Energy Bursts from Fe8 Molecular Magnet in the Quantum Tunneling Of Magnetization Process

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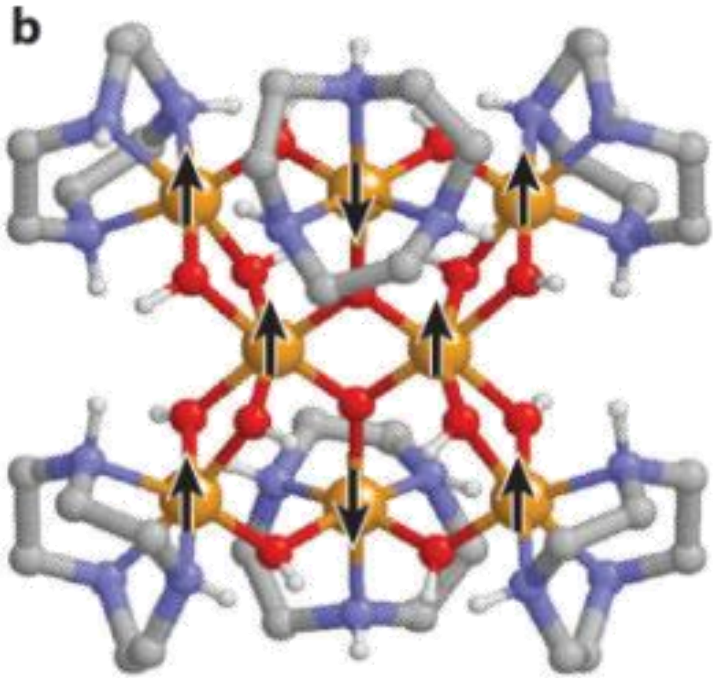
Molecular Magnets

Fe8 Molecule – Single Molecule Magnet

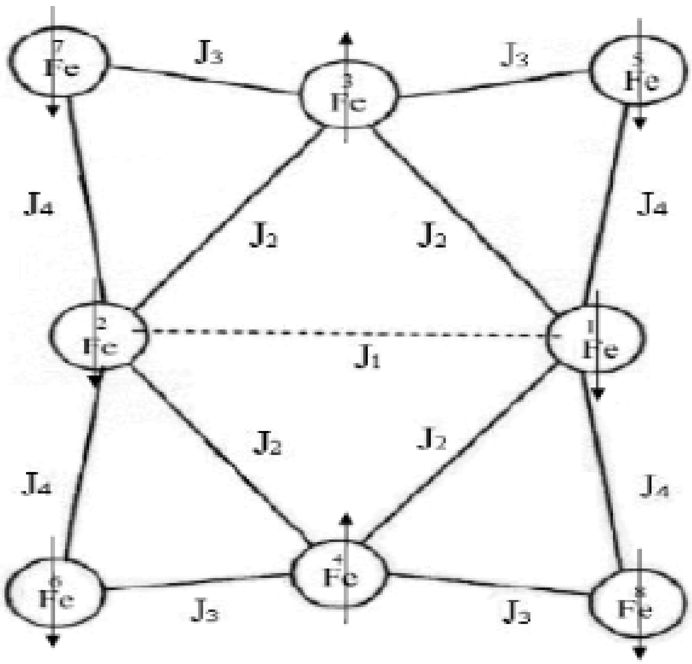
$[Fe_8O_2(OH)_{12}(C_6H_{15}N_3)_6]^{8+}$.

The antiferromagnetic interactions between the spins of the ions are between 20 to 170K.
Below that the spins are locked



● Iron
● Oxygen
● Nitrogen
○ Carbon
○ Hydrogen

J_1= -147K,
J_2= -173K,
J_3= -22K ,
J_4= -50K



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[1] K. Wieghardt, K. Pohl, I. Jibril and G. Huttner, Angew. Chem. Int. Ed. Engl. 23 (1984), 77.
[2] C. Delfs, D. Gatteschi, L. Pardi, R. Sessoli, K. Wieghardt and D. Hanke, Inorg. Chem. 32, 3099 (1993).

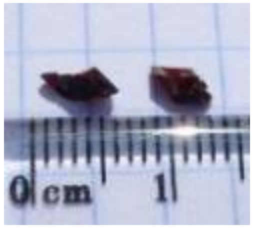
Fe8 Molecule – Single Molecule Magnet

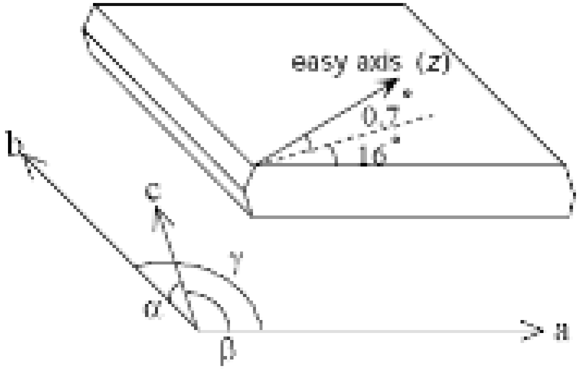
- 8 Fe^{3+} ions (S = 5/2):
  - 2 ions with spin down: S = 5
  - 6 ions with spin up: S = 15
  - Total **S = 10**

- Magnetic interactions between the molecules are small (~0.05K).

- Single crystals of Fe_8 are grown from a solvent.

- Orientation is made by a microscope.





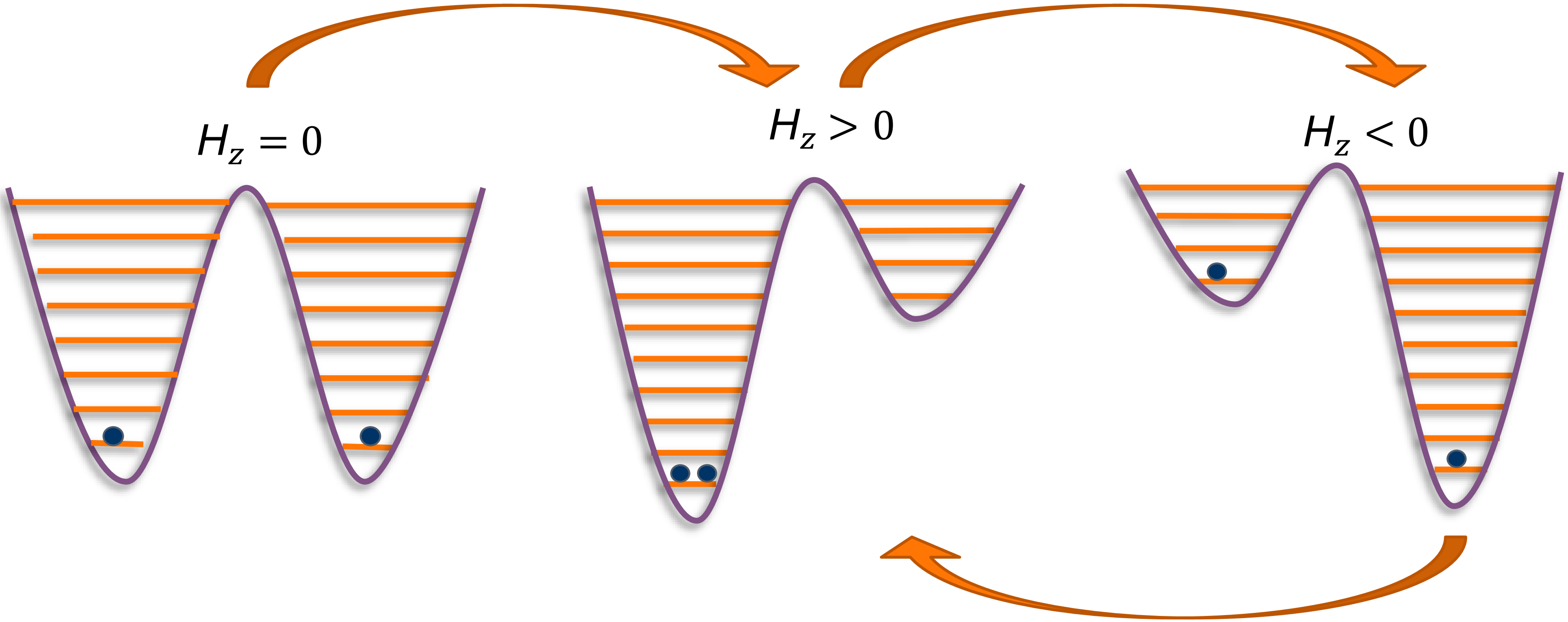
M. Ueda & S. Maegawa, J. Phys. Soc. Jpn. 70 (2001)

The Crystal field Hamiltonian

$$\mathcal{H} = DS_z^2 + E\left(S_x^2 - S_y^2\right) - g\mu_B S_z H_z$$

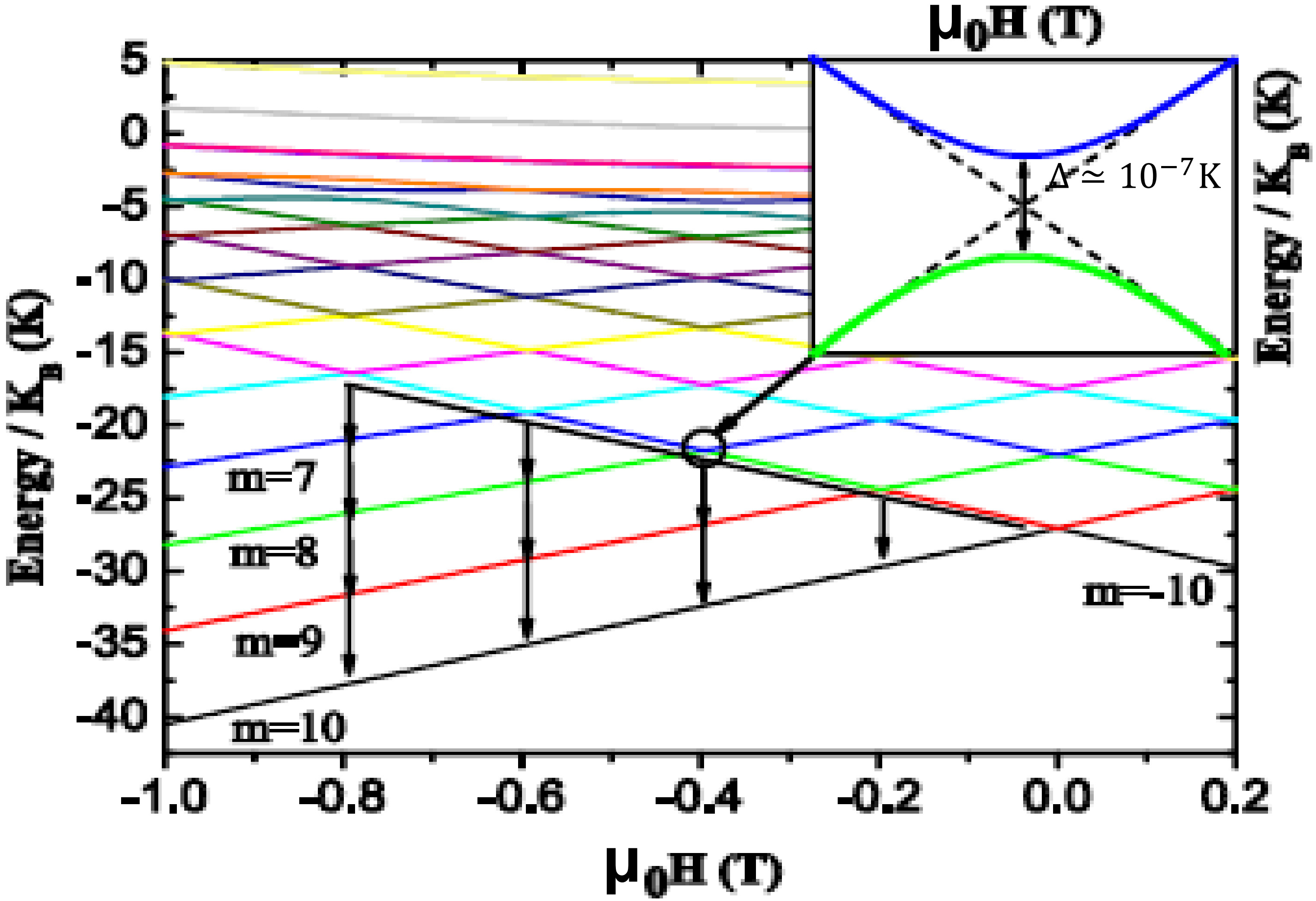
D ~0.29 K E ~0.045 K

The Experiment



Relaxation processes

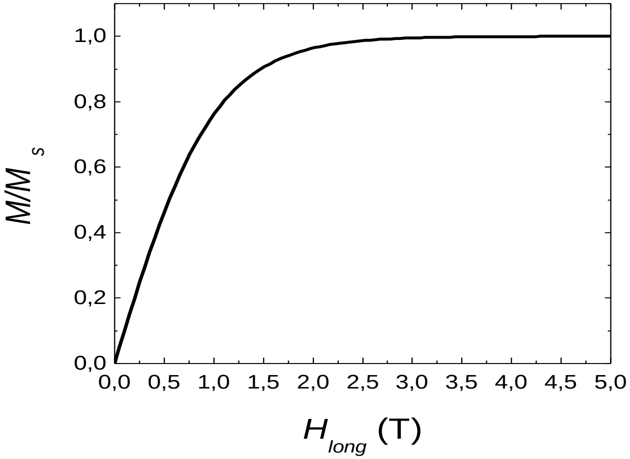
Energy Levels for S=10



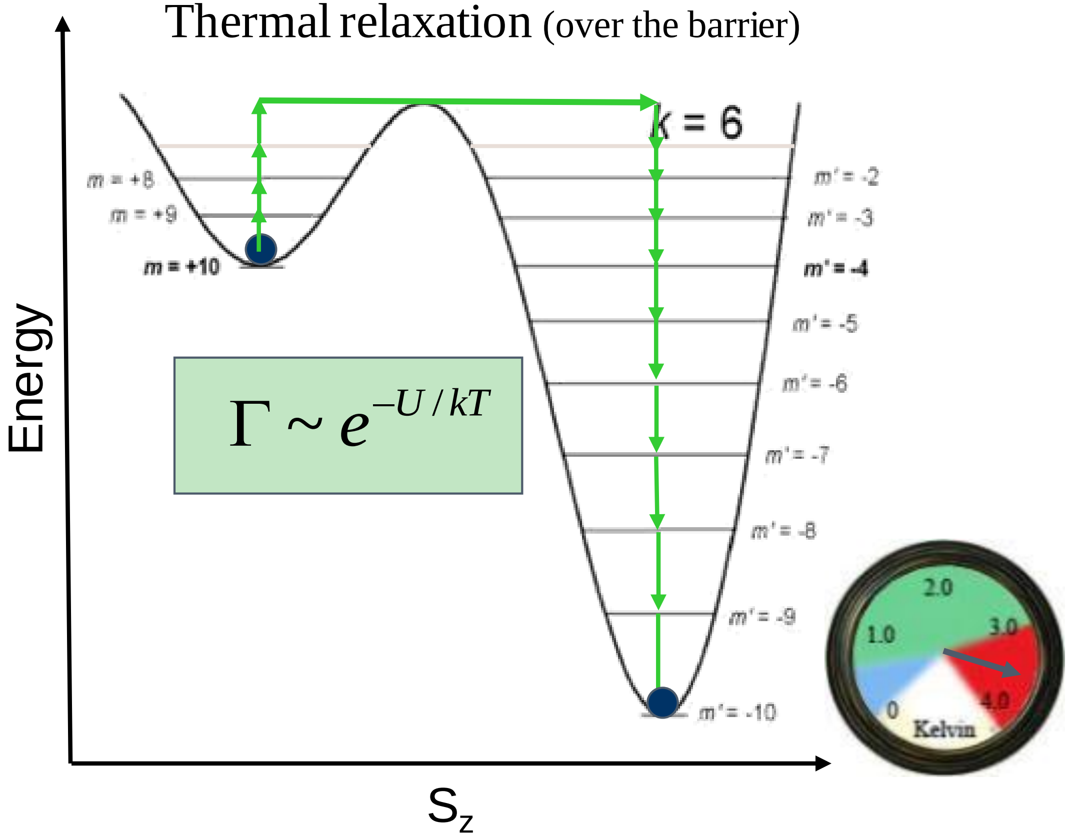
Wernsdorfer et al. (J. Appl. Phys. **87** (2000), 5481)

Relaxation processes

Magnetic field relaxation at high temperature for a single molecule



Thermal relaxation (over the barrier)

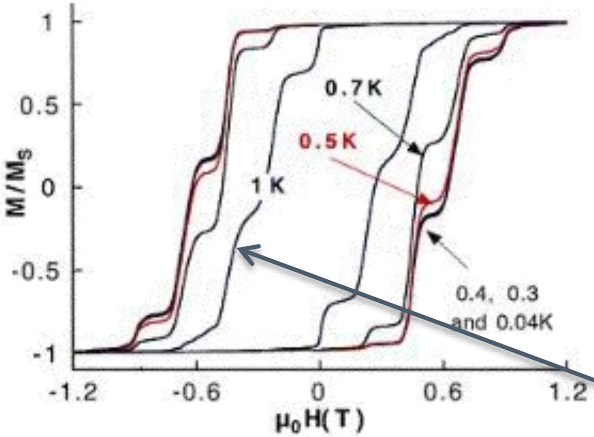


$$\Gamma \sim e^{-U/kT}$$

Relaxation processes

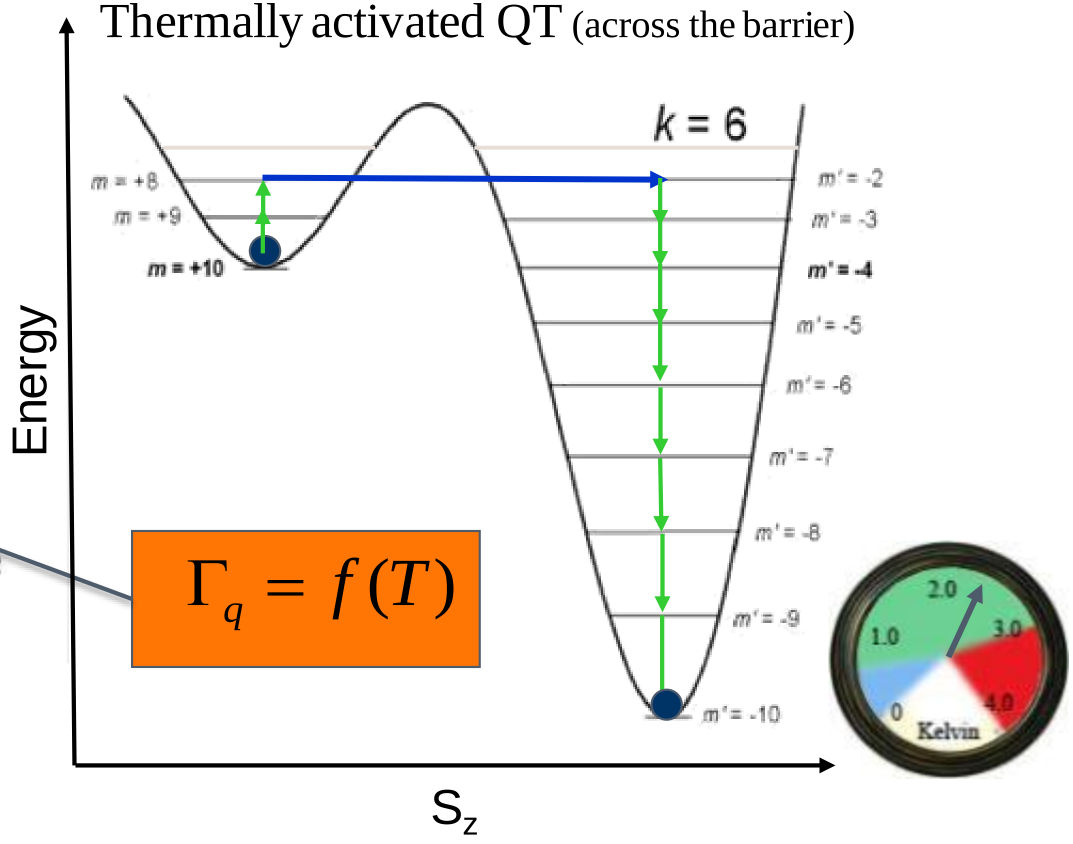
Magnetic field relaxation at low temperature for a single molecule

Hysteresis loop of Fe8



W . Wernsdorfer et al. J. Appl. Phys. 87 (2000), 5481

Thermally activated QT (across the barrier)

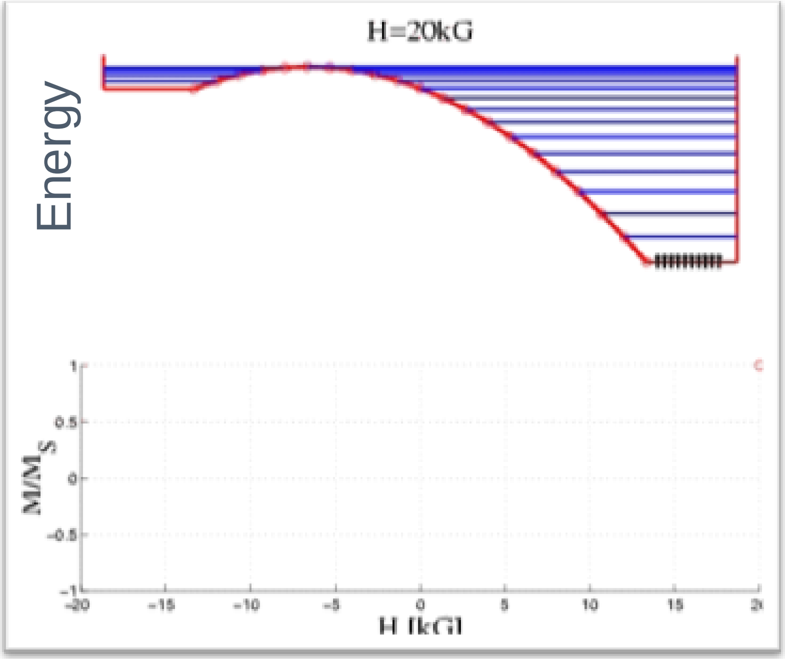


$$\Gamma_q = f(T)$$

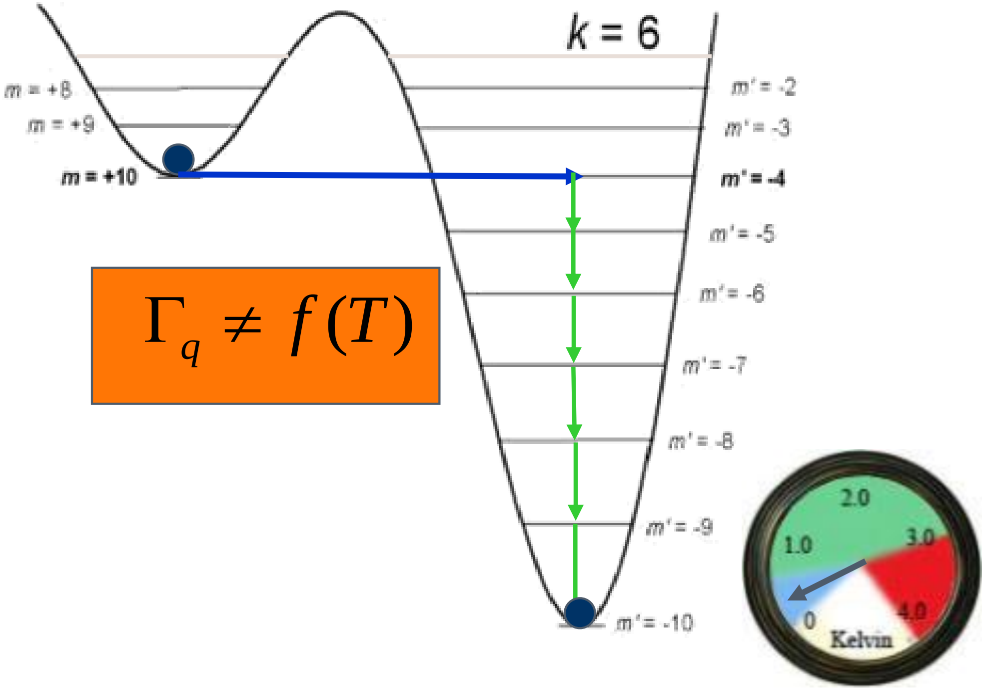
Relaxation processes

Magnetic field relaxation at low temperature for a single molecule

Hysteresis loop of Fe8



Pure Quantum Tunneling (across the barrier)



$$\Gamma_q \neq f(T)$$

Relaxation processes

Crystal of Molecular Magnets

At low-temperature magnetic relaxation of whole crystal can occur via two mechanisms:

A. **Staircase magnetization curve** - Slow mechanism involves quantum transitions at unrelated spatial points.

B. **Magnetic Avalanche** - Abrupt reversal of the magnetization.

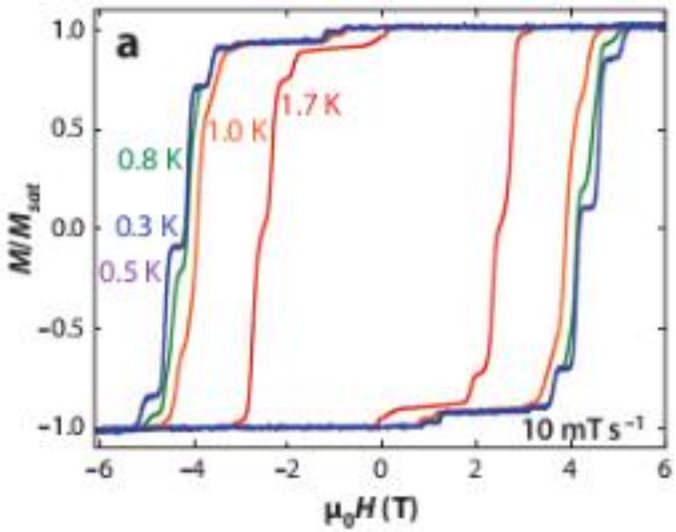


Figure A

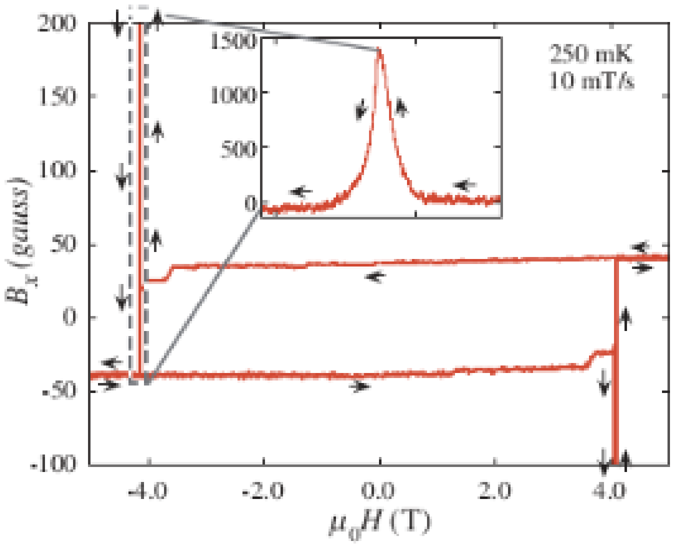
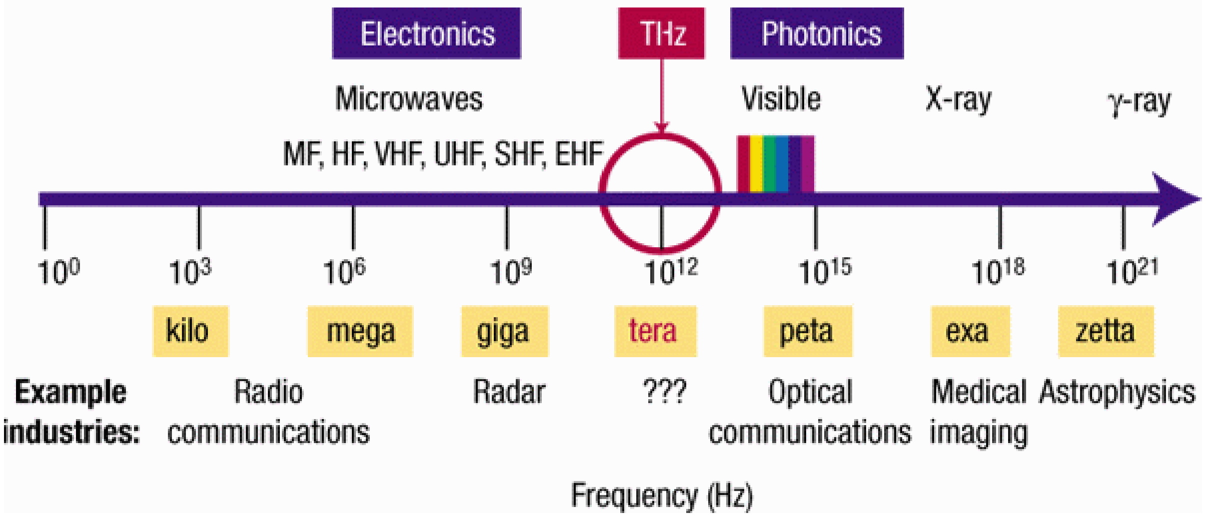


Figure B

Suzuki Y, Sarachik MP, Chudnovsky EM, McHugh S, Gonzalez-Rubio R, et al. 2005. Phys. Rev. Lett. 95:147201

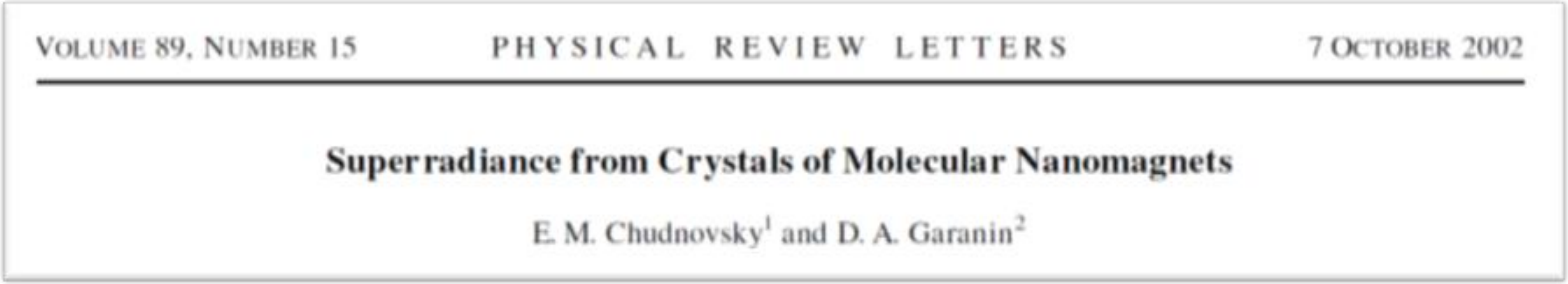
Radiation Spectrum

- The expected energy release after the tunneling is twice $\simeq 5K$ $\simeq 109.5Ghz$ with photon wavelength of about 2.7mm.
  - Cosmic Background radiation is at 160.2 GHz.
  - At frequencies within this range, useful power generation and receiver technologies are inefficient and impractical.



- Our sample size is also about 3x3x1mm which is the elementary condition for **Dicke Superradiance**.

Plan A : Is There Radiation emanating from Fe8 ?



VOLUME 89, NUMBER 15 PHYSICAL REVIEW LETTERS 7 OCTOBER 2002

Superradiance from Crystals of Molecular Nanomagnets

E. M. Chudnovsky[1] and D. A. Garanin[2]

APPLIED PHYSICS LETTERS VOLUME 84, NUMBER 13

Electromagnetic radiation produced by avalanches in the magnetization reversal of Mn_{12}-acetate

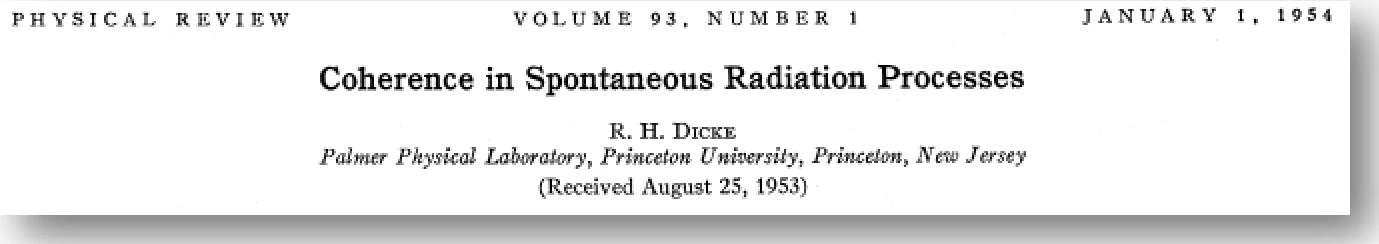
J. Tejada[a)]

Department de Fisica Fonamental, Universitat de Barcelona, Diagonal 647, 08024 Barcelona, Spain

PHYSICAL REVIEW B **79**, 180404(R) (2009)

Electromagnetic radiation emanating from the molecular nanomagnet Fe_8

Oren Shafir and Amit Keren



PHYSICAL REVIEW VOLUME 93, NUMBER 1 JANUARY 1, 1954

Coherence in Spontaneous Radiation Processes

R. H. Dicke
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey

(Received August 25, 1953)

(Enhanced spontaneous emission)

- S&S-radiance arises from spontaneous organization and phase-locking between initially independent systems.

- The effect occurs when the molecules are confined to a volume smaller than radiation wavelength cubed.

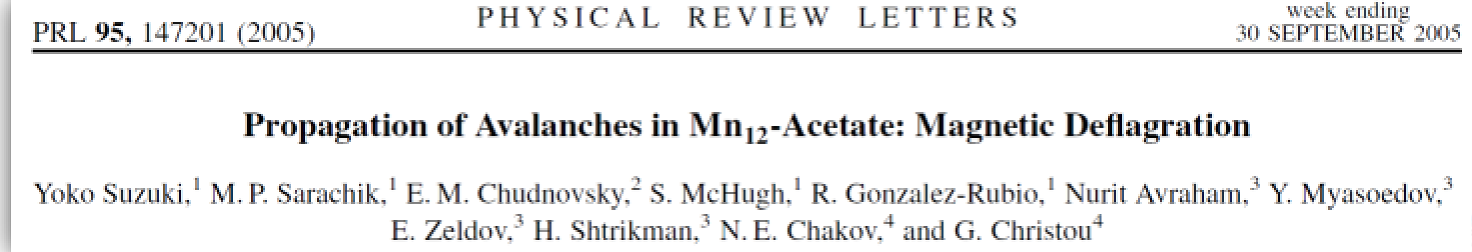
- The spontaneous emission is enhanced when the molecules’ dipole moments are in phase (super-radiance) and inhibited when they are out of phase (sub-radiance).

- Both effect are universal and fundamental, allowing SR phenomena to occur in a variety of physical systems.

$$\Gamma \simeq \frac{N^2}{4}\Gamma_0$$

Plan B: Are there Avalanches in Fe8?

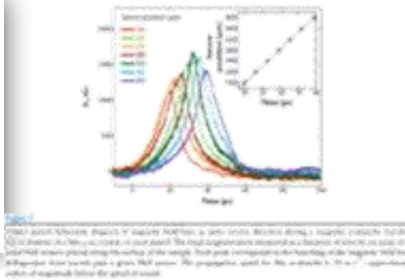
Avalanche in Mn_{12}



PRL **95,** 147201 (2005) PHYSICAL REVIEW LETTERS week ending 30 SEPTEMBER 2005

Propagation of Avalanches in Mn_{12}-Acetate: Magnetic Deflagration

Yoko Suzuki,[1] M. P. Sarachik,[1] E. M. Chudnovsky,[2] S. McHugh,[1] R. Gonzalez-Rubio,[1] Nurit Avraham,[3] Y. Myasoedov,[3] E. Zeldov,[3] H. Shtrikman,[3] N. E. Chakov,[4] and G. Christou[4]

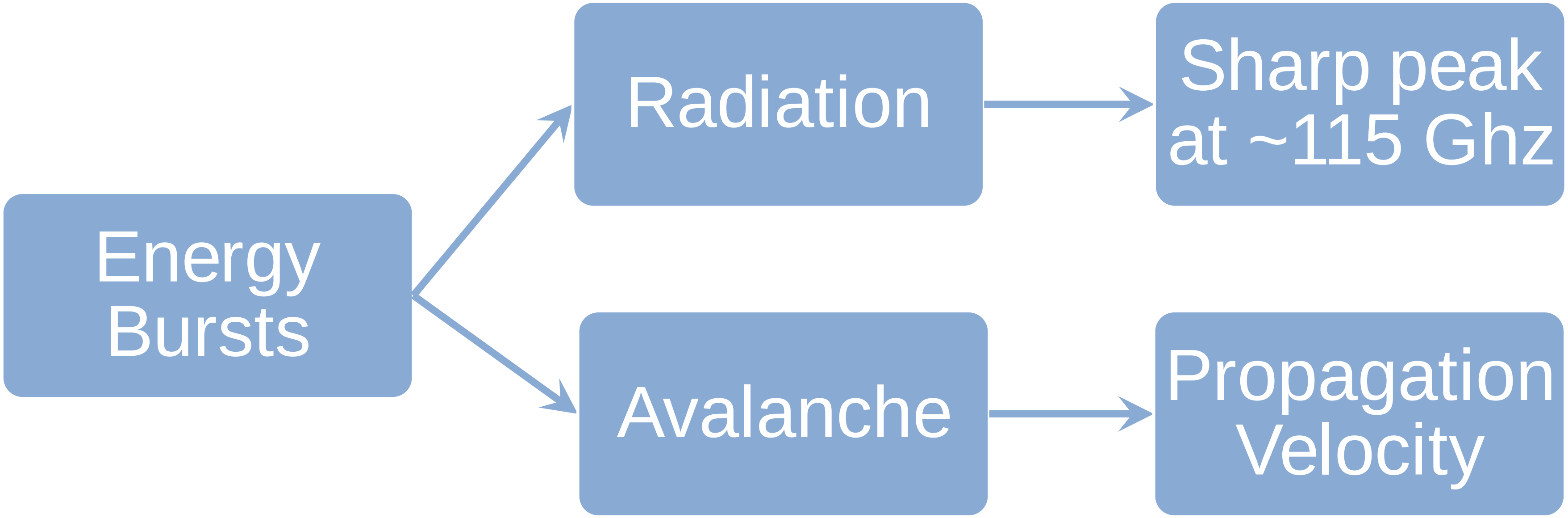


- The Hysteresis loop is temperature depended down to 60 mK
- Avalanches occur in a stochastic way both at resonant magnetic fields (where energy levels on opposite side of the barrier match), and away from resonance.
- The avalanche propagates through the crystal at a constant velocity that is roughly two orders of magnitude smaller than the speed of sound ≈10 (m/s).

Suzuki Y, Sarachik MP, Chudnovsky EM, McHugh S, Gonzalez-Rubio R, et al. 2005. Phys. Rev. Lett. 95:147201

J. A. A. J. Perenboom, J. S. Brooks, S. Hill, T. Hathaway, N. S. Dalal, Phys. Rev. B 58, 330 (1998).

Experimental Motivation



Energy Bursts

Radiation

Sharp peak at ~115 Ghz

Avalanche

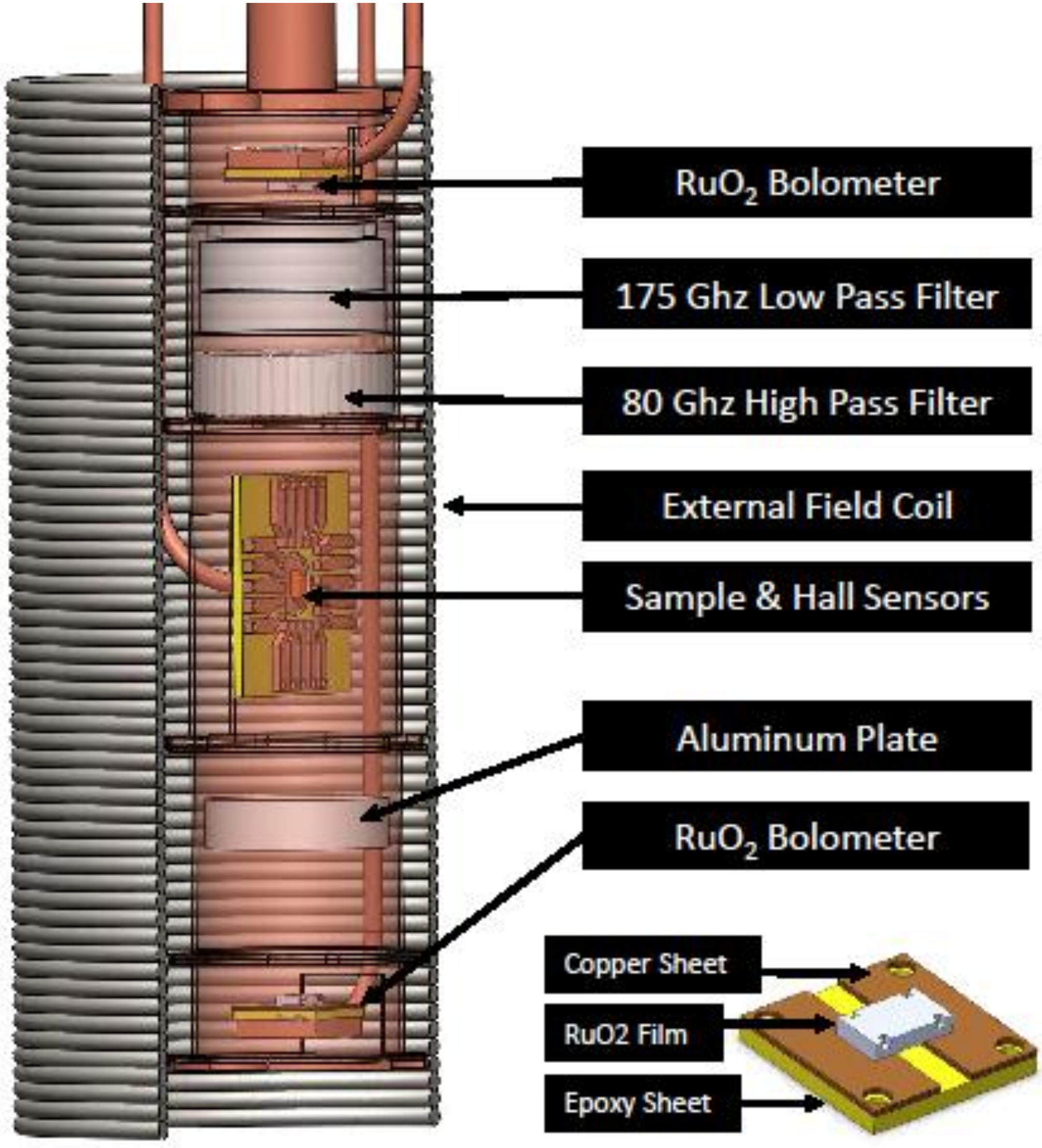
Propagation Velocity

Questions:

1. Is there an EM radiation? What is it frequency?
2. How does the magnetization depends on location on the sample? Is there Avalanche in Fe_8?

Experimental Setup.

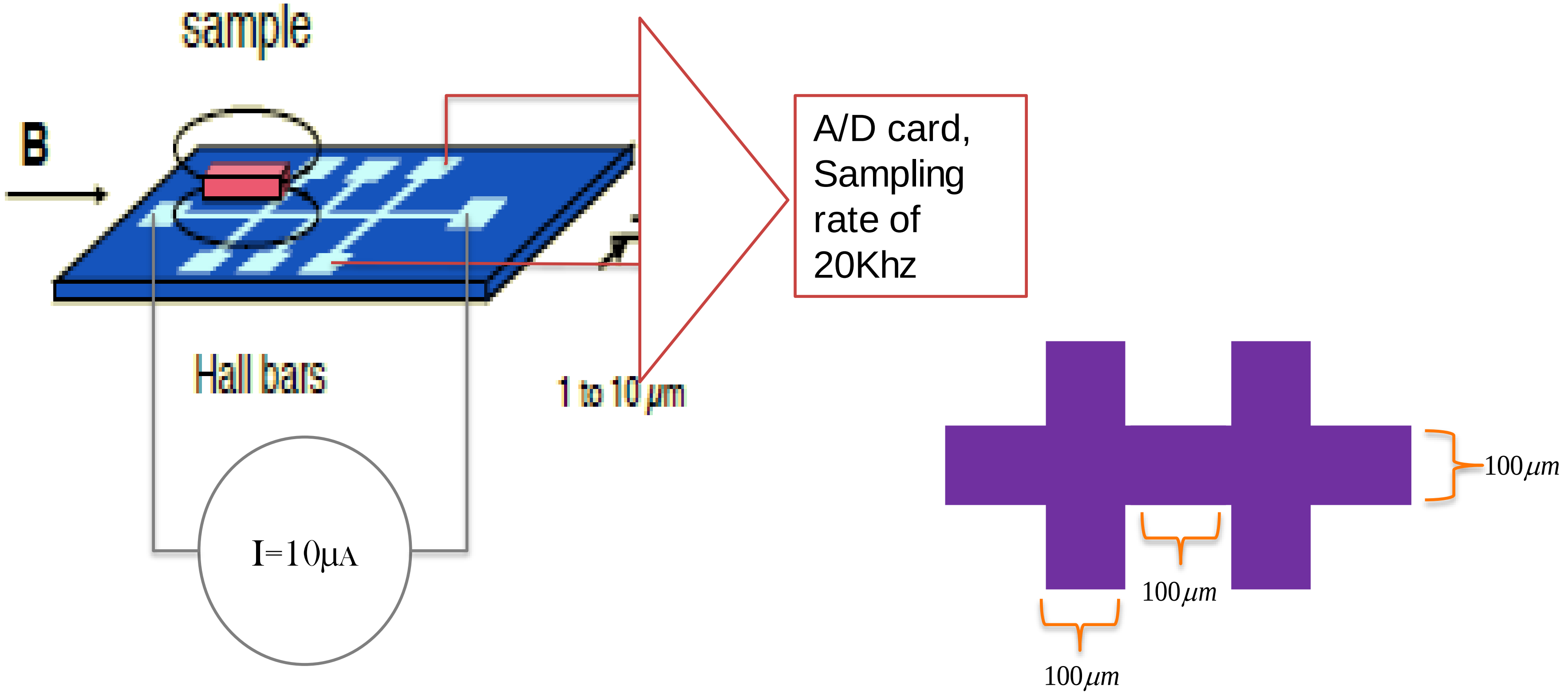
The Experiment conducted in Dilution Refrigerator at 100mK



Experimental Setup.

Hall Sensors - Form Eli Zeldov ,Weitzman institute.

- Two-dimensional electron gas (2DEG) formed at the interface of GaAs/AlGaAs heterostructures.



A/D card,
Sampling
rate of
20Khz

Experimental Setup.

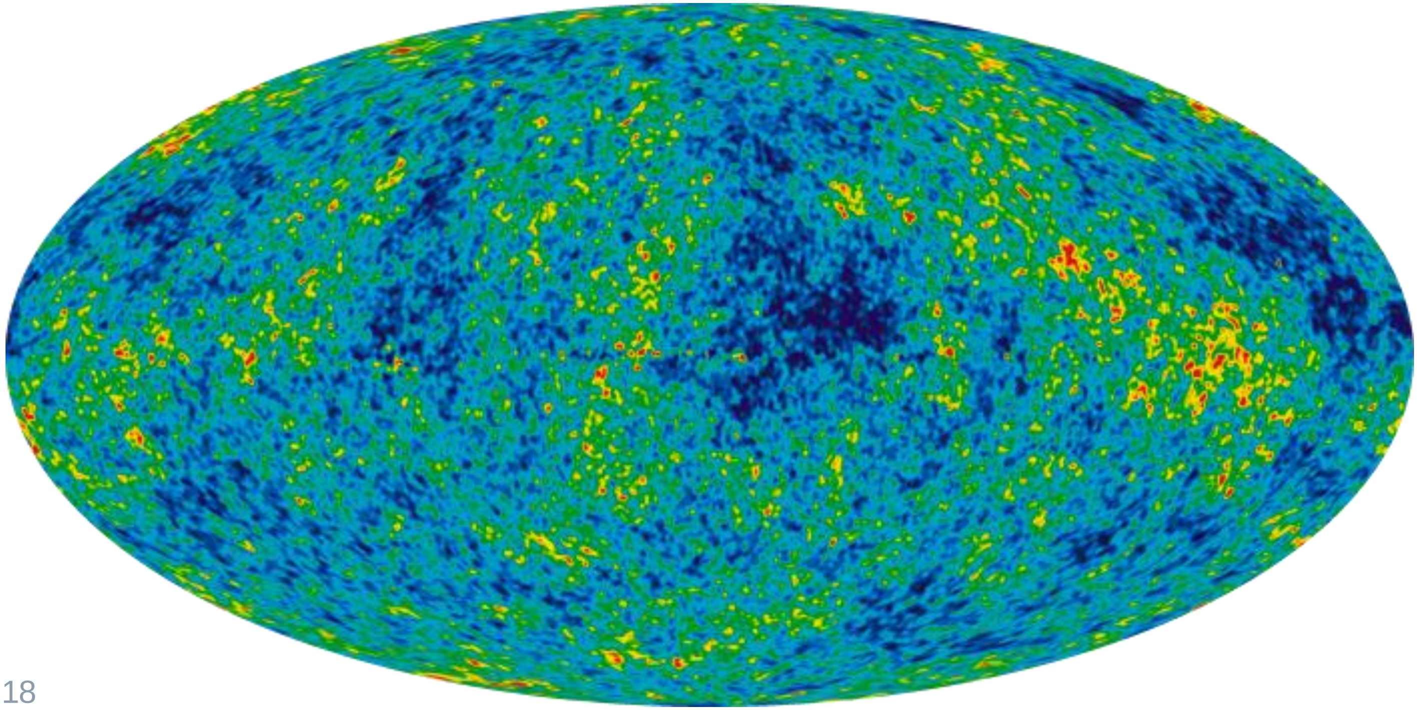
Filters

Used for cosmic background radiation measurement.

From Shaul Hanany – University of Minnesota

MAXIMA

Millimeter Anisotropy eXperiment IMaging Array

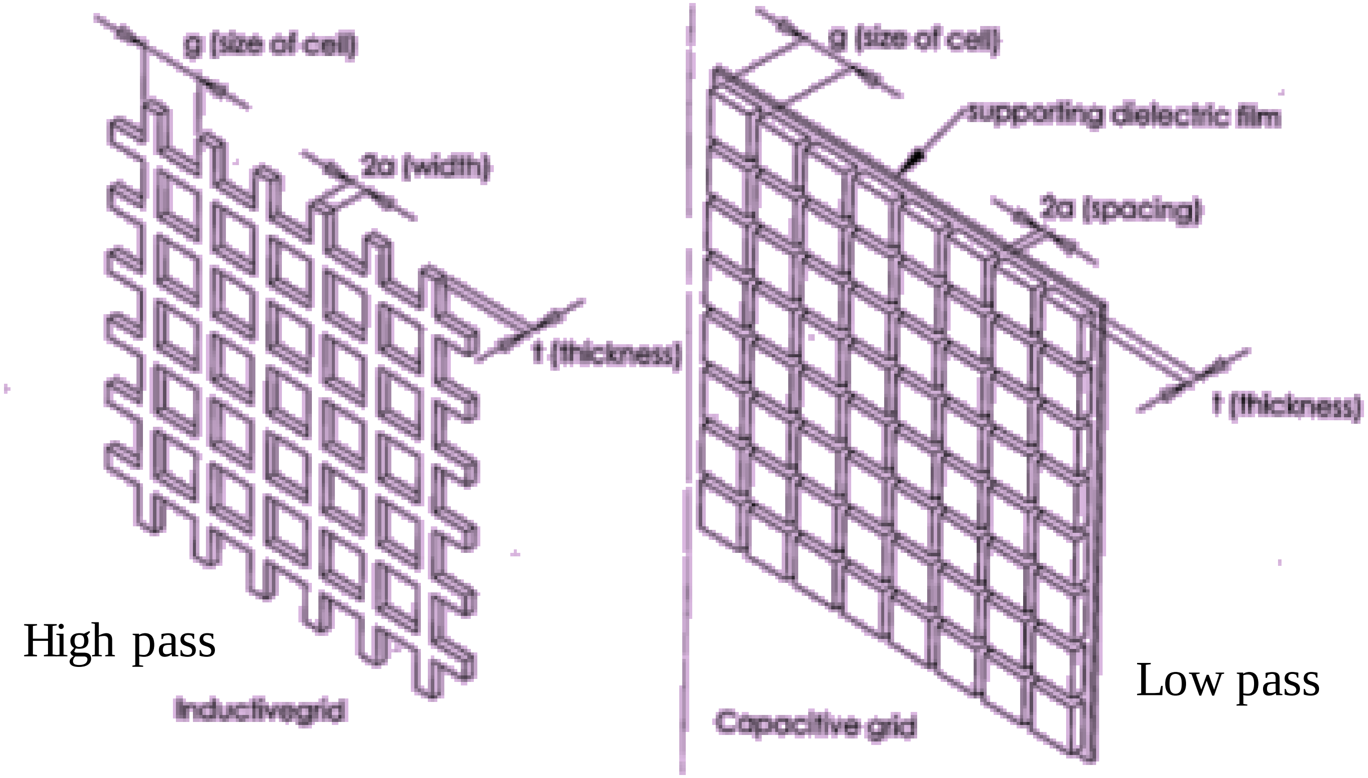


Experimental Setup.

Filters

- In an inductive grid the metal is continuous, so at low frequency it must reflect all the incident wave (high pass).

- Because the grids are complements of each other, a capacitive mesh acts as a low pass filter.



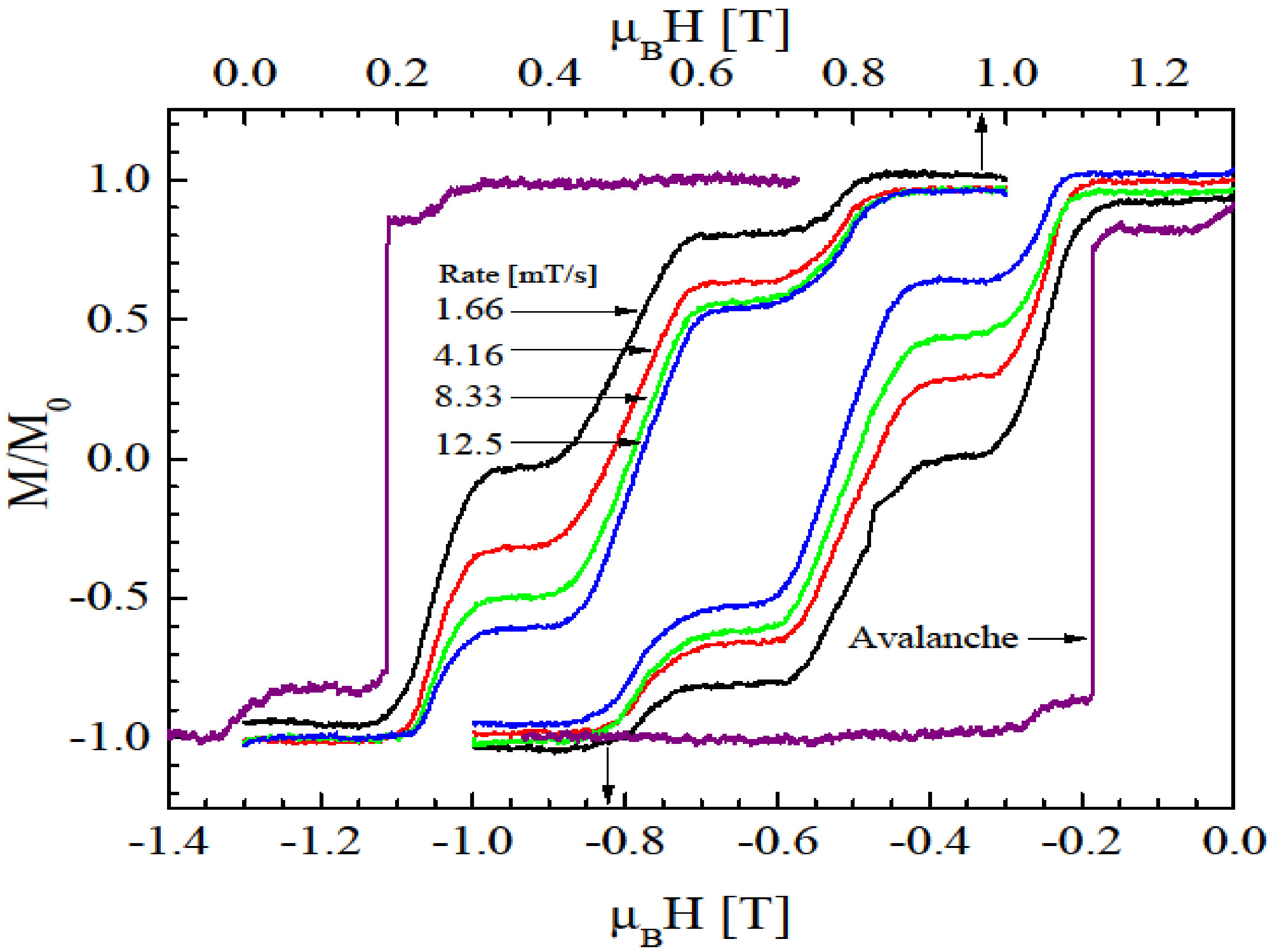
High pass

Low pass

Max Born and Emil Wolf (1999). Principles of Optics 7th Edition

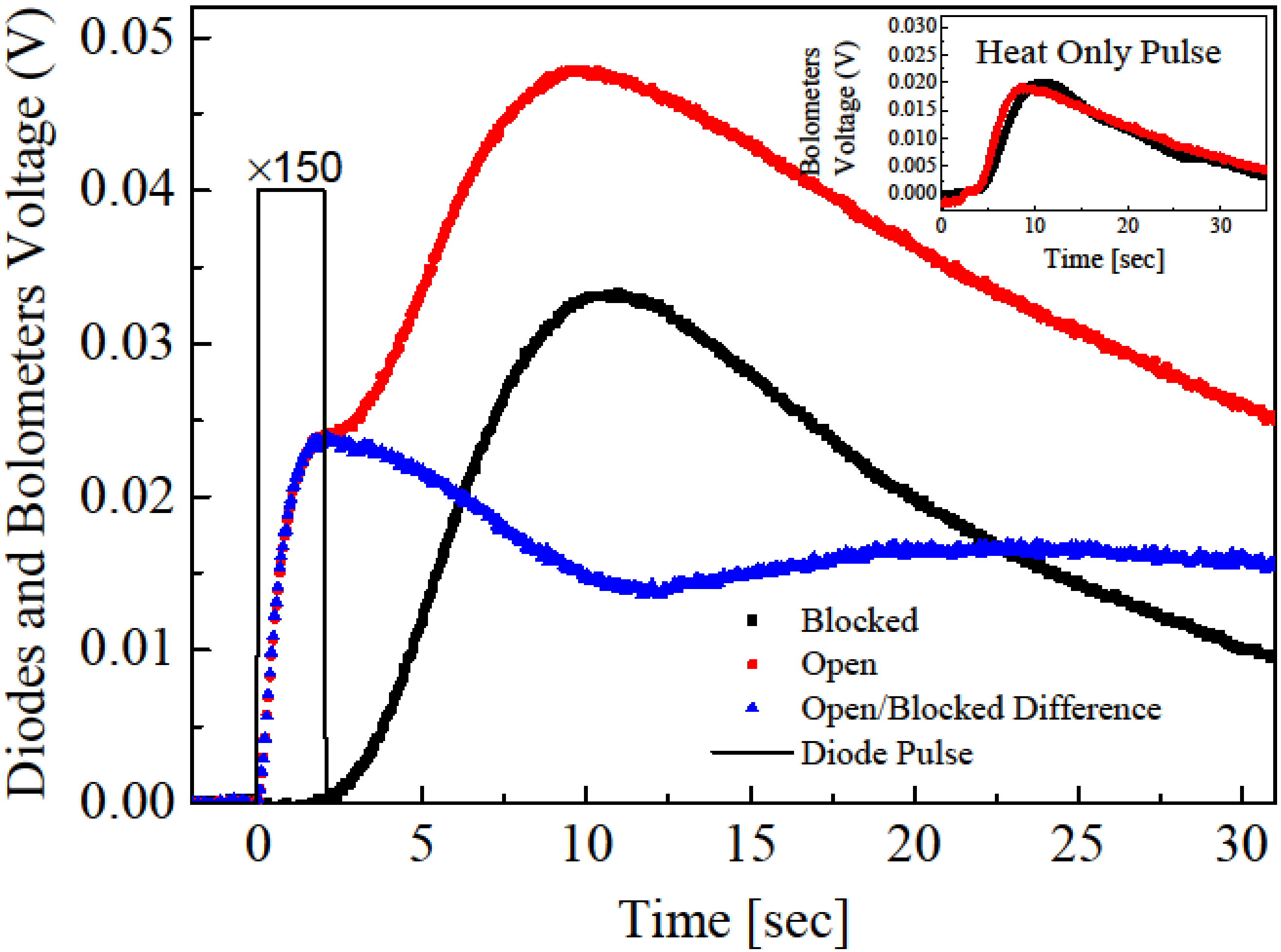
Results – Plan A

Magnetization Measurements



- Two types of relaxation are detected.
- Avalanche is new, Staircase is similar to previous work.
- Magnetic sweep rate dependence is seen.

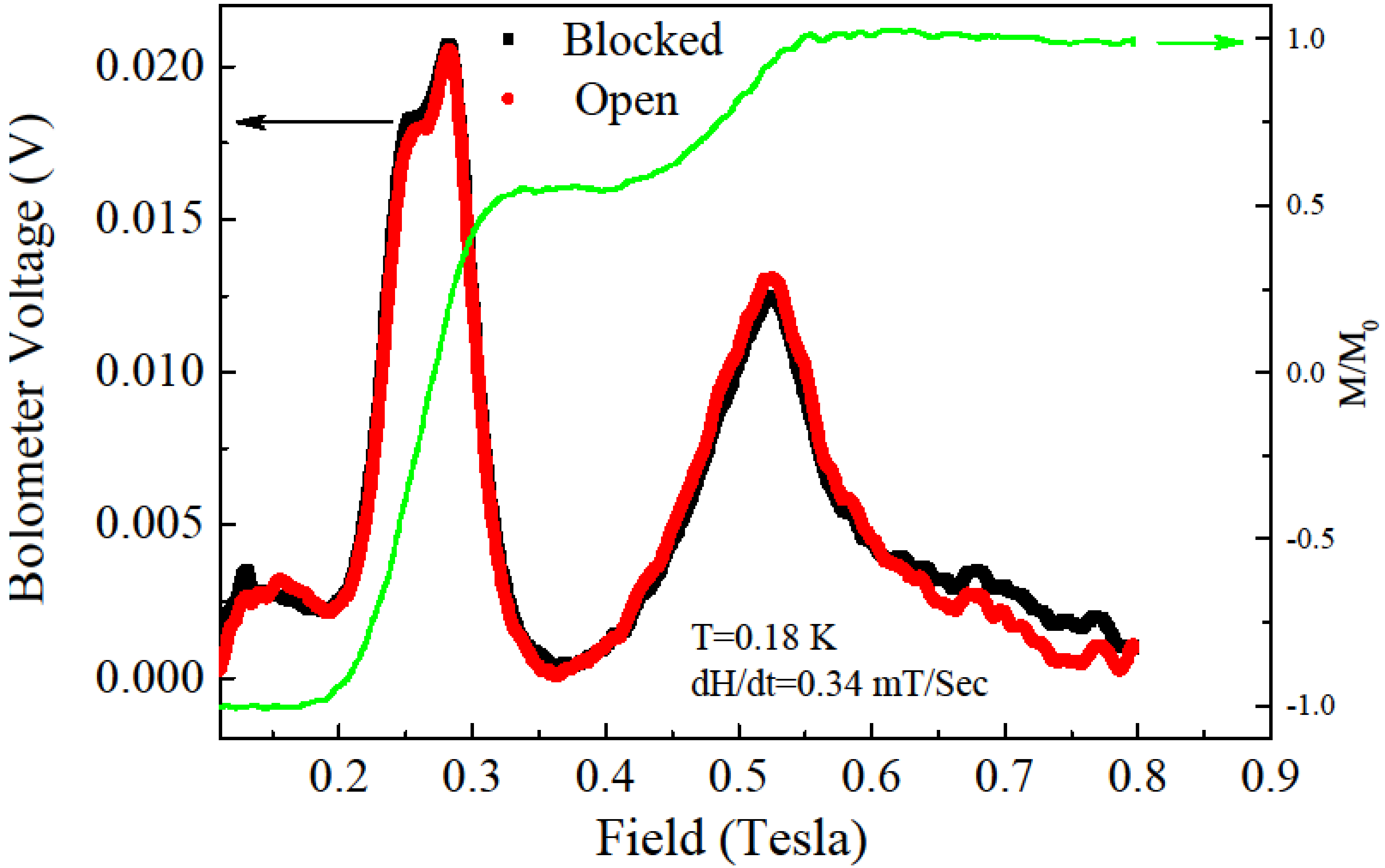
Radiation Test Experiment



Setup was tested by replacing the sample with 2 LEDs and a heater.

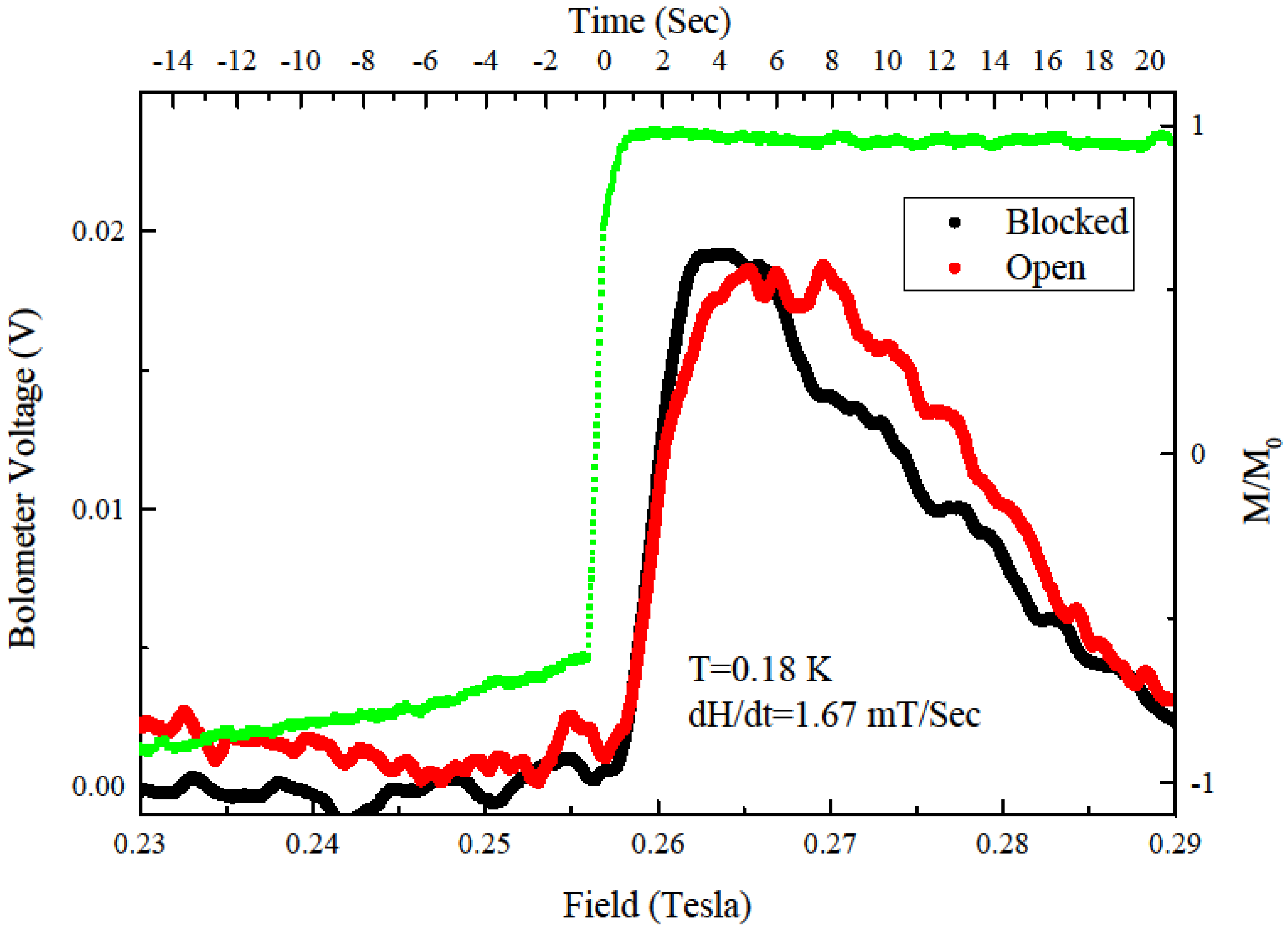
Bolometer which sees the Diode rises earlier and higher.

No Radiation has been Detected
At the staircase regime ☹



- Question: Why the second transition lasts longer than the first after tunneling have finished?

No Radiation has been Detected At avalnche regime ☹



- Question: Where energy in avalanche? Same voltage change, much shorter time.

Plan A conclusions

- ✓ We found no evidence for release of electromagnetic radiation.
- ✓ Why the second transition lasts after the magnetization stops?
- ✓ Where is the energy in avalanche case?

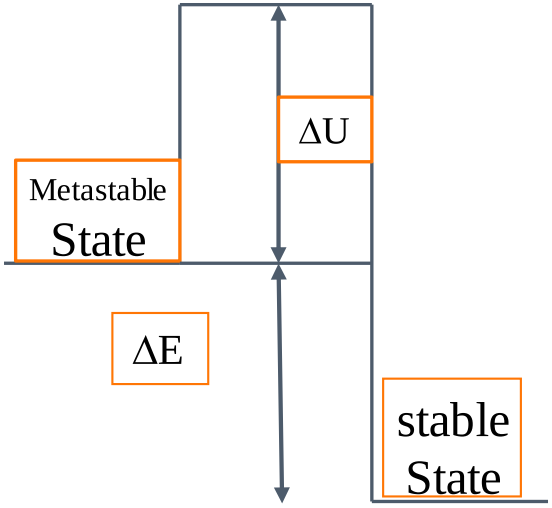
Introduction to Magnetic Deflagration

Magneto: The master of magnetism

What is a magnetic deflagration?

- Subsonic combustion propagating through heat transfer.
- Burning material heats the next layer of cold material and ignites it.
- In magnetic deflagration Zeeman energy playing the role of the chemical energy.
- Magnetic deflagration is nondestructive and reversible.

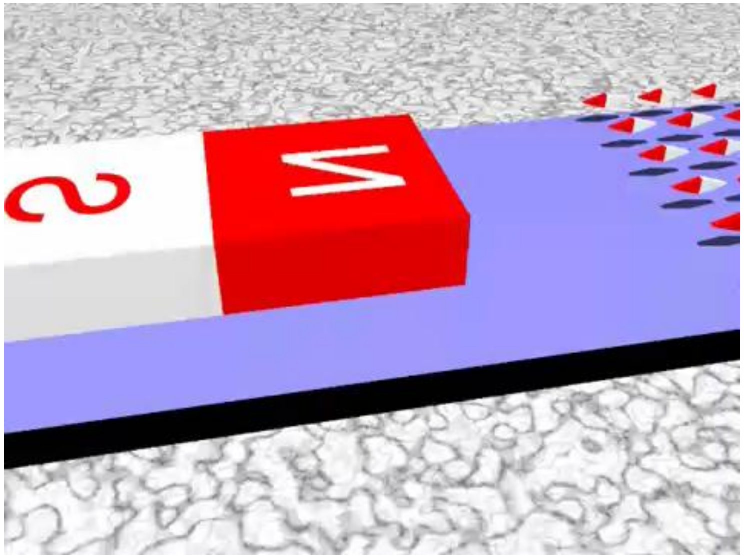
What is a magnetic deflagration?


ΔU

Metastable State

ΔE

stable State



- ➢ When a molecule makes a transition from the metastable state $S_z = \pm 9$ to the absolute energy minimum $S_z = \mp 10$, ΔE energy is released.

- ➢ This leads to local temperature change $\Delta T = \frac{\Delta E}{C_{ph}}$, where is C_{ph} the phonon heat capacity per magnetic molecule.

What is a magnetic deflagration?

- Heat flows according to heat equation:

$$\frac{dT}{dt} = \nabla\kappa(T)\nabla T - \frac{\Delta E}{C_{ph}}\frac{dn}{dt}$$

$$\frac{dn}{dt} = -\Gamma(n - n^{eq})$$

- There are two possible scenarios:

  A. The heat defuses away – **No Ignition**. (Slowly moving match.)

  B. The heat cumulates, this rises the transition rates and causes **Deflagration**. (Match moves fast, heat creation is larger)

What is a magnetic deflagration?

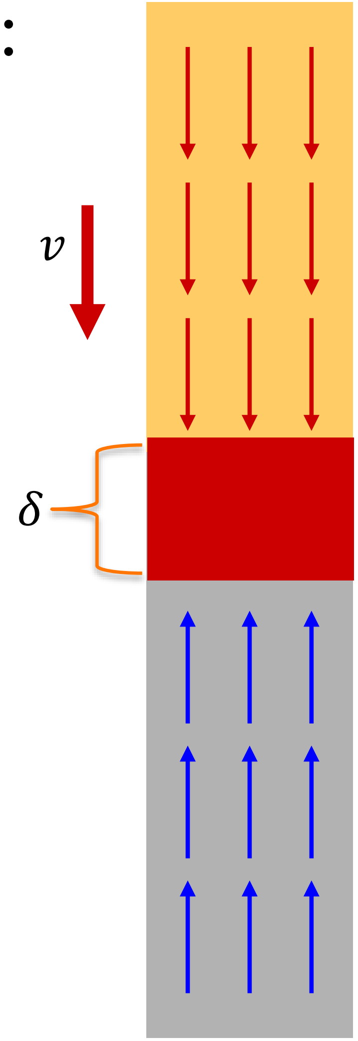
When the transition rate of a single molecule $\Gamma(H_0, T_0)$

(Heat creation) exceeds a critical value:

$$\Gamma_c = \frac{8\kappa(T_0)k_B T_0^2}{U(H_0)\Delta E(H_0) n_i l^2}$$

Notice no sweep rate dependence

- $U(H_0)$ is the energy barrier.
- $\Delta E(H_0)$ it the energy released decaying to the ground state.
- n_i the initial population of the exited state.
- l caratiristic length of the sample.
- T_0 the ignition temperature.

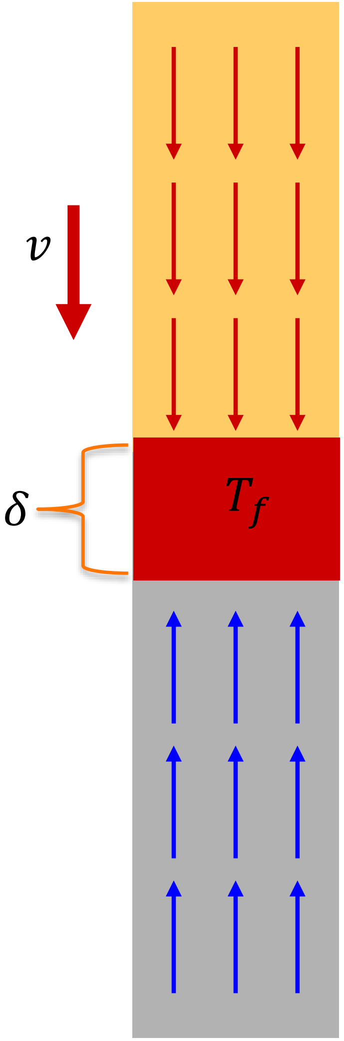


D. A. Garanin and E.M. Chudnovsky, Phys. Rev. B76, 054410 (2007)

What is a magnetic deflagration?

There are two characteristic timescales:

1. Thermal diffusion timescale: $\tau_d \simeq \frac{\delta^2}{\kappa}$
2. Burning timescale which is the chemical reaction timescale given by: $\tau_b = \tau_0 \exp\left(\frac{U(H)}{k_B T_f}\right)$



The width of the front δ, can be approximated by equalization the two timescales $\tau_d \simeq \tau_b$ leading to:

$$\delta = \sqrt{\kappa\tau_b}$$

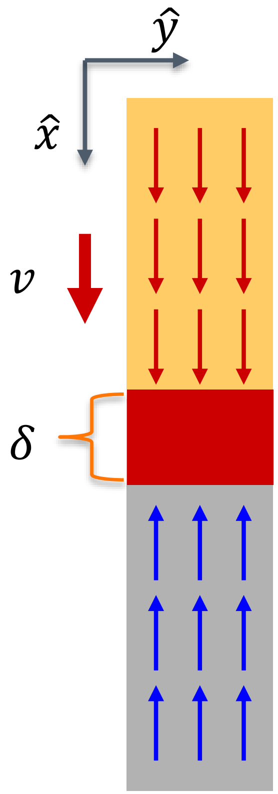
The front propagates at a characteristic speed which equals to the width over the burning rate:

$$v \simeq \sqrt{\frac{\kappa}{\tau_b}} = \sqrt{\frac{\kappa}{\tau_0}} \exp\left(-\frac{U(H)}{2k_B T_f}\right)$$

D. A. Garanin and E.M. Chudnovsky, Phys. Rev. B76, 054410 (2007)

Hall Signal Calculation

Can we calculate the deflagration front width from our data?



Magnetic diploe: $\vec{B} = \frac{\mu_0}{4\pi}\left(\frac{3\vec{r}(\vec{\mu}\cdot\vec{r})}{r^5} - \frac{\vec{\mu}}{r^3}\right)$

Assuming the sample is on XY plane with the magnetization

pointing in $\hat{x}$ direction: $\vec{\mu} = m\, dx\, dydz\, \hat{x}$,m is dipole density.

$$-L < x < x'(t)$$

$$-w < y < w$$

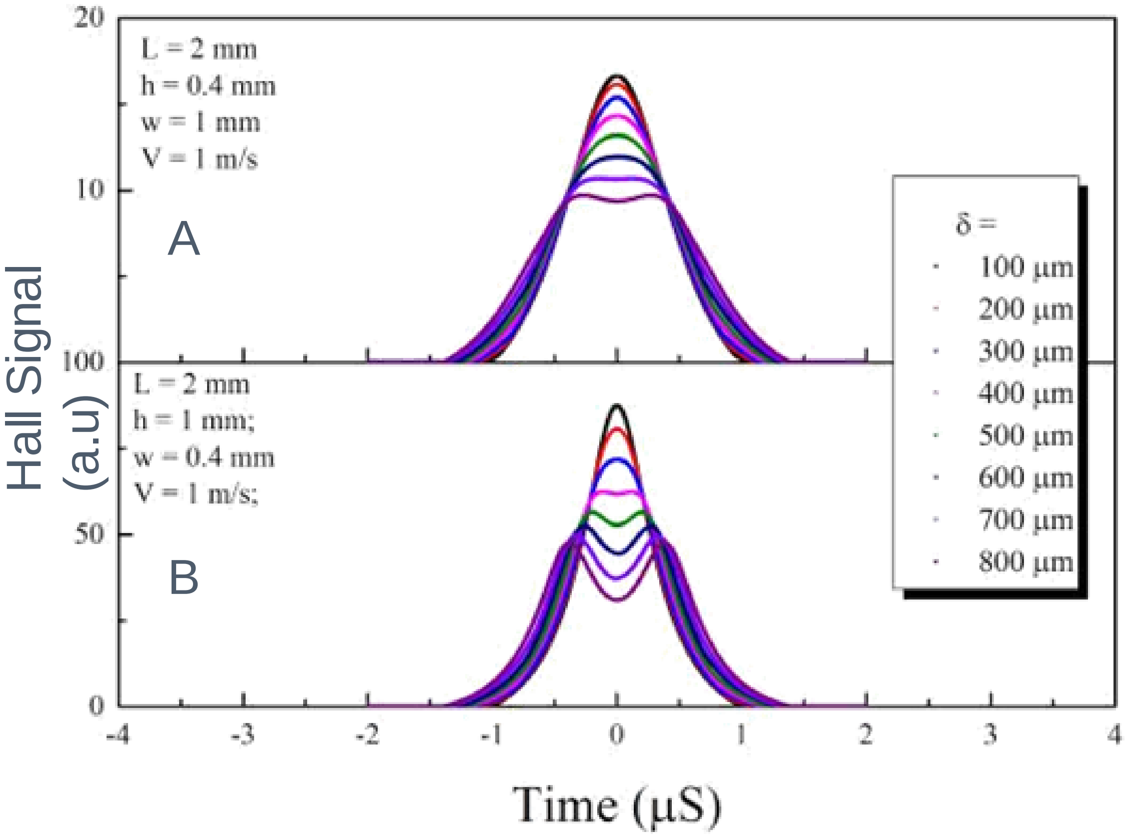
$$d_1 < z < d_2$$

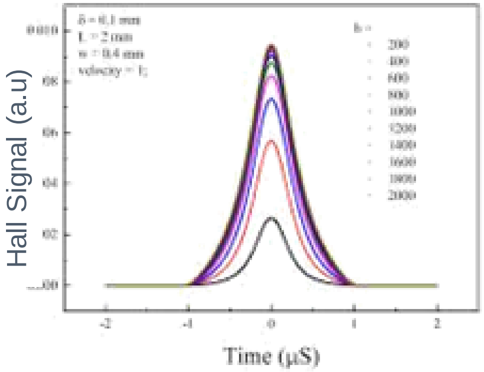
The Field in z direction can be calculated analytically:

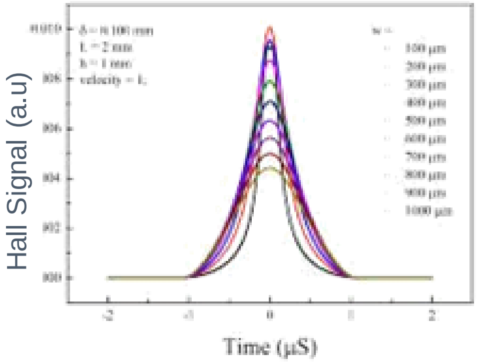
$$B_z = \left[Arc\tan\left(\frac{\sqrt{w^2 + x^2 + d_2^2}}{w}\right) - Arc\tan\left(\frac{\sqrt{w^2 + x^2 + d_1^2}}{w}\right)\right]_{x=-L}^{x=x'(t)}$$

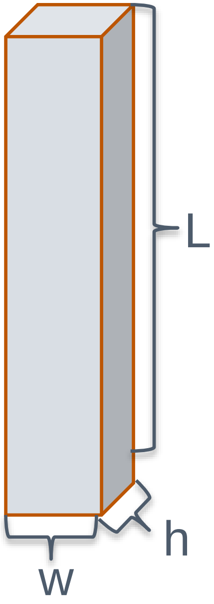
Hall Signal Calculation

Integration over the Hall Sensor for parameters similar to the experiment:









Things that can split the peak:

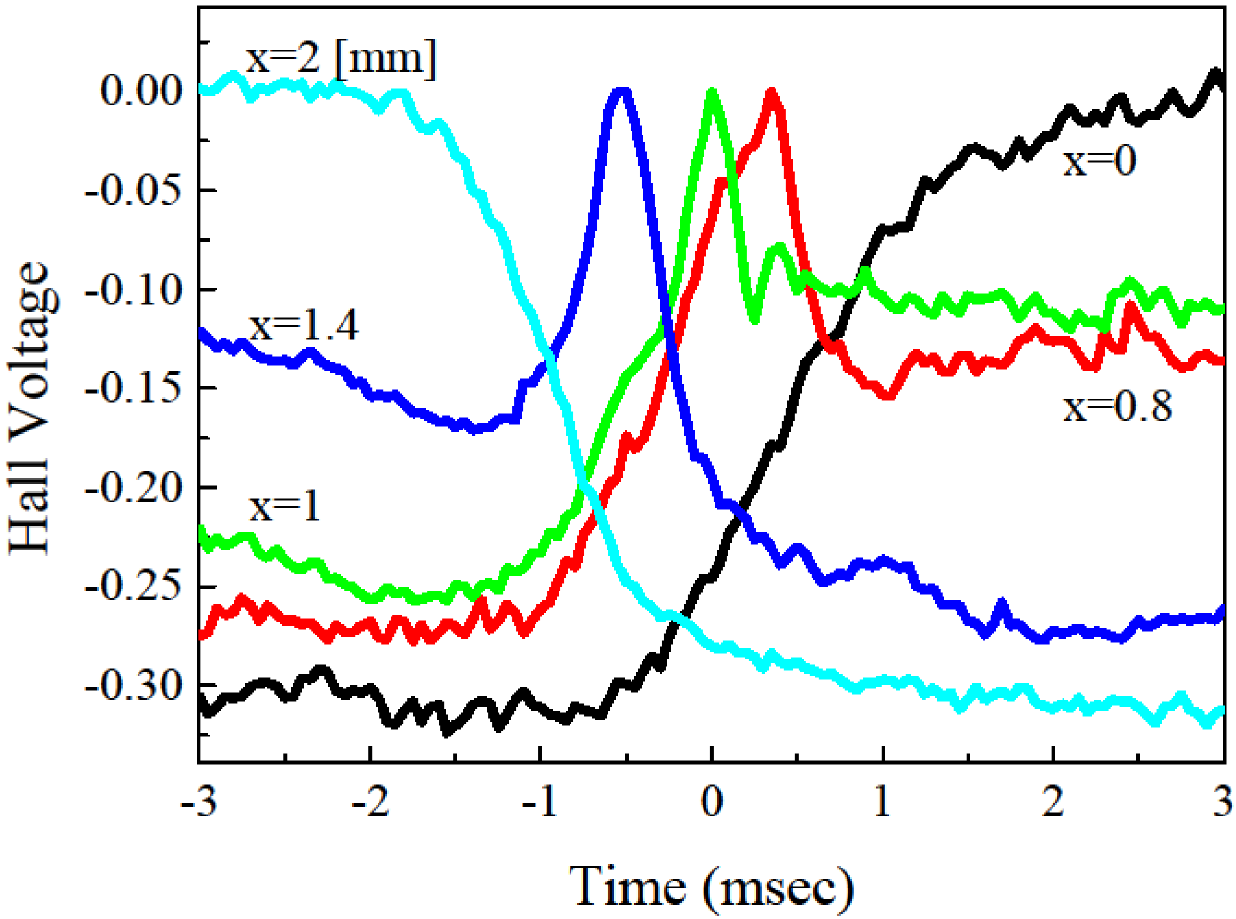
A. Deflagration front bigger than ~700µm.

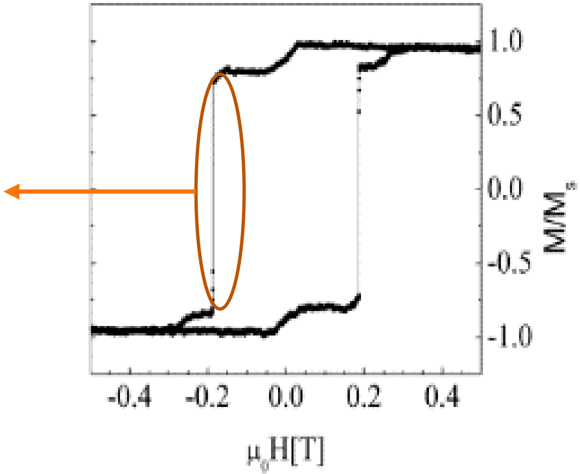
B. The deflagration runs through a strip smaller than the sample.

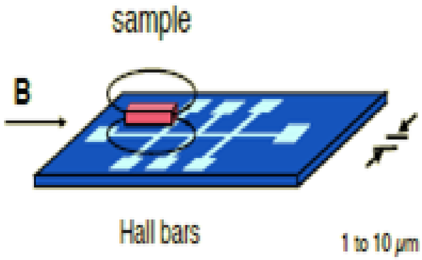
Results – Plan B

Results with Deflagration

Above the critical sweep rate:





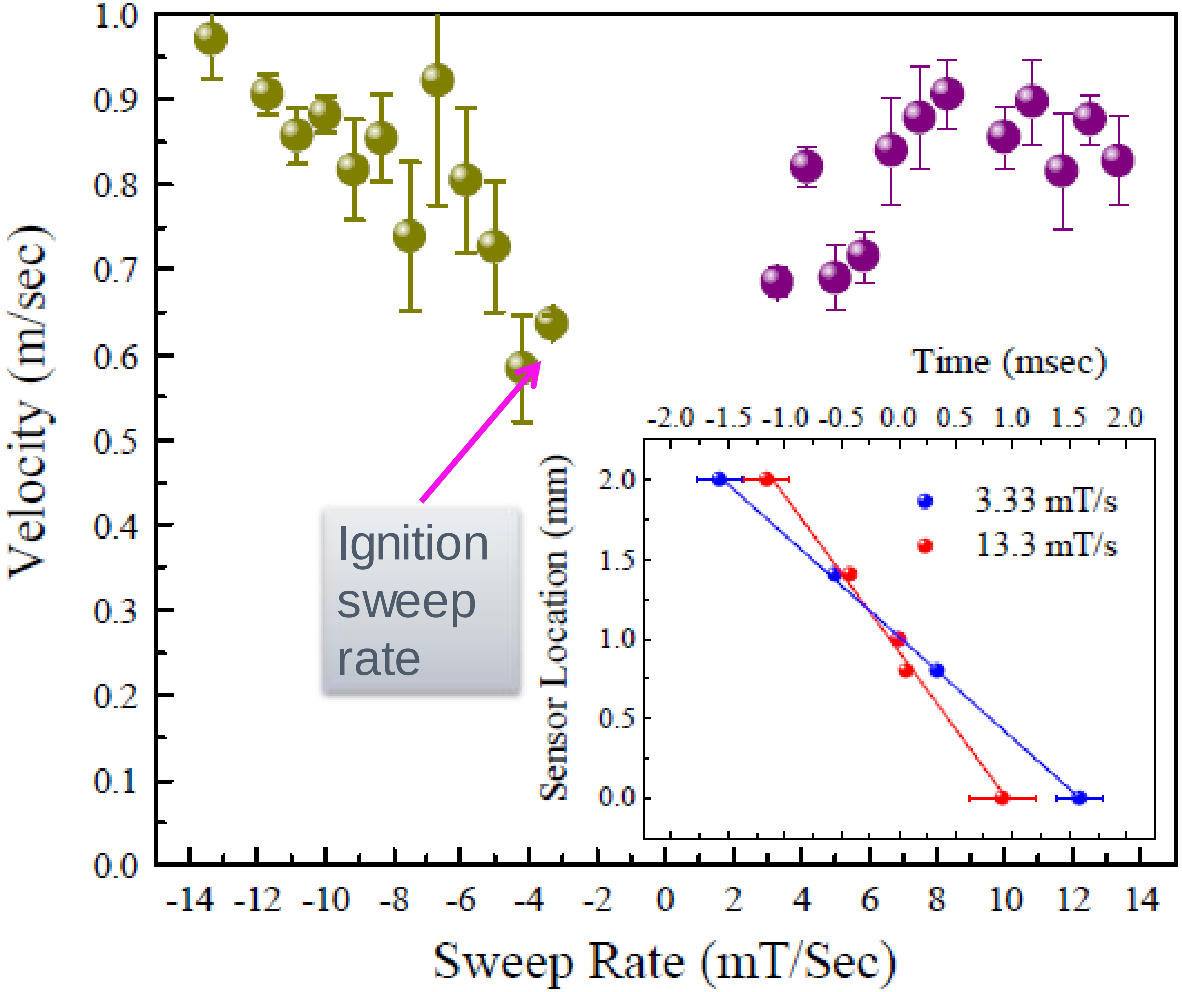


- The Deflagration ignites each time at first resonant field.
- The evolution of the peaks and cusps provide the avalanche propagation velocity.

Results

Deflagration velocity as a function of magnetic field sweep rate

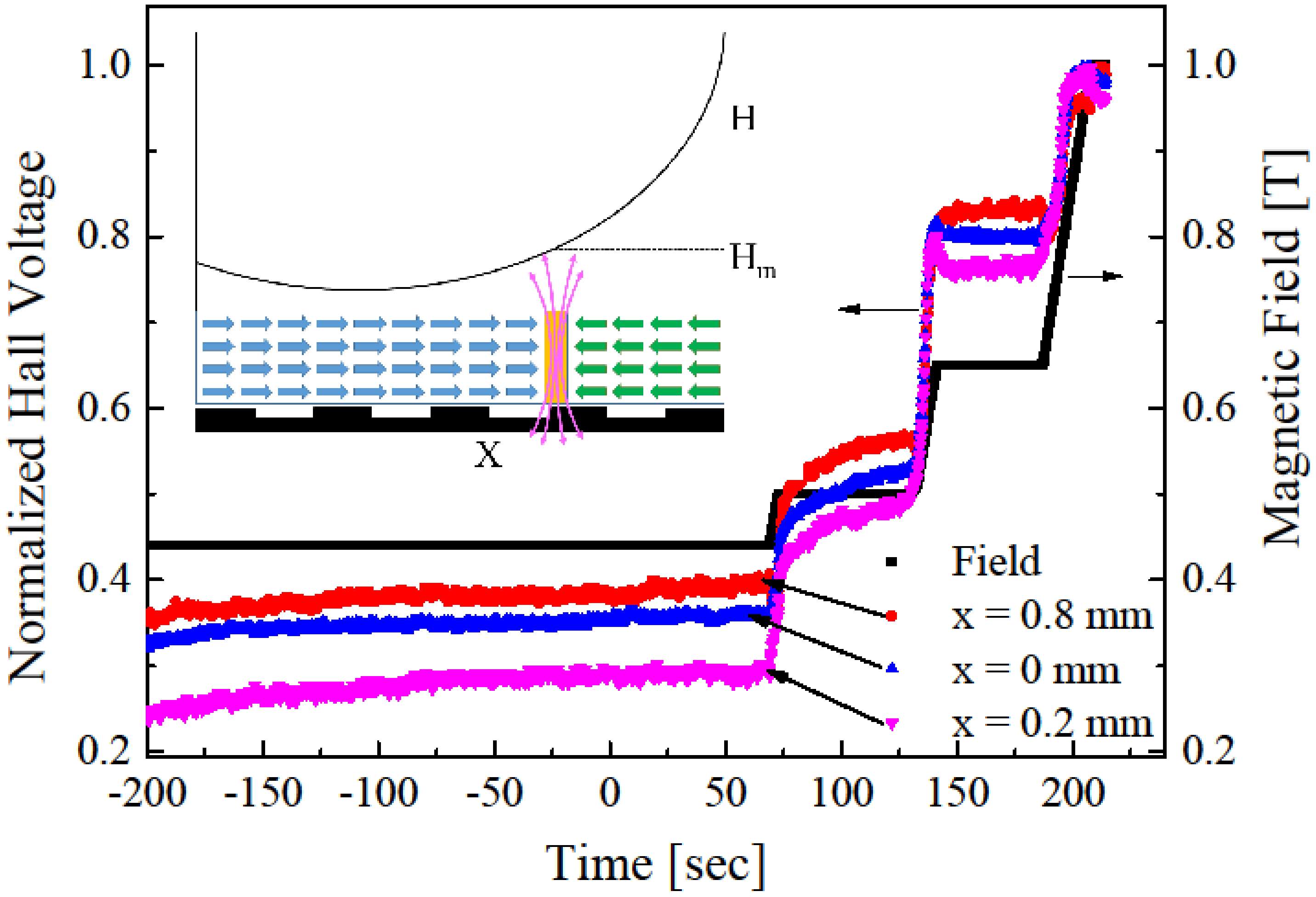
No theoretical treatment



v = 0.5 - 1 m/sec

Avalanche velocity as a function of magnetic field sweep rate at zero gradient. The field is swept from positive to negative and vice versa.

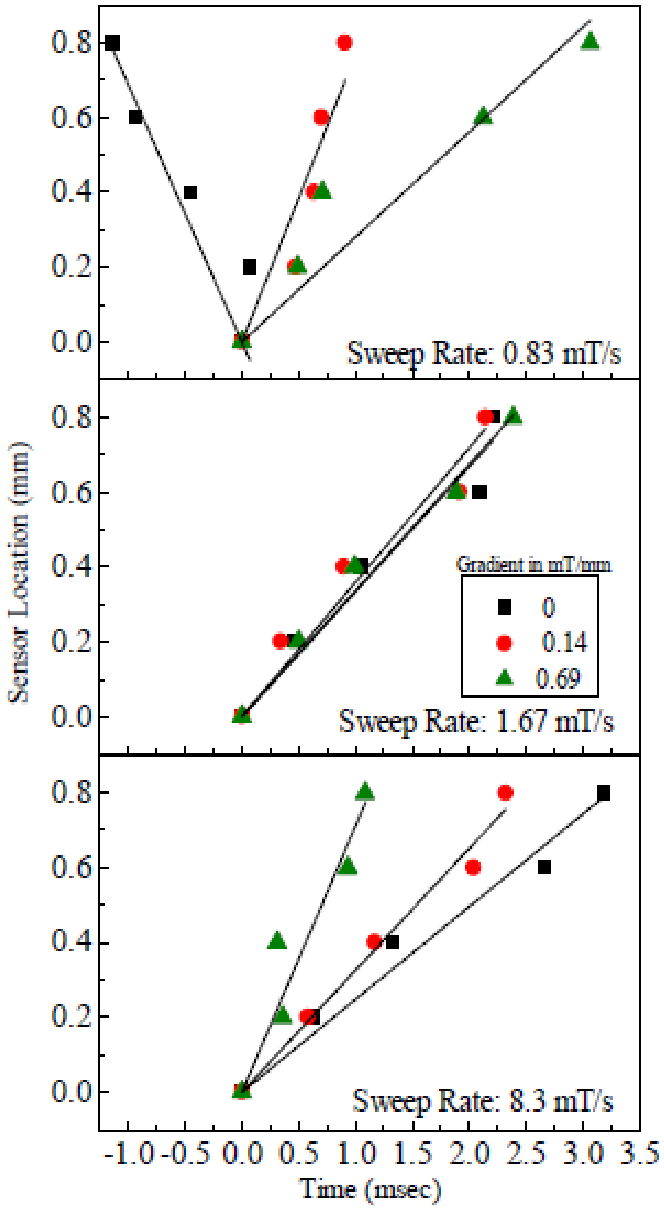
Result without Deflagration



- If the symmetry point, we might have a front of resonant area.
- In this case, pausing the field sweep will stop the magnetization evolution.
- For a typical sweep rate this speed is $V_m \sim 1.5 \times 10^{-4}$

Results

Different Gradients – No theoretical treatment



- The velocity not caused by inhomogeneity of the field.
- The velocity changes its size and direction as a function field gradient.
- Field Gradient can change the ignition point, the critical values and the propagation velocity, by changing the amount of molecules at resonance.

Deflagration Velocity Calculation

The velocity is given by: $$v = \sqrt{\frac{\kappa}{\tau_0}} \exp\left(-\frac{U(H)}{2k_B T_f}\right)$$

Known:

$$\tau_0 = 3.4 \cdot 10^{-8} sec$$

$$U(H) = 24.5 \text{ Kelvin}$$

$$v = 0.5 \text{ - } 1 \text{ m/sec}$$

Not Know (yet):

$$\kappa = ?$$

$$T_f = ?$$

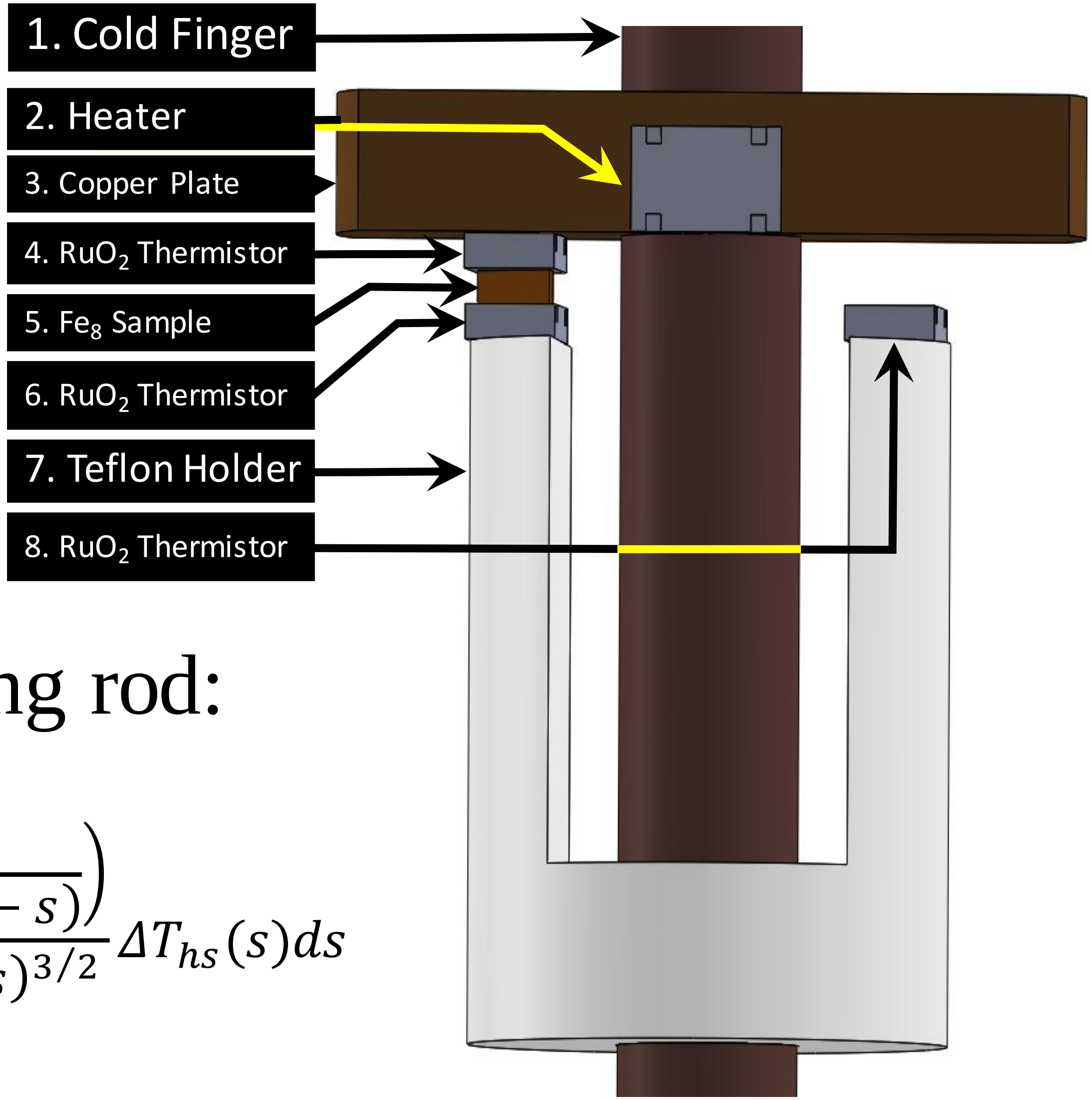
C. Sangregorio, T. Ohm, C. Paulsen, R. Sessoli, and D. Gatteschi, Phys. Rev. Lett. 78, 4645 –1997

Thermal Diffusivity Measurements

Experimental Setup

Thermal diffusivity is defined via the heat equation:

$$\frac{\partial T}{\partial t} = k\nabla^2 T$$

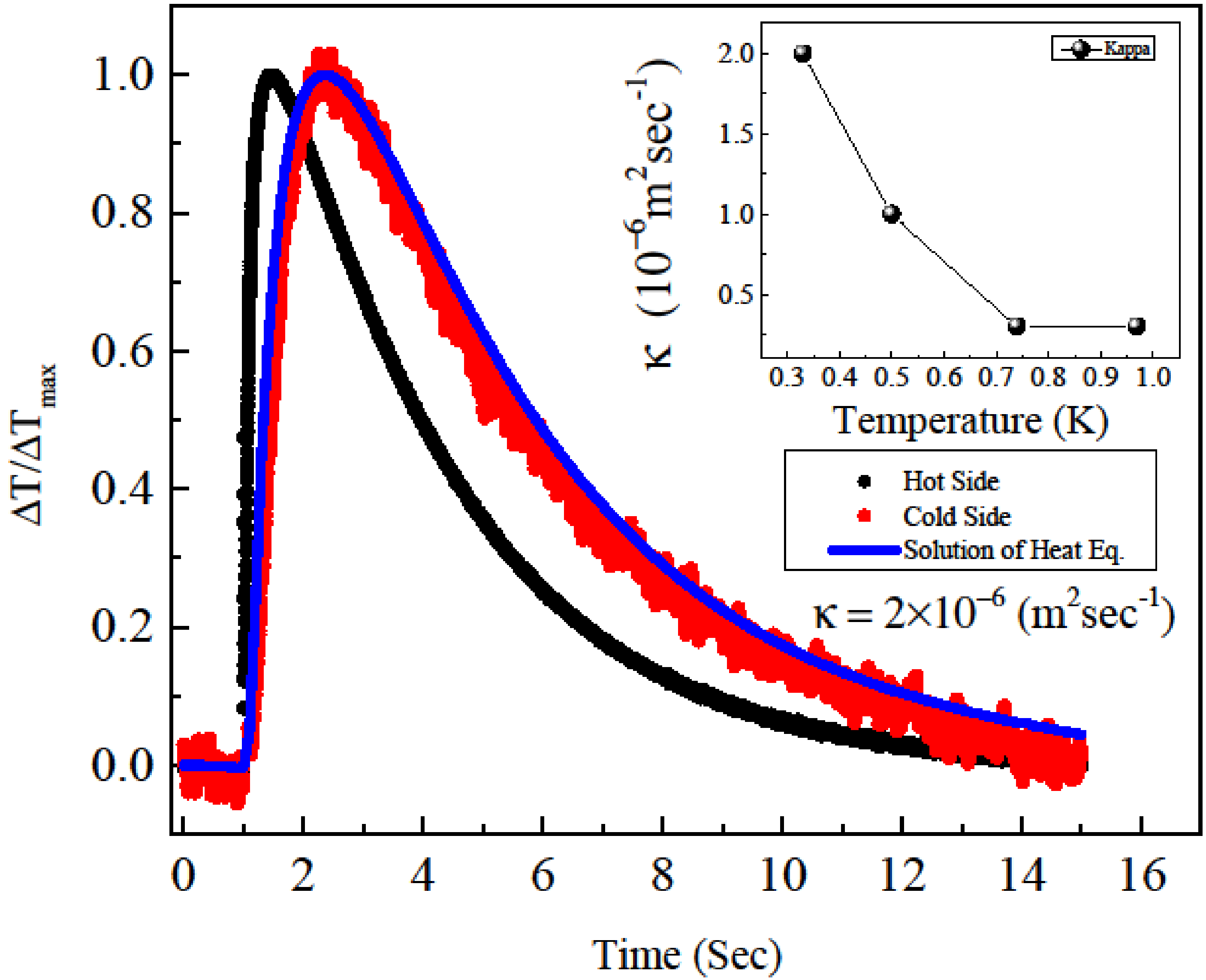


1. Cold Finger
2. Heater
3. Copper Plate
4. RuO$_2$ Thermistor
5. Fe$_8$ Sample
6. RuO$_2$ Thermistor
7. Teflon Holder
8. RuO$_2$ Thermistor

Solution for 1D long rod:

$$\Delta T_{cs}(t) = c\int_0^t \frac{x\exp\left(-\frac{x^2}{4\kappa(t-s)}\right)}{(4\pi\kappa)^{1/2}(t-s)^{3/2}}\Delta T_{hs}(s)ds$$

Thermal diffusivity measurements



Calculation of Flame Temperature

$$T_f = \frac{U}{k_B \ln\left(\frac{\kappa}{v^2 \tau_0}\right)} = \frac{24.5}{\ln\left(\frac{2 \cdot 10^{-6}}{0.6^2 \cdot 3.4 \cdot 10^{-8}}\right)} = 4.8K$$

- ✓ Seems a bit high for a sample at 180-300 mK.
- ✓ But still is lower than the barrier.

Measuring T_f will be Maayan’s Project

Results Summary

- ✓ We found no evidence for release of electromagnetic radiation.

- ✓ We found deflagration in Fe_8 crystals.

- ✓ The deflagration front velocity is of the order of 1 m/sec and is sensitive to field gradients and sweep rates.

- ✓ We also measured the thermal diffusivity of Fe_8 and predicted The flame temperature.

Open Questions

- The heat released during avalanche is smaller than during staircase magnetization change, while it should be otherwise. Where did this energy go?
- Why is there critical sweep rate for deflagration?
- Why deflagration Velocity increases with sweep-rate?
- What is the field gradient dependence?
- Estimated flame temperature is around 5K which is pretty high and still lower that the barrier.

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