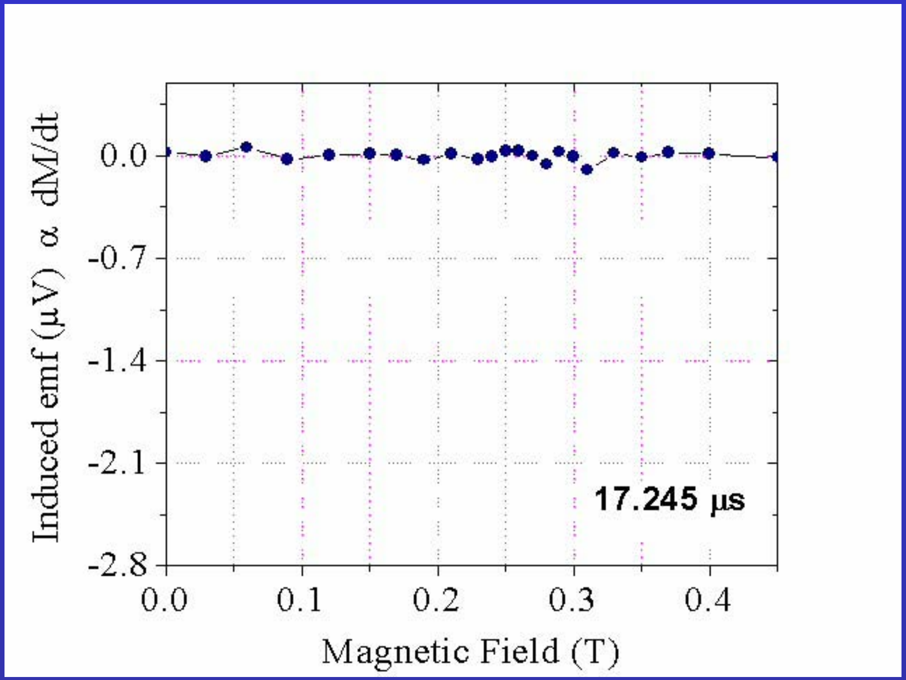
Short (2 μs) Pulses

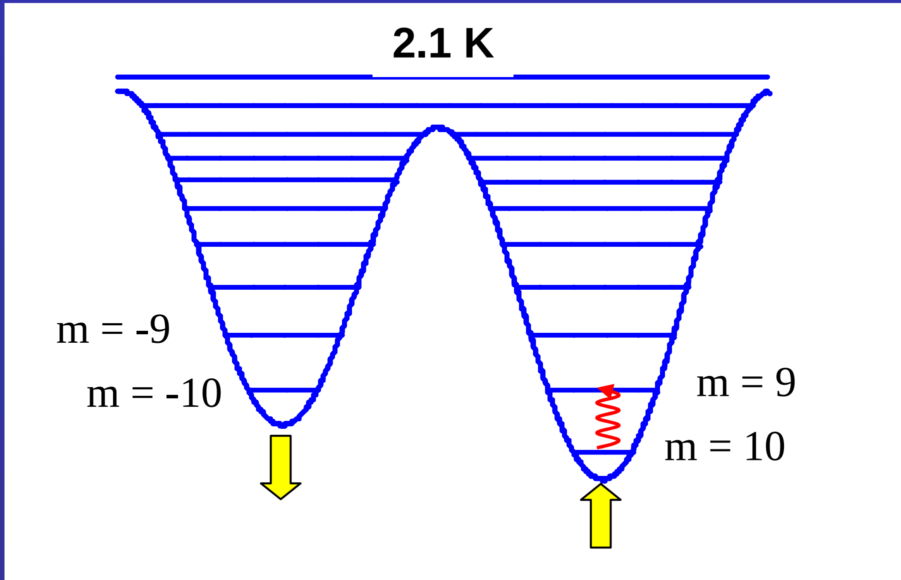
10-to-9 Transition

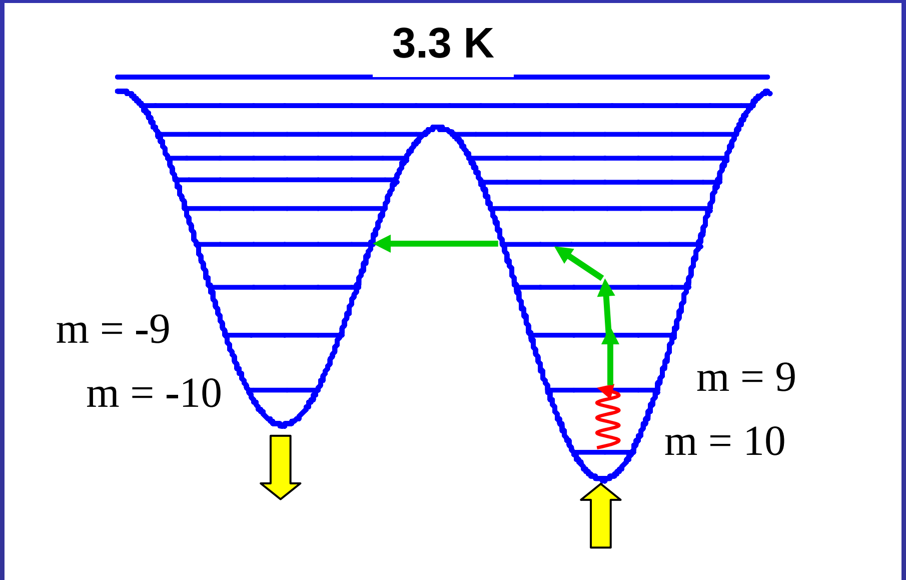


Magnetization Dynamics in "Real Time"

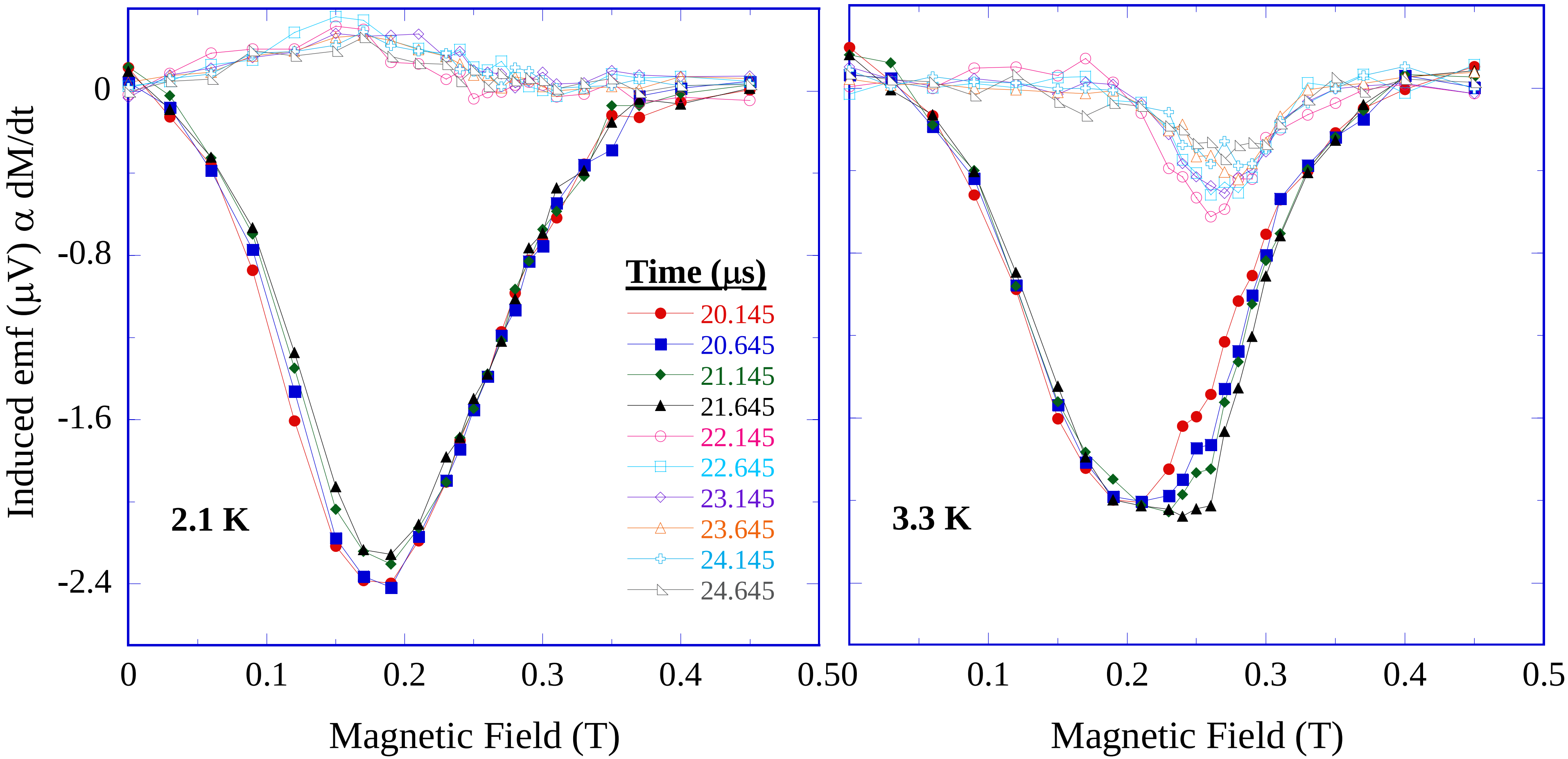








Photon/Phonon Assisted Tunneling



- Tunneling begins during radiation pulse and continues after pulse is turned off.

Simulations

Phonon transition rates

$$\frac{dP_i}{dt} = -\sum_{\substack{j=1 \\ i\neq j}}^{21} \left(\gamma^{+}_{i,j} + \gamma^{-}_{i,j} + w_{i,j}\right) P_i + \sum_{\substack{j=1 \\ i\neq j}}^{21} \left(\gamma^{+}_{j,i} + \gamma^{-}_{j,i} + w_{j,i}\right) P_j$$

Photon transition rates

- Work in energy eigenbasis (not spin eigenbasis)
- Tunneling included automatically

$$w_{i,j} = \frac{\pi (H_1 g \mu_B)^2}{2\hbar^2} \left|\langle i | S_x | j \rangle\right|^2 F(\omega)$$

$$\gamma^{\mp}_{i,j} = \frac{g_{ph}}{24\pi\rho c_s^5 \hbar^4} \left|\langle i | \{S_{\mp}, S_z\} | j \rangle\right|^2 \left(\varepsilon_i - \varepsilon_j\right)^3 N^{ph}_{i,j}$$

Phonon number for i -> j transition

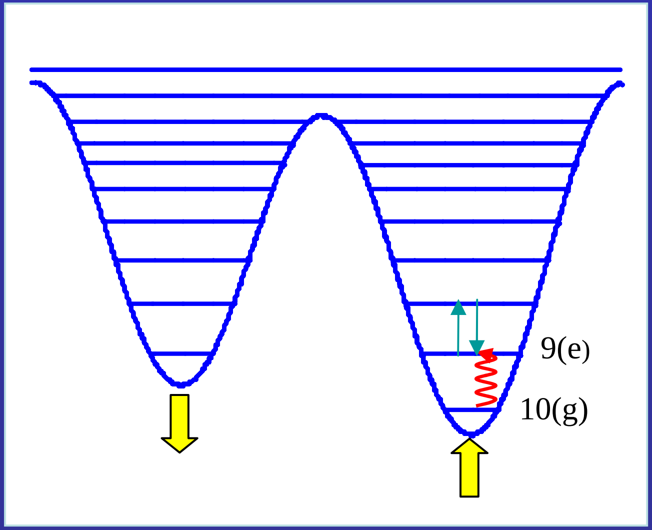
$$N^{ph}_{i,j} = \frac{1}{e^{\frac{(\varepsilon_i - \varepsilon_j)}{T}} - 1}$$

(thermal, all but 10 (g) <-> 9 (e))

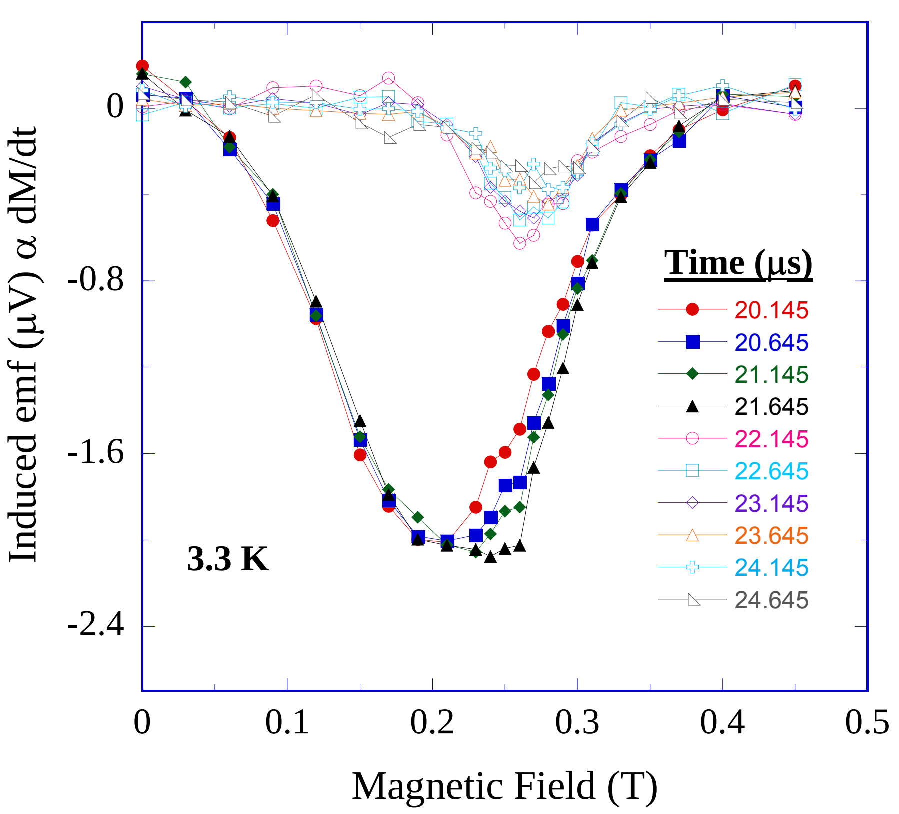
Phonon bottleneck number (for 10 (g) <-> 9 (e)):

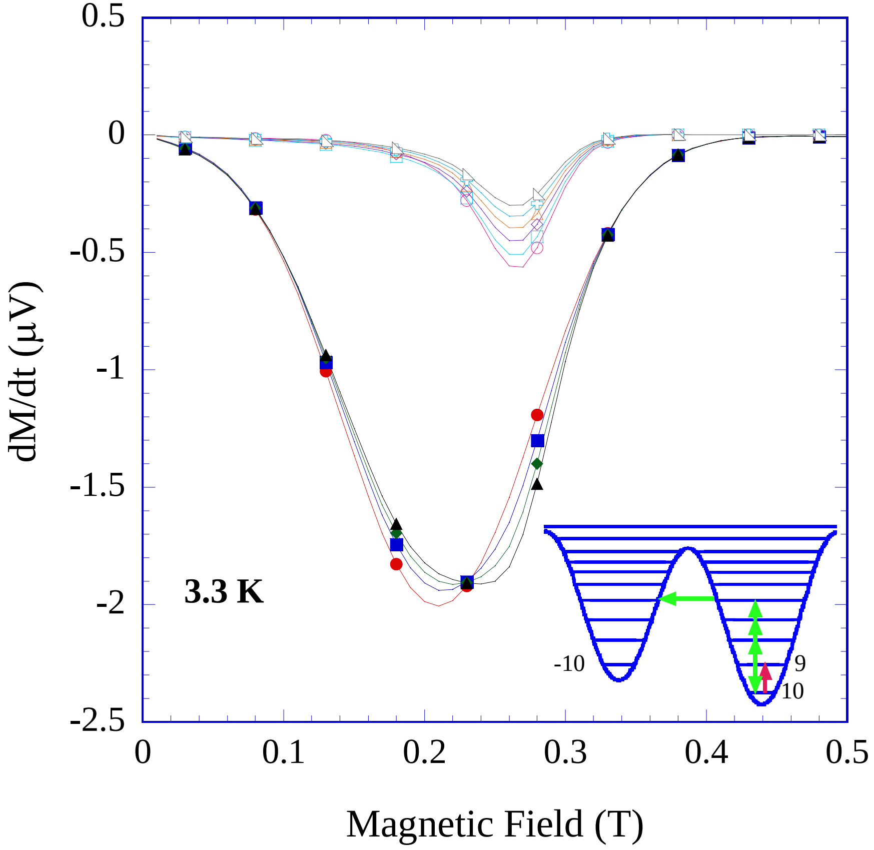
$$\frac{dn_{pb}}{dt} = -P_g \frac{\left(\gamma^{+}_{g,e} + \gamma^{-}_{g,e}\right)}{N^{ph}_{g,e}} n_{pb} + P_e \frac{\left(\gamma^{+}_{e,g} + \gamma^{-}_{e,g}\right)}{N^{ph}_{e,g}} \left(n_{pb} + 1\right) - \frac{\left(n_{pb} - N^{ph}_{e,g}\right)}{t_{pb}}$$

Absorption — Emission — Decay



Simulation Results





Parameters: Anisotropy D = 0.290 K

Line width σ = 650 Oe

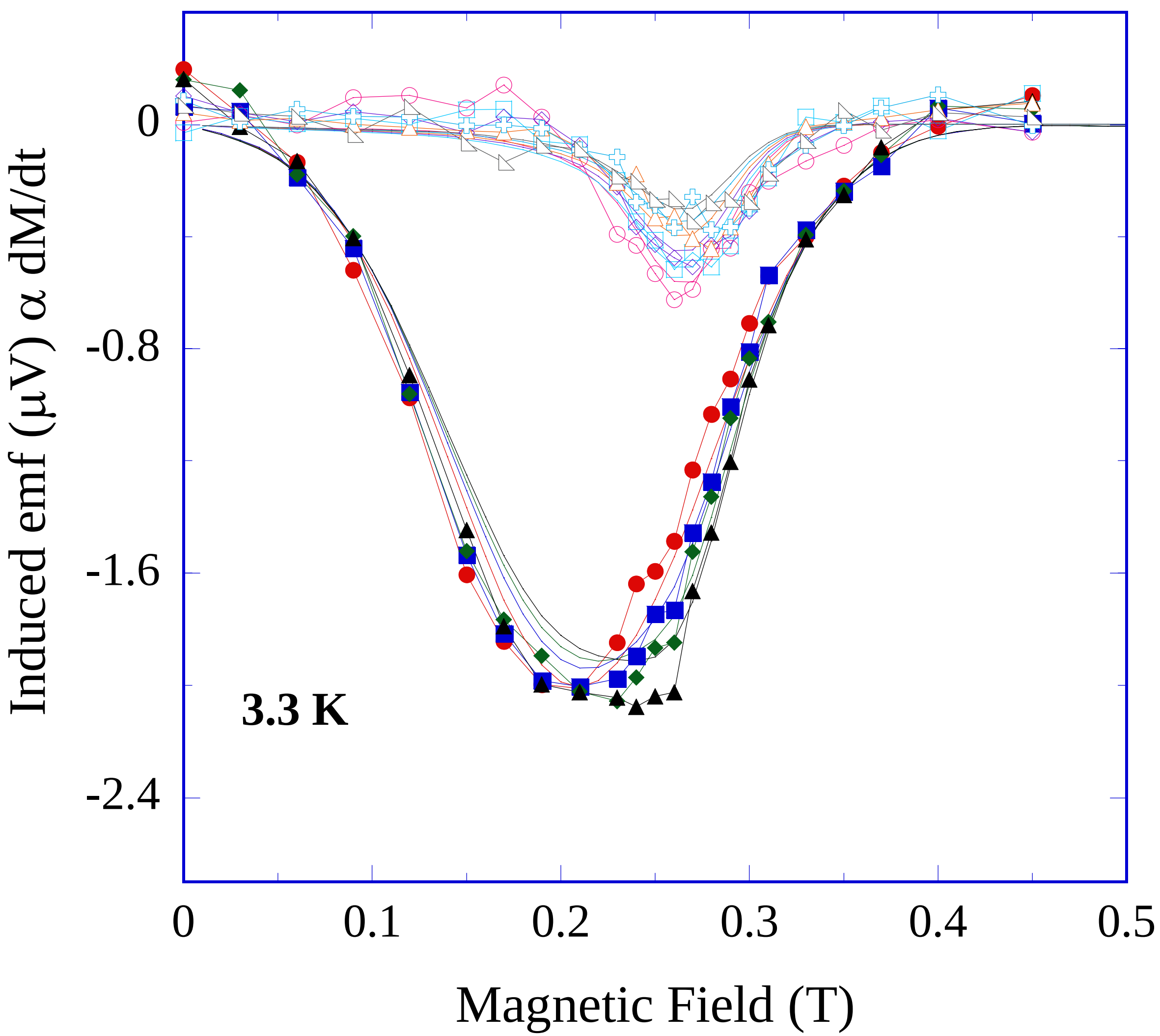
rf field H_1 = 1.0 Oe

speed of sound c_s = 670 m/s

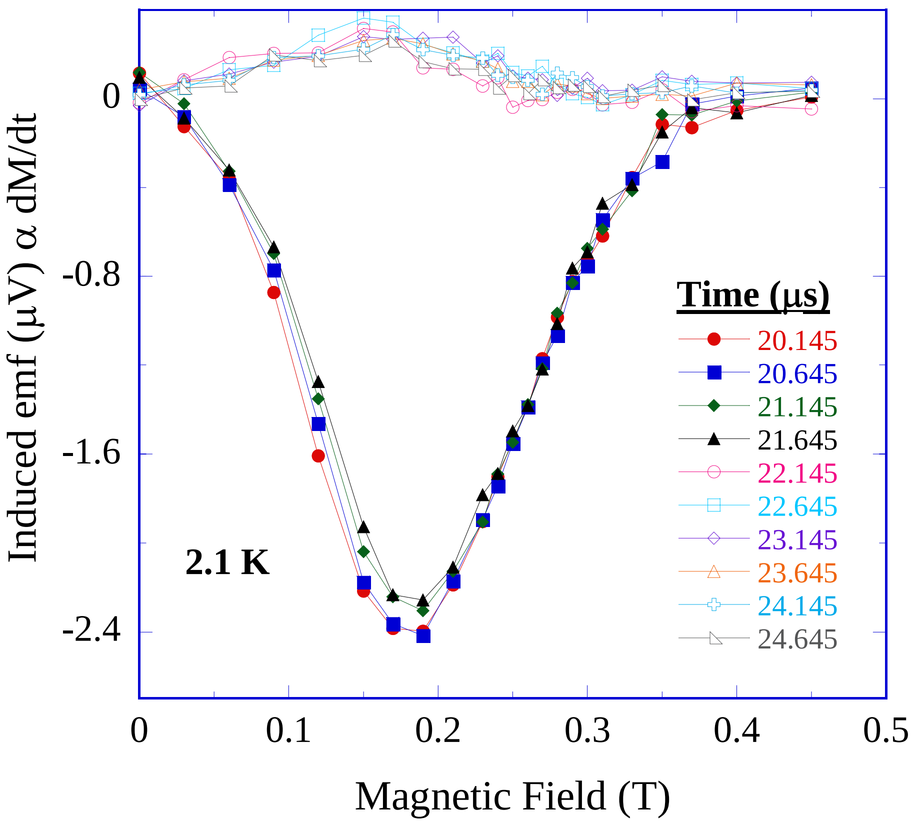
[800 m/s in Evangelisti, et al., PRL 2005]

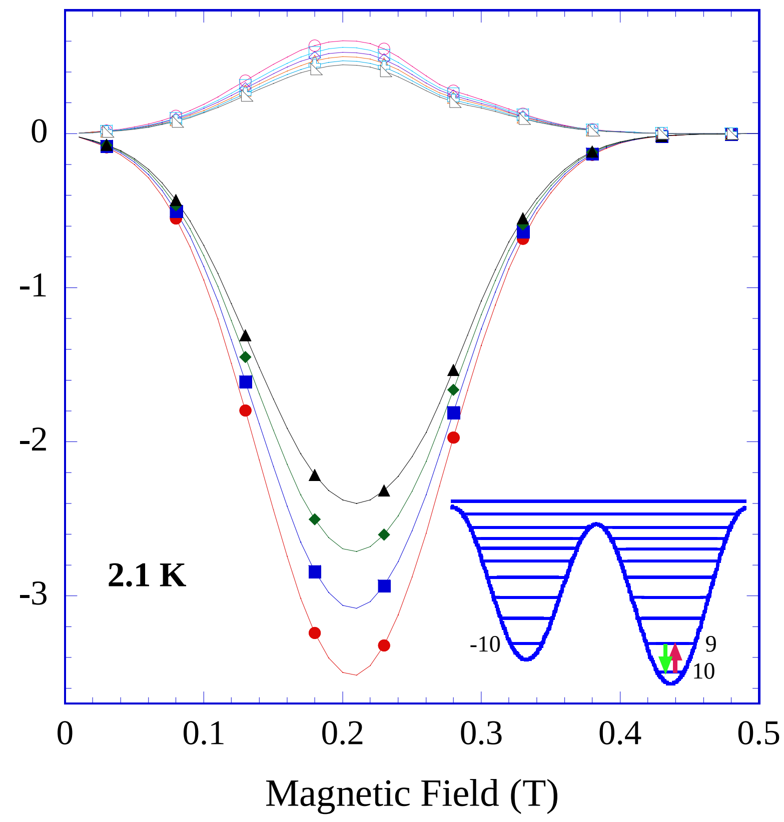
inhomogeneous broadening: 200 Oe

Simulation Results

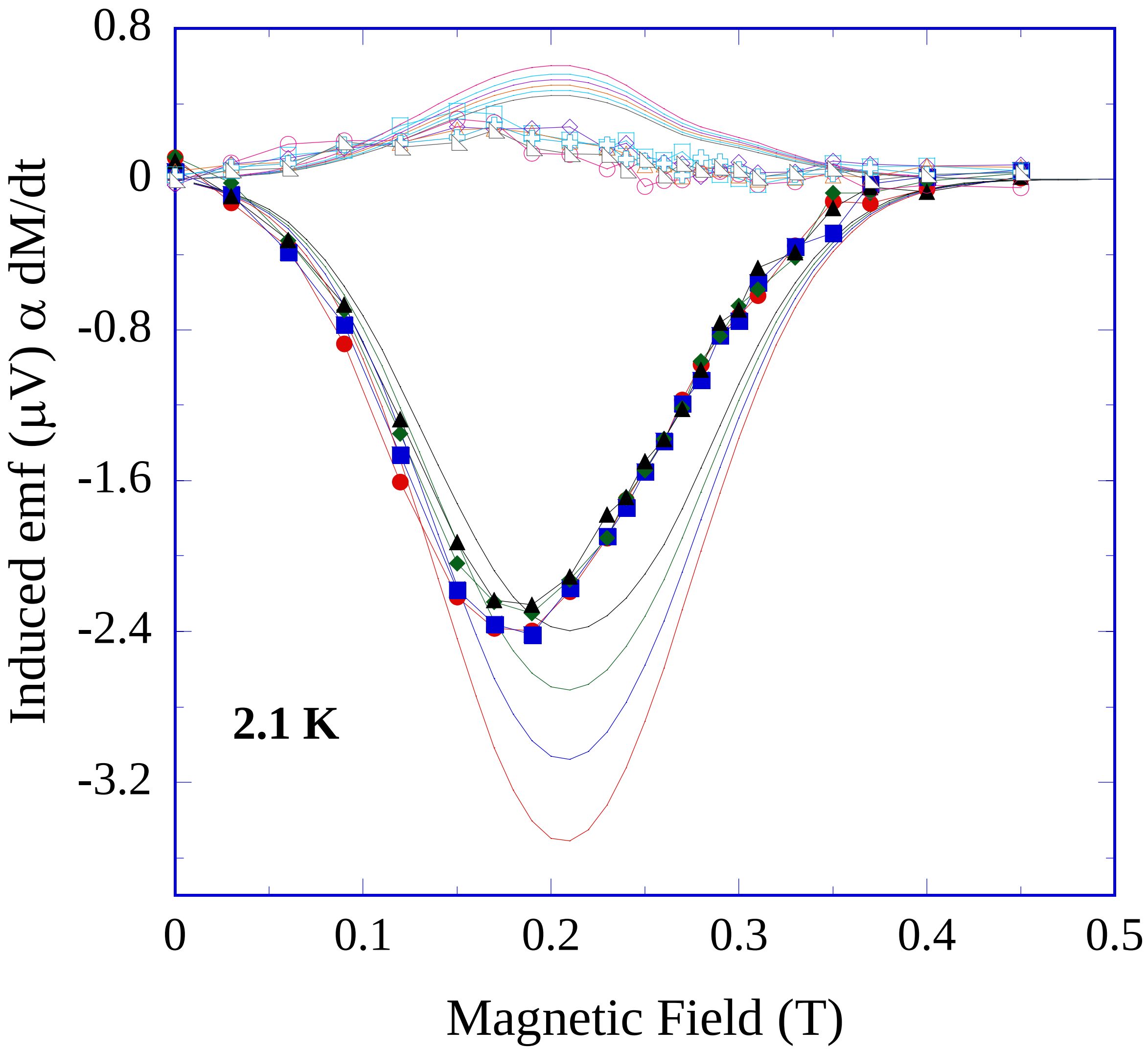


Simulation Results





Simulation Results



Line shift also observed in ESR: Park, et al., PRB (2002).

Conclusions

- Resonant radiation drives the spins and phonons out of thermal equilibrium – Heating.
- Phonon bottleneck in Fe_8 with decay time ~5 μs.
- Phonon bottleneck and thermally assisted tunneling from excited state induced by resonant radiation.