Joachim Hemberger

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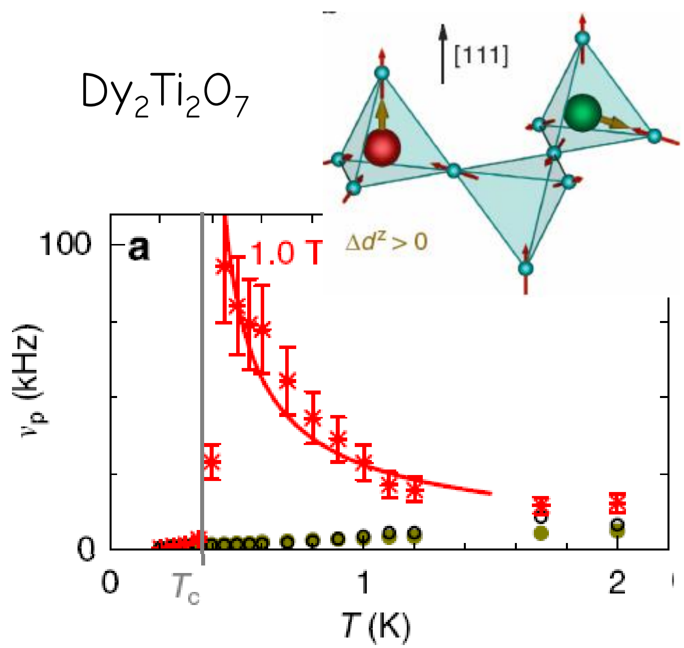
Critical speeding-up in magneto-electric spin-ice

$Dy_2Ti_2O_7$



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Critical speeding-up in magneto-electric spin-ice

- Introduction – Critical Dynamics of Complex Matter
  (e.g. slowing down of magnetoelectric fluctuations in $MnWO_4$)
- Spin-Ice $Dy_2Ti_2O_7$

$\Rightarrow$ magneto-electric response of magnetoelectric monopoles

$\Rightarrow$ Dynamics near critical end-point

Motivation from "real life":

If it's getting critical,

slow down,

or

speed up !

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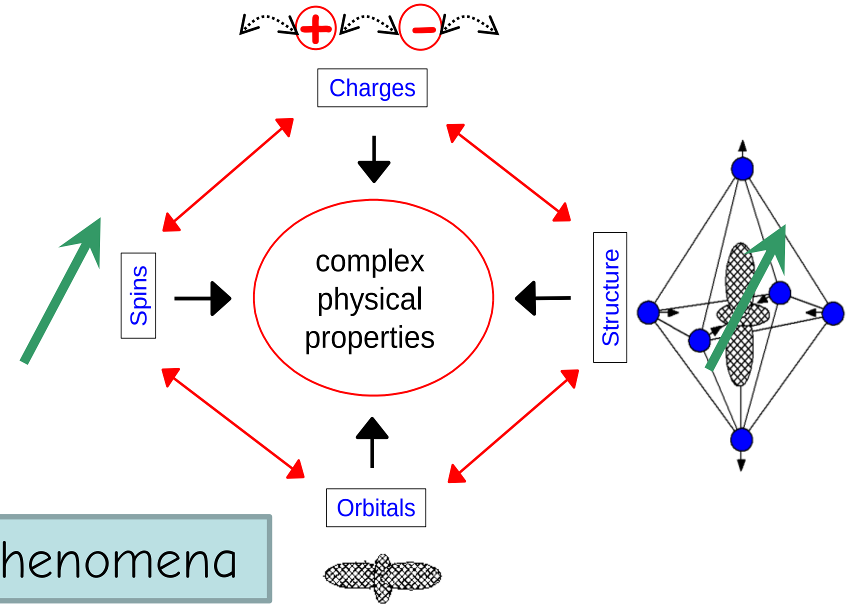
Physical motivation/outline

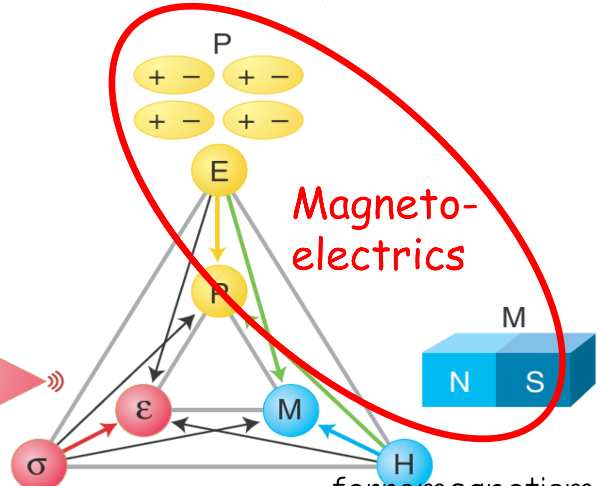
Competing interaction
of correlated
microscopic degrees of freedom



Complex (Quantum) Materials

Complex order phenomena

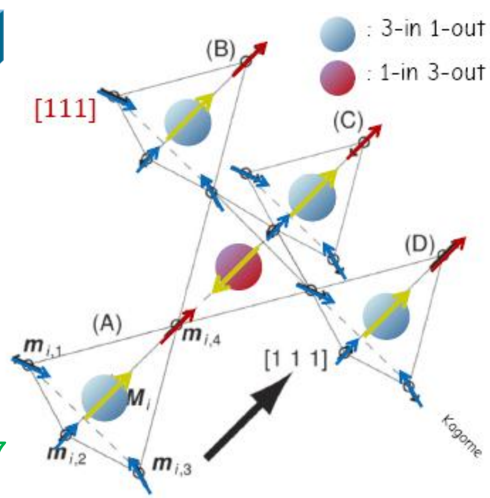
ferroelectricity



ferromagnetism

[Spaldin&Fiebig, Science 15, 5733 (2005)]

e.g.
$Dy_2Ti_2O_7$



(Critical) Dynamics ?

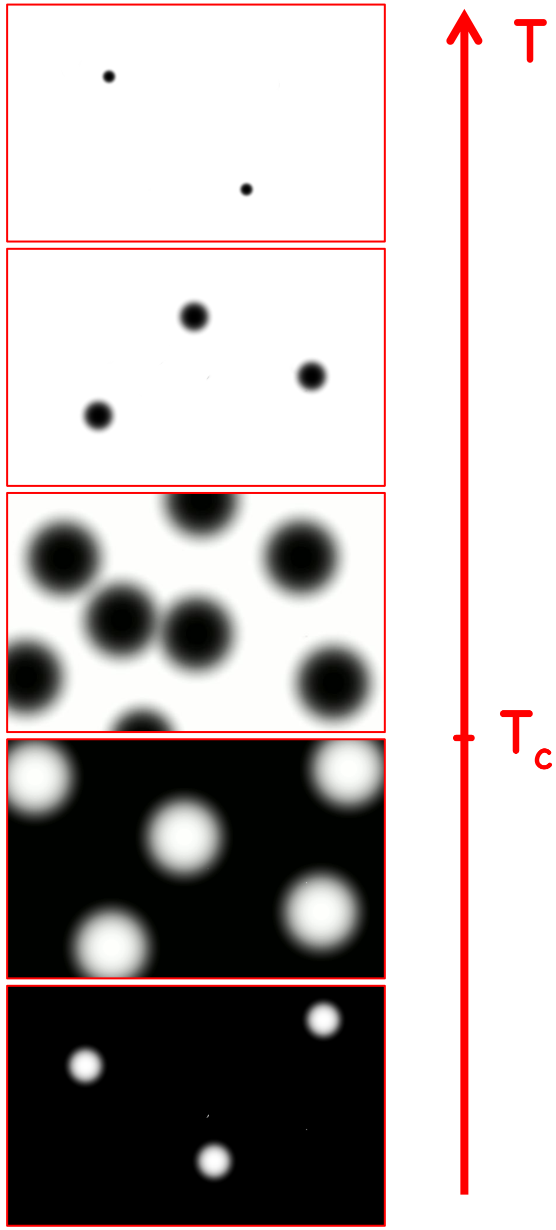
"slow" due to correlation ... ?

Critical Dynamics: What to expect ?

Fluctuations near (continuous) phase transition:

-> Increase of correlation length

Divergence at T_c -> long range order



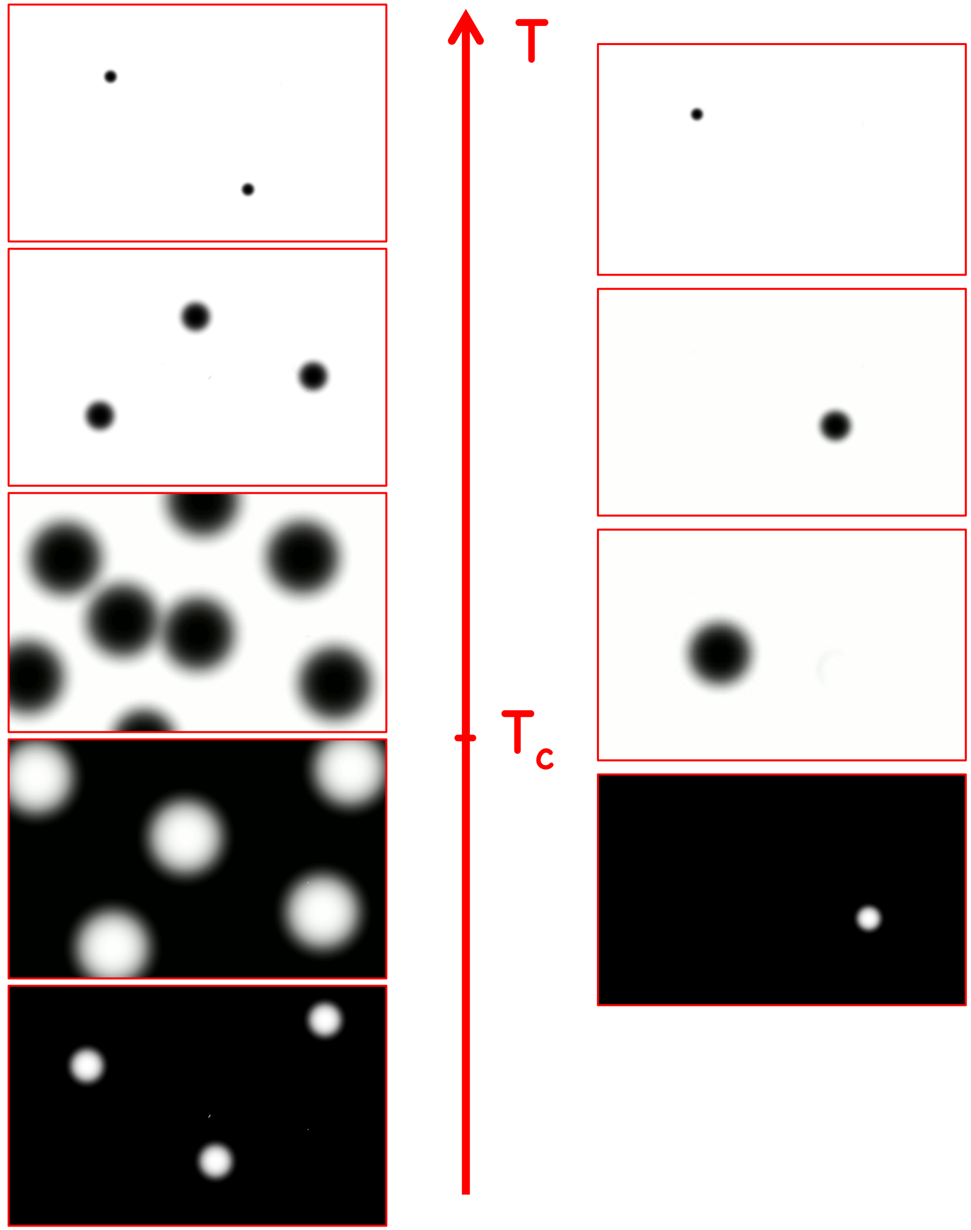
Interaction vs temperature

Critical Dynamics: What to expect ?

Fluctuations near (continuous) phase transition

-> Increase of correlation length

Divergence at T_c
->
long range order



-> Increase of correlation time

Divergence at T_c
->
Static order

J. Hemberger, Kolymbari, 2015

Critical Dynamics: What to expect ?

Description via critical exponents: (Landau mean field)

correlation length	$\xi \propto \left(\frac{T-T_C}{T_C}\right)^{\nu_\xi}$	$\nu_\xi = 1/2$
susceptibility	$\chi \propto \left(\frac{T-T_C}{T_C}\right)^{\gamma}$	$\gamma = 2\nu_\xi = 1$
correlation time (relaxation time)	$\tau_\xi \propto \xi^z \propto \left(\frac{T-T_C}{T_C}\right)^{\nu_\xi z}$	$\nu_\xi z = 1$

Correlation function:
(e.g. spin orientation)

$$G(r) = \langle \mathbf{a}(R) \cdot \mathbf{a}(R+r)\rangle - \langle \mathbf{a}(R)\rangle \cdot \langle \mathbf{a}(R+r)\rangle ,$$

$$G(r) \propto \exp\left(-\frac{r}{\xi}\right)$$

(same for *G(t)* …)

[Details: Hohenberg and Halperin, Rev. Mod. Phys. 49, 435 (1977)]

Critical Dynamics: What to expect ?

Spectroscopic approach:

Spectral weight $S \sim \frac{ne^2}{m} \sim \int_0^\infty \sigma' d\omega \int_0^\infty \varepsilon'' \omega d\omega \sim \Delta\varepsilon\, \omega_0{}^2 \sim const.$

Dielectric strength $\Delta\varepsilon \sim \frac{ne^2}{m} \sim \int_0^\infty \frac{\varepsilon''}{\omega} d\omega$

Softmode ("Lyddane-Sachs-Teller"):

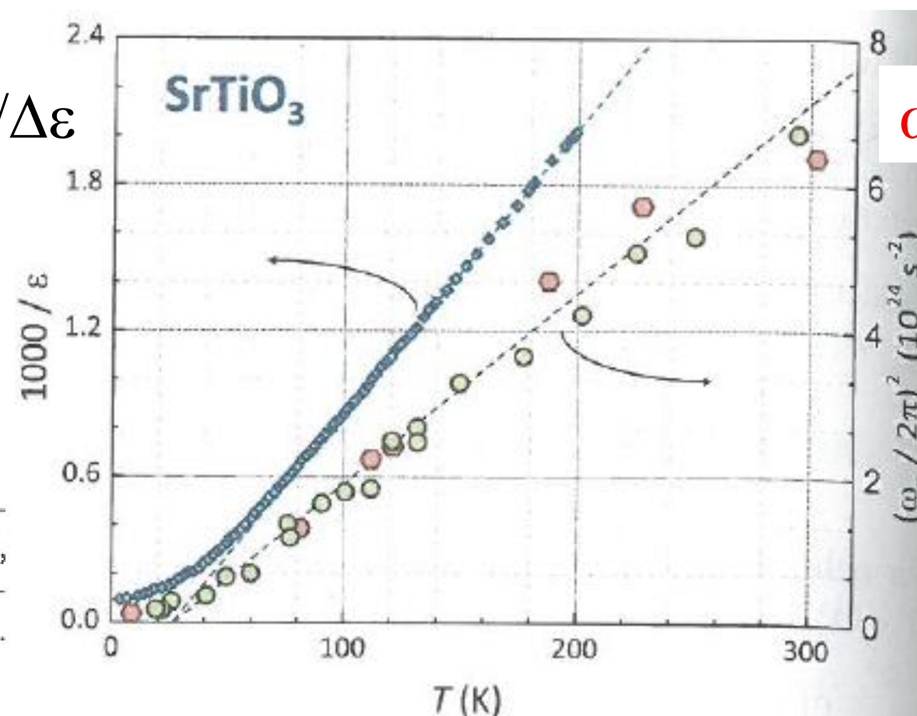
permittivity diverges at e.g. continuous **FE** phase transition

-> **polar** phonon mode gets slow

$$\to \; \omega_0{}^2 \sim 1/\Delta\varepsilon$$

$$\left(\tau_\xi{}^{-1} = \omega_0^2 / \gamma\right)$$

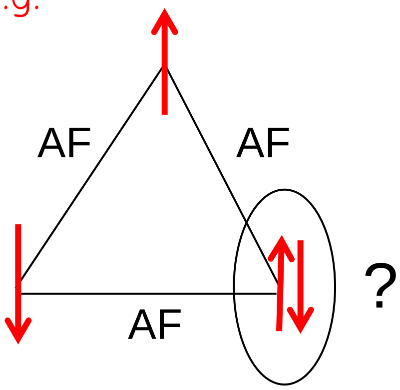
$1/\Delta\varepsilon$ $\omega_0{}^2$



(nach T. Sakudo, H. Unoki, Phys. Rev. Lett. 26, 851 (1971) und Y. Yamada, G. Shirane, J. Phys. Soc. Jpn 26, 396 (1969)).

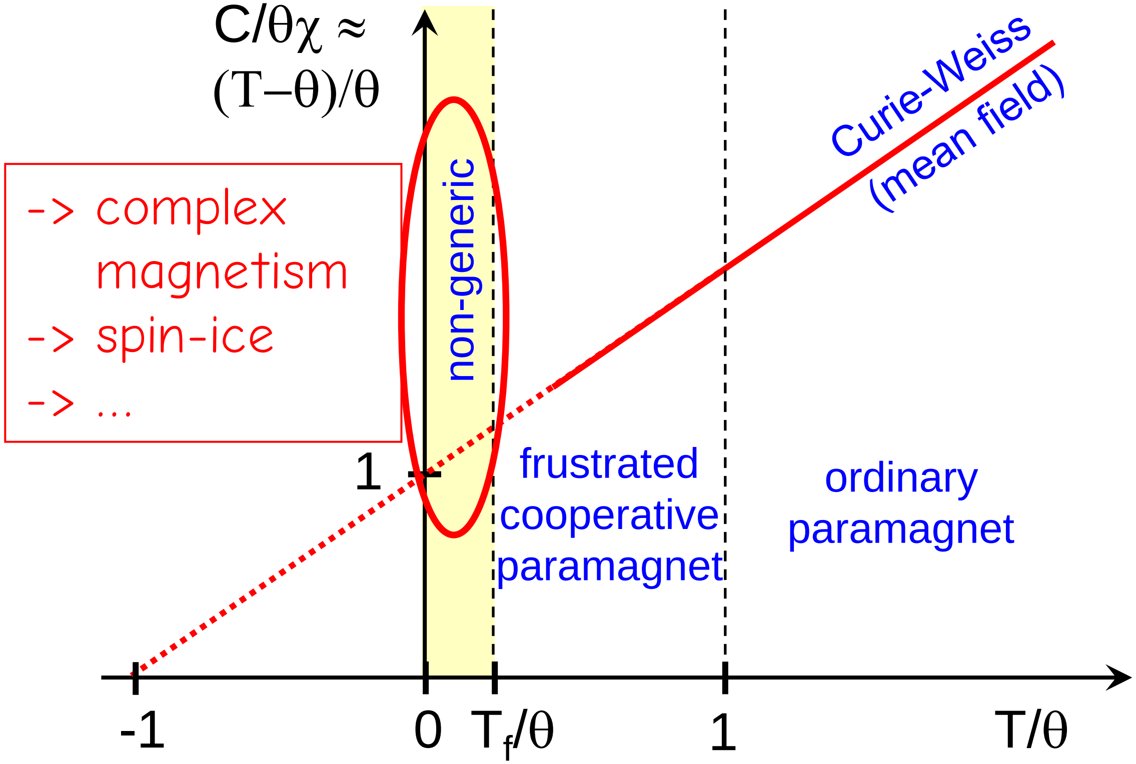
Additional impact: Frustration

e.g.



"not all competing interactions can be satisfied"

Normalized inverse susceptibility:



Accentuation of small energy scales ...

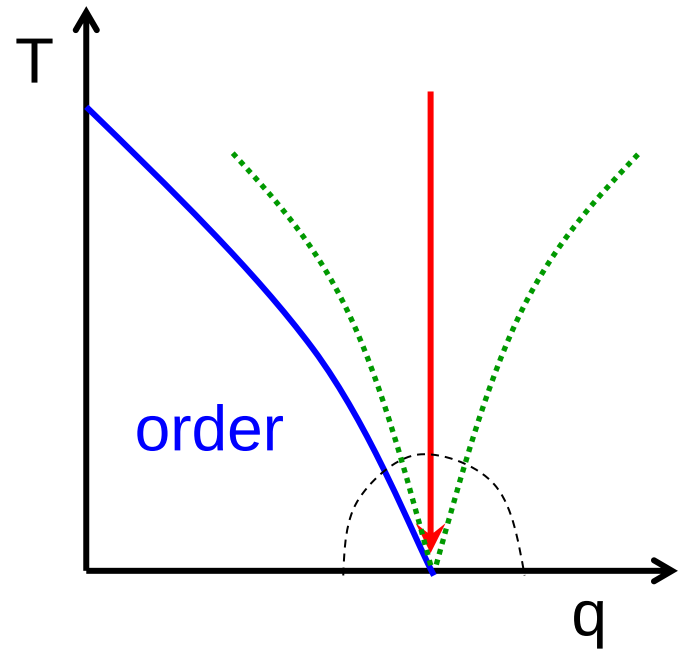
→ low lying excitations, degenerate ground states, fluctuations
often sensitive to only small perturbation

Additional impact: Quantum fluctuations

Classical limit: $k_B T_c \gg \hbar/\tau_\xi$ -> Temperature drives fluctuations

Suppression of the energy scale for the correlated state:

$T_c \to 0$: $k_B T_c < \hbar/\tau_\xi$



-> Quantum Phase Transition

-> other control parameter ...

-> altered dynamics ...

Experimental

Broadband spectroscopy
in electric and magnetic fields*

to investigate

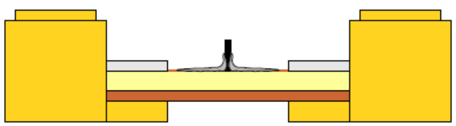
Dynamics of
Correlated Matter

LF/RF/MW/THz/...

mHz ... continuous broadband ... 1 THz

(1 GHz ~ 4 μeV)

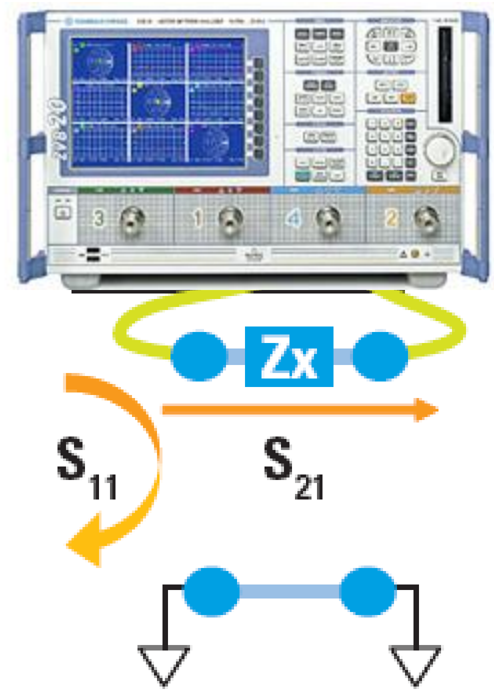
from 30 mK to 400 K,
Magnetic fields up to 14 T



Sample prepared as capacitor (or inductor) in transmission configuration

E.g. microwave spectroscopy using micro-strip setup

... measurement of complex scattering parameters S_{11} & S_{21} -> complex sample impedance **Zx.**



$$S_{11} = \frac{Zx}{Zx + 100} \qquad S_{21} = \frac{100}{Zx + 100}$$

Coworkers/Collaboration

Spin-Ice
$Dy_2Ti_2O_7$

LF,RF,MW,THz

Christoph Grams
Daniel Niermann
Manuel Pietsch
Steffen Harms

Samples Martin Valldor
(now MPI-CPfS, Dresden)
Thomas Lorenz
Simon Scharfe

Theory Markus Garst (Cologne)
Daniel Khomskii
Simon Trebst

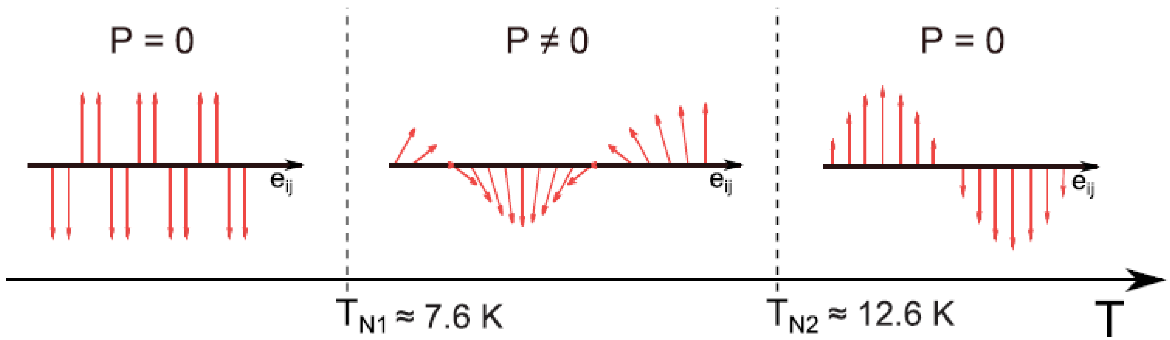
Funding:

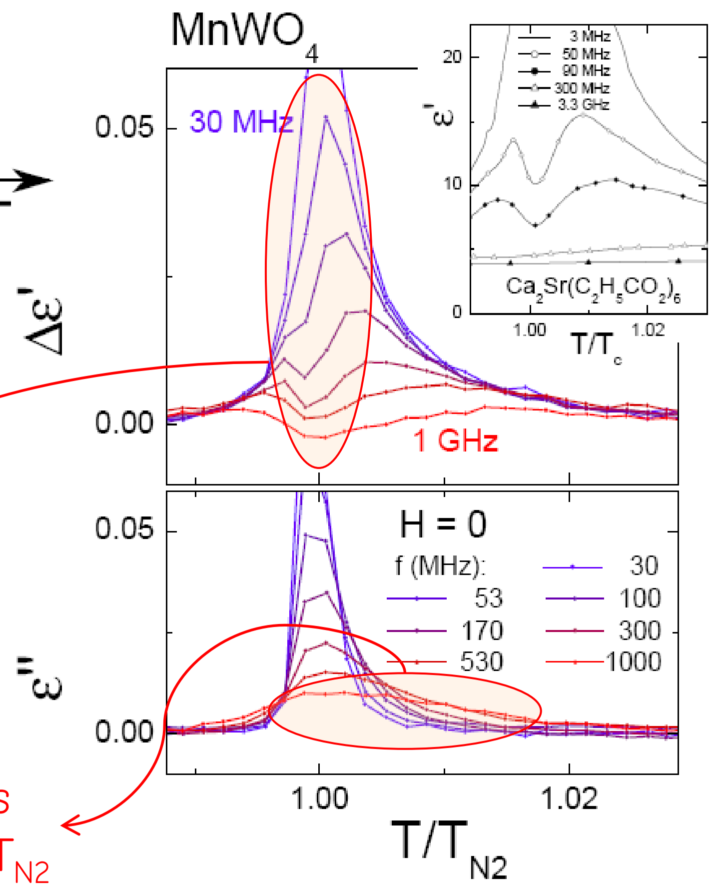
Quantum Matter and Materials
Center of Excellence
at the University of Cologne
QM²

Deutsche
Forschungsgemeinschaft
research grant HE-3219/2-1

A "usual" scenario: e.g.: magnetoelectric multiferroic $MnWO_4$

Critical slowing-down on entering the spiral, multiferroic phase





Characteristic minimum
in $\varepsilon'(T)$ at T_{N2}
-> critical damping

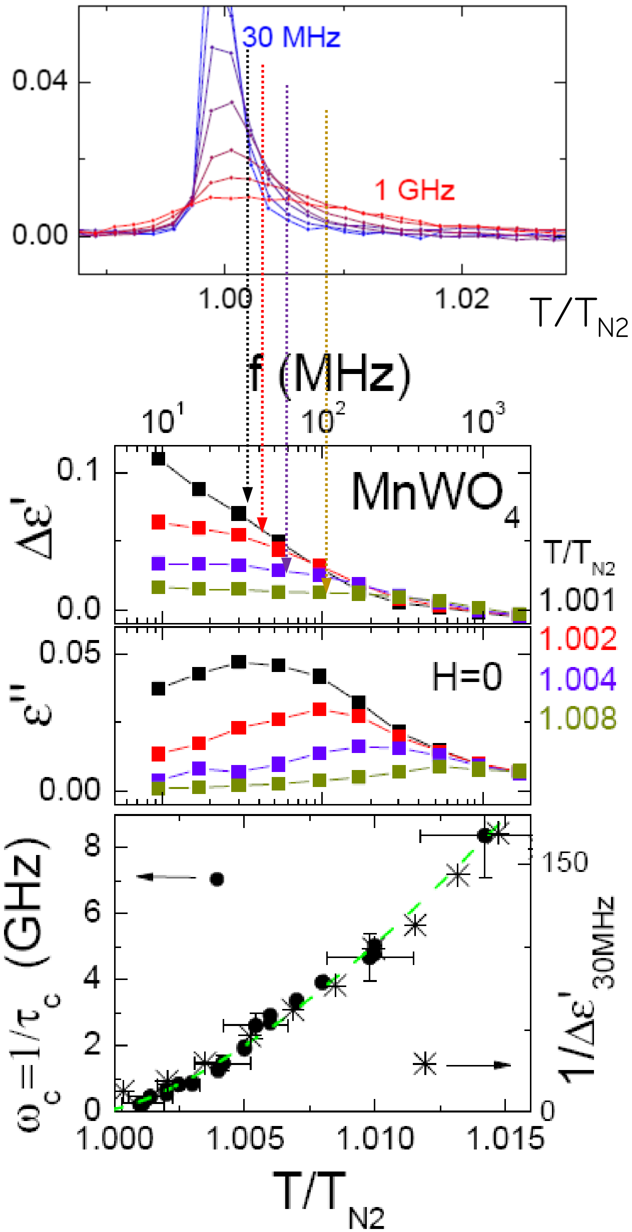
Known from proper ferroelectrics
with over-damped soft-mode
(but much smaller response !!!)

Blinc, Zeks: *Soft modes in ferroelectrics and antiferroelectrics*, North-Holland, 1974

Dispersive loss
already above T_{N2}
-> critical slowing down

[PRL 114, 037204 (2015)]

Critical slowing down on approaching T_{N2} from above



[PRL 114, 037204 (2015)]

step in $\varepsilon'(\omega)$, peak in ε'' $'(\omega)$ <-> $\omega\tau=1$

$$\tau \propto \left(\frac{T-T_{N2}}{T_{N2}}\right)^{\nu_\xi z} \quad \text{->} \quad \nu_\xi z \approx 1.3$$

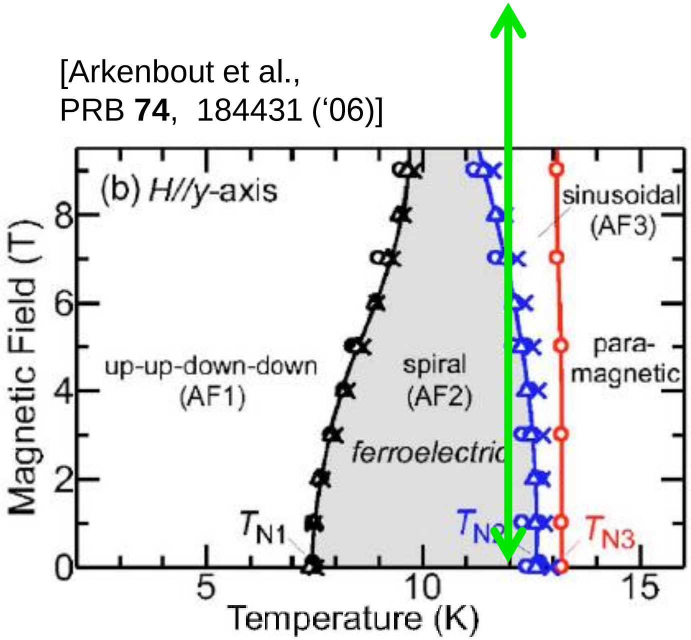
Theory:
Ginzburg-Landau exp.
3D-Ising **magnetic** order-parameter
[H. Schenk, AG Nattermann]

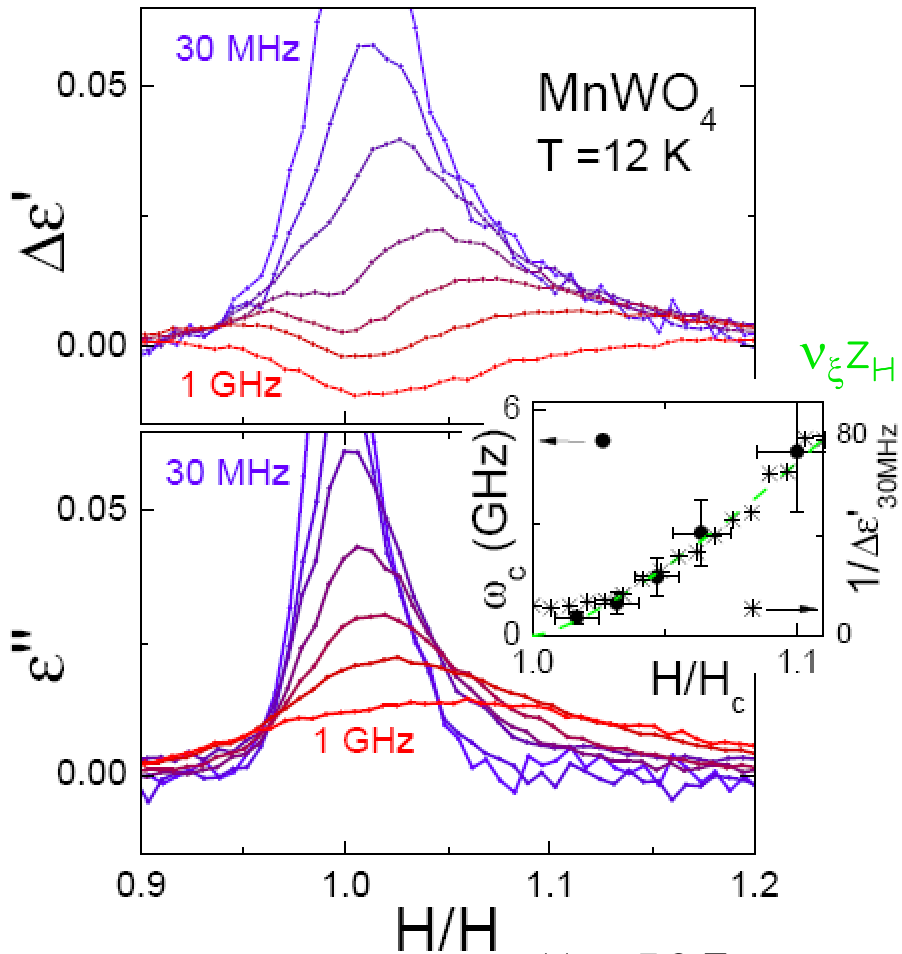
-> $\nu_\xi z = 1.272$

Softening of a electro-magnon mode (with small spectral weight due to weak magneto-electric coupling)

Magnetic field driven critical slowing down

[Arkenbout et al.,
PRB **74**, 184431 ('06)]





$\nu_\xi z_H \approx 1.3$

$H_c \approx 7.3$ T

-> Magnetic field
driven
soft mode !

[PRL 114, 037204 (2015), D. Niermann, C.P. Grams, P. Becker, L. Bohatý, H. Schenck, J. Hemberger]

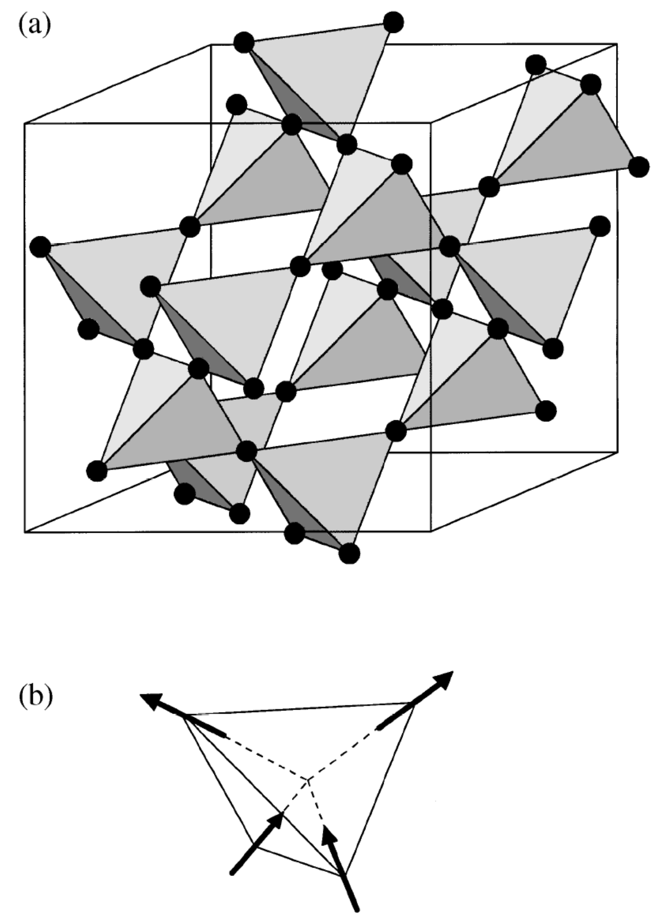
$Dy_2Ti_2O_7$

- Pyrochlore structure:
  Magnetic Dy-ions are located at corner sharing tetrahedrons

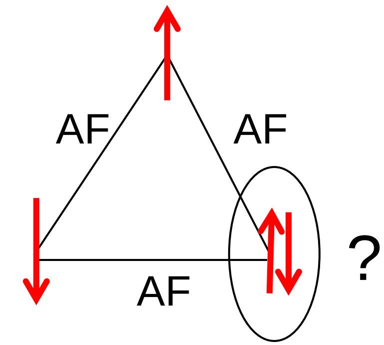
- Dy^{3+}: $4f^9$ -> $S=5/2$, $L=5$, $g=4/3$
  -> $m=10\mu_B$!

- Effective Ising spins:
  Spins point in the local [111] directions due to crystal field.

Pyrochlore lattice



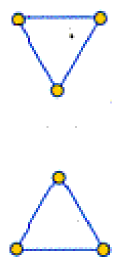
Triangles ... -> Frustration



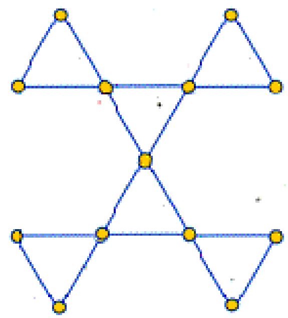
1D ("dimensionally frustrated")

2D

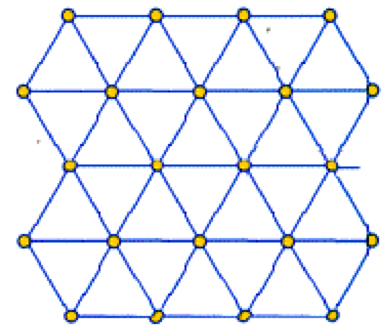
triangles



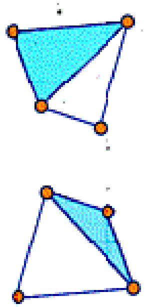
kagome



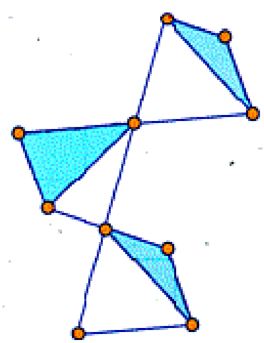
triangular lattice



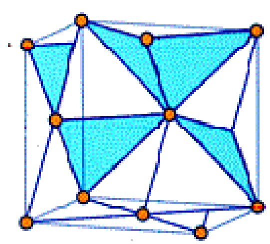
3D



tedrahedra

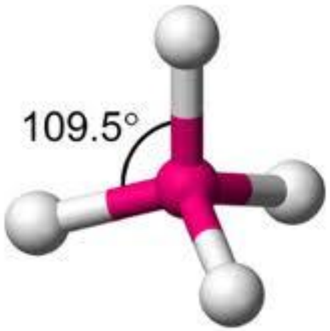


pyrochlore

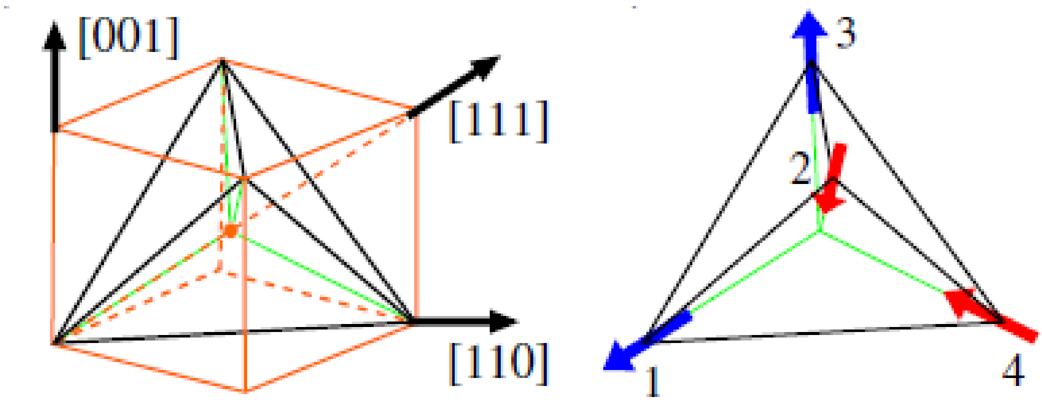


fcc

$Dy_2Ti_2O_7$ - Spin-Ice due to FM-Frustration



Interactions on a single tetrahedron
-> Ising behavior along {111}



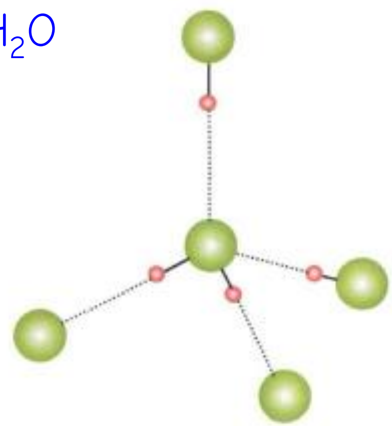
1-2 FM
1-3 AFM
1-4 FM
2-3 FM
2-4 AFM
3-4 FM

-> Optimal for 2in/2out on each tetrahedron
(energy cost for 3in/1out -> 2.2 K)

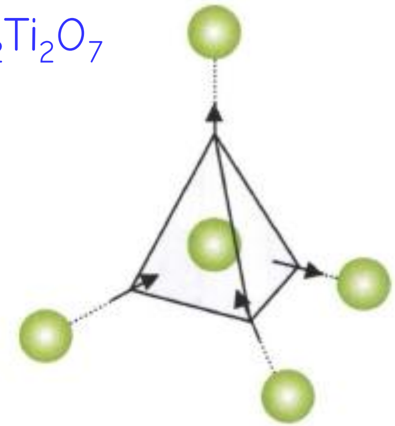
Macroscopically degenerate ground state with local order
-> well-defined zero-point (residual) entropy -> Ice

Spin Ice <-> Water Ice

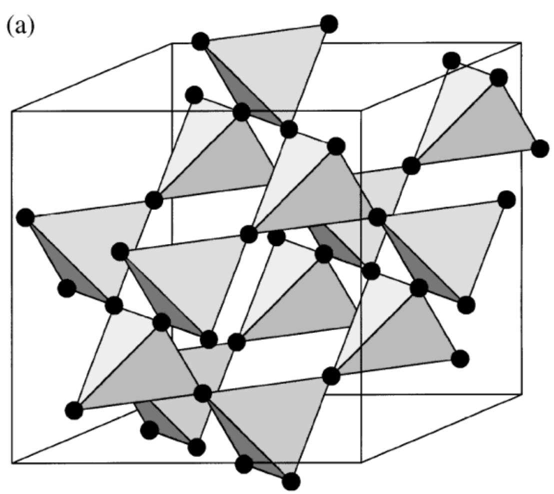
H_2O



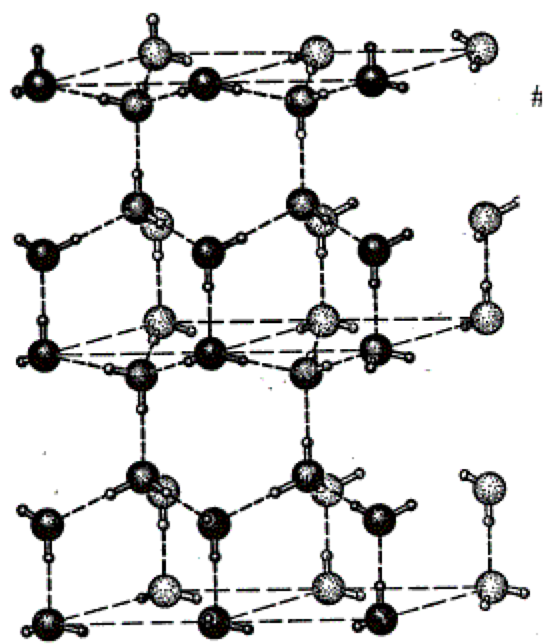
$Dy_2Ti_2O_7$



Ti <-> O
Dy-Spins <-> H bonds
In/out <-> short bond
long bond



Pyrochlore lattices



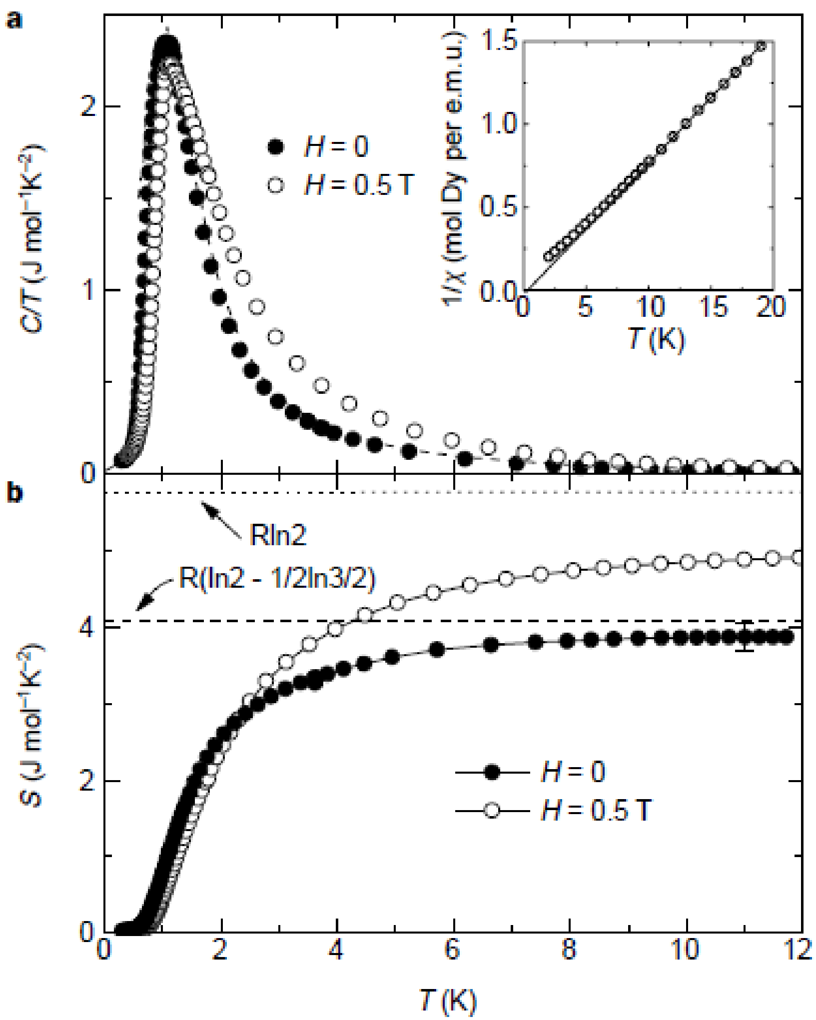
Pauling's
Ice rule:

-> 2in/2out

$Dy_2Ti_2O_7$

Zero-point entropy in 'spin ice'

A. P. Ramirez*, A. Hayashi†, R. J. Cava†, R. Siddharthan‡ & B. S. Shastry‡ Nature 399, 333 (1999)



N tedrahedra # config.

4/2=2 spins per (corner sharing) tedrahedron -> 2^{2N}

Only 6/16 config. allowed -> $\cdot(6/16)^N$

-> $2^{2N}\cdot(6/16)^N = (3/2)^N$

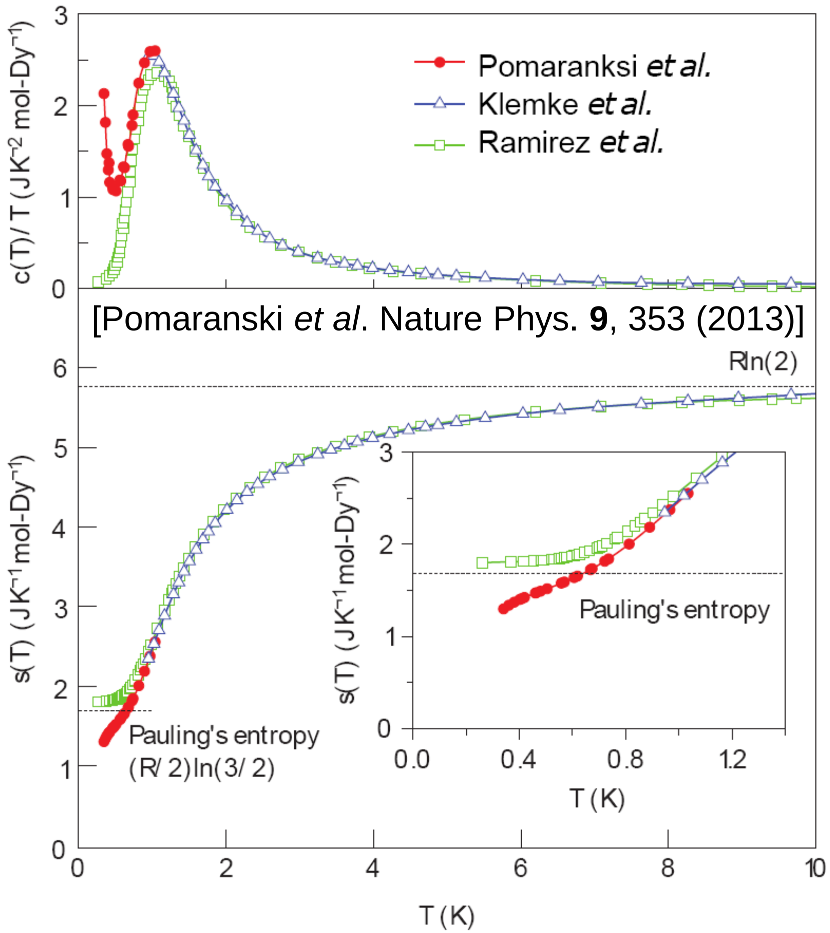
-> $\Delta S = R\ln(3/2)$

(total magnetic entropy with 2 Ising-spins per system:
-> $2R\ln(2)$

[Linus Pauling]

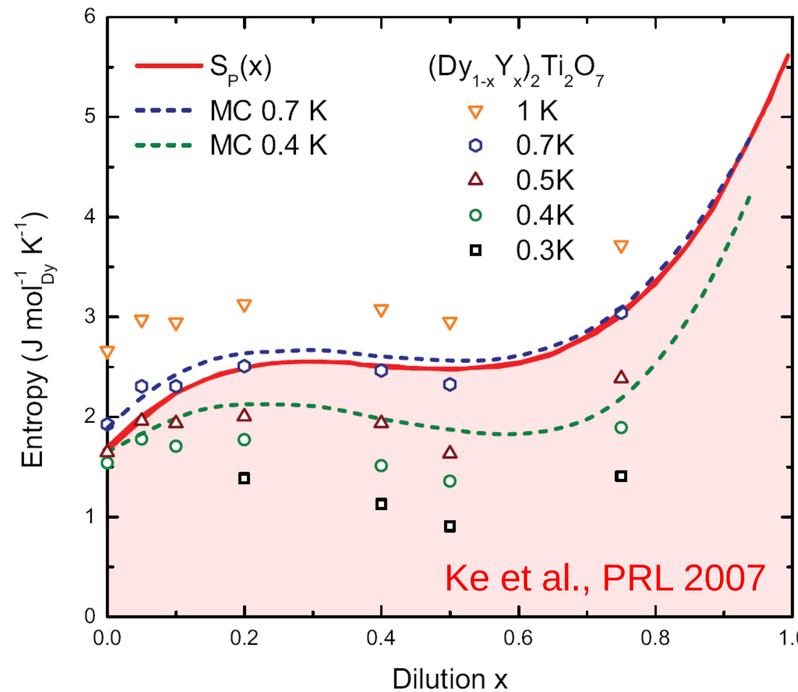
ΔS can be reduced in magnetic field

$Dy_2Ti_2O_7$ -> residual entropy ?



[Pomaranski *et al*. Nature Phys. **9**, 353 (2013)]

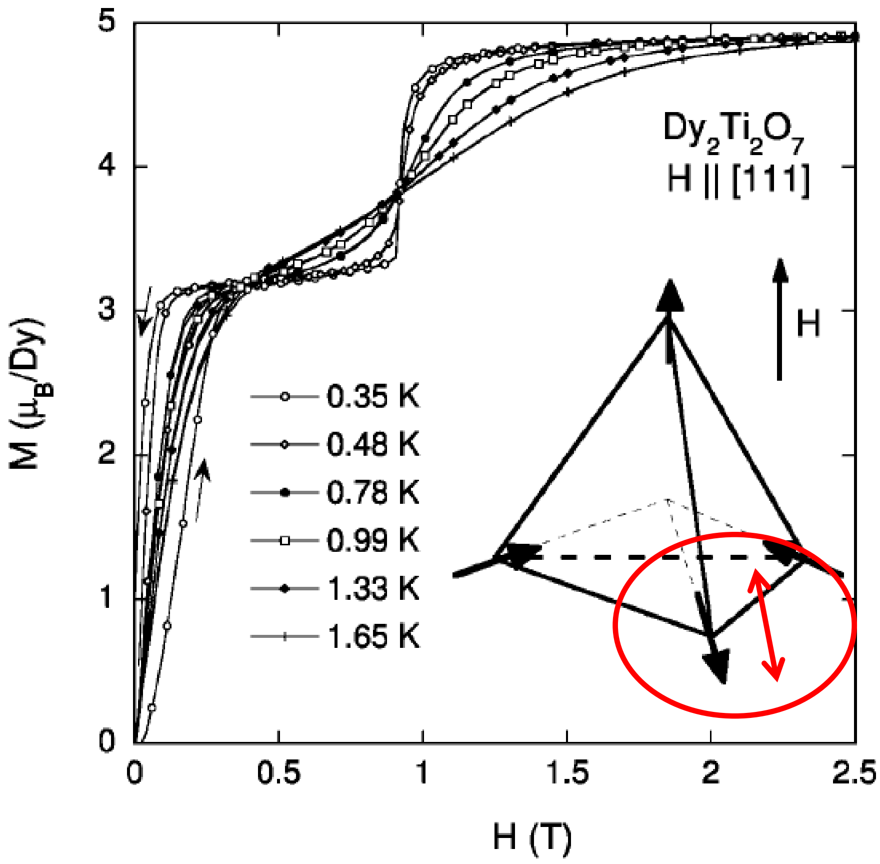
Dilution of the spin system:



[Simon Scharffe *et al*. arXiv: **1503.03856** (2015)]

-> Poster: "Specific Heat and Heat Transport in Spin-Ice Materials"

$Dy_2Ti_2O_7$ – breaking the ice-rule in magnetic field



3in/1out -> $(1 + 3\cdot1/3)\cdot 10\mu_B = 20\mu_B$ per 4 Dy

2in/2out -> $(1 + 1\cdot1/3)\cdot 10\mu_B \approx 13\mu_B$ per 4 Dy

3in/1out - 2in/2out

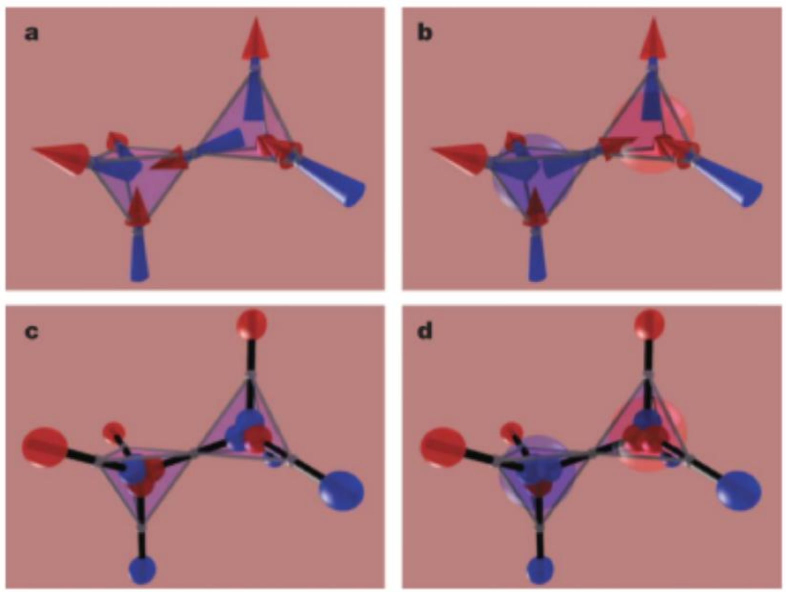
-> 2.2 K

[Sakakibara et al. PRL 90, 207205 (2003)]

$Dy_2Ti_2O_7$ spin-ice

Broken ice-rule -> **Magnetic monopoles**

(excitations of the magnetic structure)



C. Castelnovo, R. Moessner, S. L. Sondhi, ***Nature***, **451**, 42 (2008)

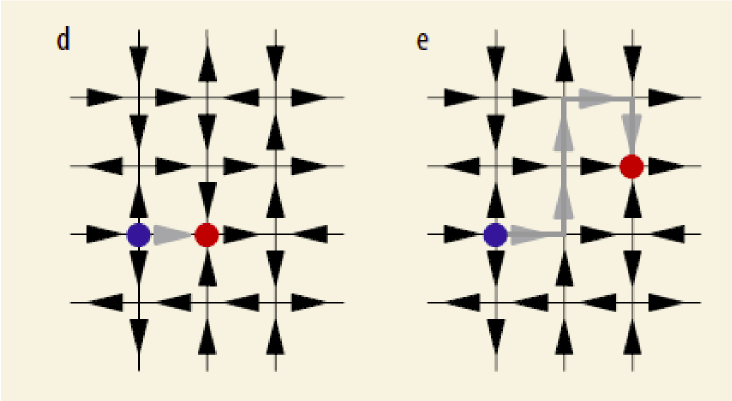
Always pairs of mono- & anti-monopoles

$$\mathrm{div}\,\mathbf{E} = \rho/\varepsilon_0$$

$$\mathrm{div}\,\mathbf{B} \neq 0 \quad ?$$

Postulated by Dirac ...

Can move without further breaking of ice-rule



Magnetic Monopoles …

nature **451**, 42 (2008)

nature Vol 451|3 January 2008|**doi:10.1038/nature06433**

LETTERS

Magnetic monopoles in spin ice

C. Castelnovo[1], R. Moessner[1,2] & S. L. Sondhi[3]

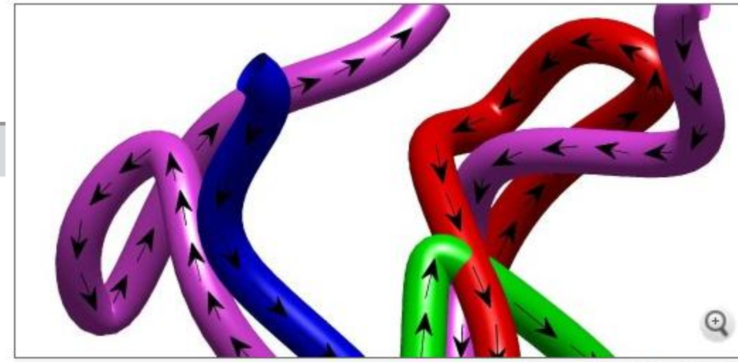
Electrically charged particles, such as the electron, are ubiquitous. In contrast, no elementary particles with a net magnetic charge have ever been observed, despite intensive and prolonged searches (see ref. 1 for example). We pursue an alternative strategy, namely that of realizing them not as elementary but rather as emergent particles—that is, as manifestations of the correlations present in a strongly interacting many-body system. The most prominent examples of emergent quasiparticles are the ones with fractional electric charge *e*/3 in quantum Hall physics[2]. Here we propose that magnetic monopoles emerge in a class of exotic magnets known collectively as spin ice[3–5]: the dipole moment of the underlying electronic degrees of freedom fractionalises into monopoles. This would account for a mysterious phase transition observed experimentally in spin ice in a magnetic field[6,7], which is a liquid–gas transition of the magnetic monopoles. These monopoles can also be detected by other means, for example, in an experiment modelled after the Stanford magnetic monopole search[8].

04.09.2009 Drucken | Senden | Feedback | Merken

Spiegel Online

Monopole

Forscher entdecken lang gesuchte Magnetismus-Exoten



HZB / D.J.P. Morris & A. Tennant

"Spin-Spaghetti": An den Enden bildeten sich jeweils magnetische Monopole

Sie sind wie Yin und Yang: Nord- und Südpol eines Magneten können nur zusammen auftreten. Doch einem Forscherteam mit deutscher Beteiligung ist es nun zum ersten Mal gelungen, diese Regel außer Kraft zu setzen - und sogenannte magnetische Monopole herzustellen.

Dirac Strings and Magnetic Monopoles in the Spin Ice $Dy_2Ti_2O_7$

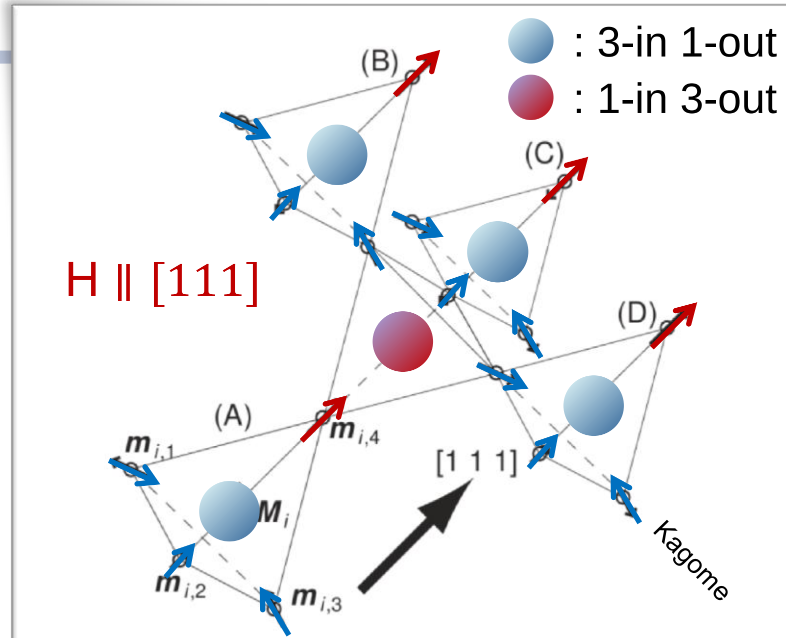
D. J. P. Morris,[1*] D. A. Tennant,[1,2*] S. A. Grigera,[3,4*] B. Klemke,[1,2] C. Castelnovo,[5] R. Moessner,[6] C. Czternasty,[1] M. Meissner,[1] K. C. Rule,[1] J.-U. Hoffmann,[1] K. Kiefer,[1] S. Gerischer,[1] D. Slobinsky,[3] R. S. Perry[7]

Sources of magnetic fields—magnetic monopoles—have so far proven elusive as elementary particles. Condensed-matter physicists have recently proposed several scenarios of emergent quasiparticles resembling monopoles. A particularly simple proposition pertains to spin ice on the highly frustrated pyrochlore lattice. The spin-ice state is argued to be well described by networks of aligned dipoles resembling solenoidal tubes—classical, and observable, versions of a Dirac string. Where these tubes end, the resulting defects look like magnetic monopoles. We demonstrated, by diffuse neutron scattering, the presence of such strings in the spin ice dysprosium titanate ($Dy_2Ti_2O_7$). This is achieved by applying a symmetry-breaking magnetic field with which we can manipulate the density and orientation of the strings. In turn, heat capacity is described by a gas of magnetic monopoles interacting via a magnetic Coulomb interaction.

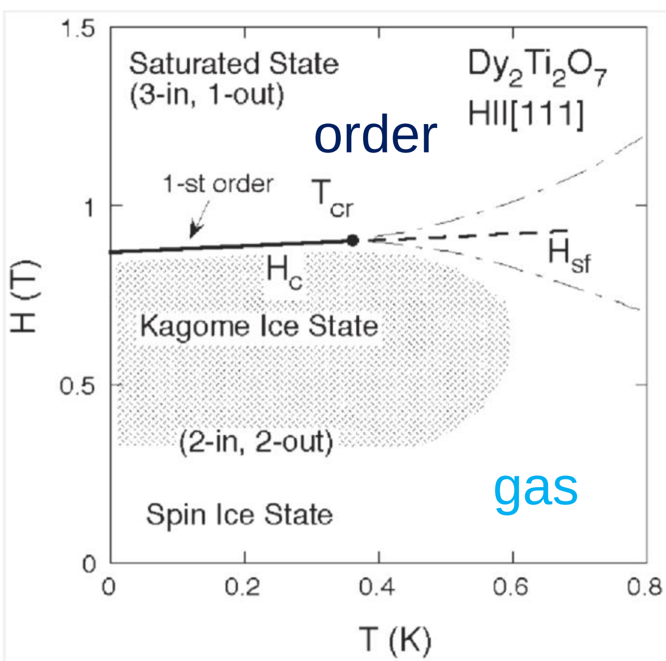
Science **326**, 411 (2009)

$Dy_2Ti_2O_7$

H || [111] fixes one **spin** per tetrahedron:
-> H enhances monopole density
(like p in the case of H_2O gas)
-> Kagome Ice state for intermediate fields
(disorder restricted to Kagome planes)
-> Monopole-Anti-monopole order
at high fields

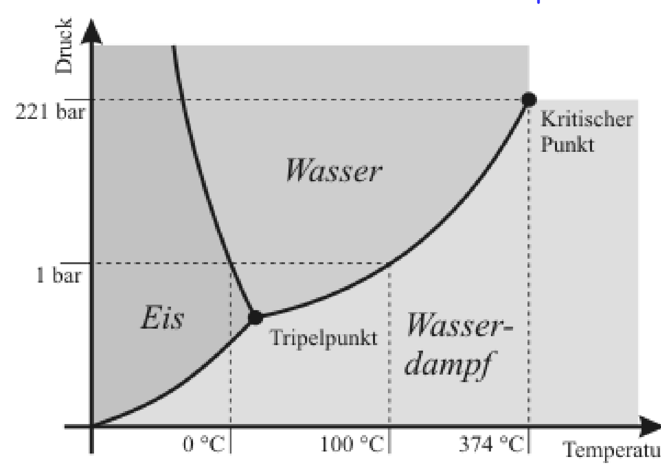


M. Saito, R. Higashinaka, Y. Maeno, *PRB*, **72**, 144422 (2005)



H. Aoki, T. Sakakibara,K. Matsuhira, Z. Hiroi, *JPSJ*, **73(10)**, 2851-2856. (2004)

-> monopole gas to monopole order transition with critical endpoint



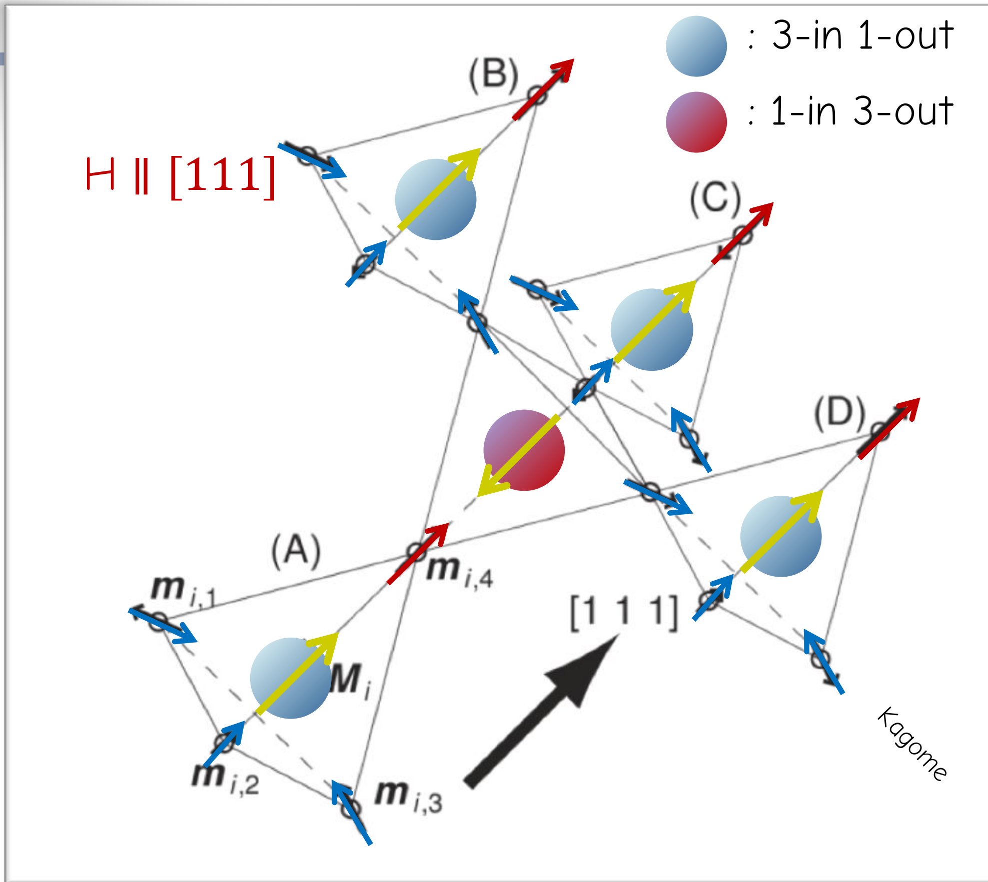
In analogy to water …

Dipoles on monopoles

Magnetic monopoles carry
electric dipoles
[D.I. Khomskii, ***Nature Comm.***, **3**, 1 (2012)]

Spin-orbit coupling:

-> Electric dipole moment points to point the one different spin in the 3in/1out or 1in/3out tedrahedra
-> magneto-electric coupling



M. Saito, R. Higashinaka, Y. Maeno, *PRB*, **72**, 144422 (2005)

Magnetic monopole order
<->
Anti-ferroelectric order

-> Monopole **dynamics** accessible
with **magnetic** and **dielectric** spectroscopy !

Electric dipoles on magnetic monopoles

H ‖ [111]

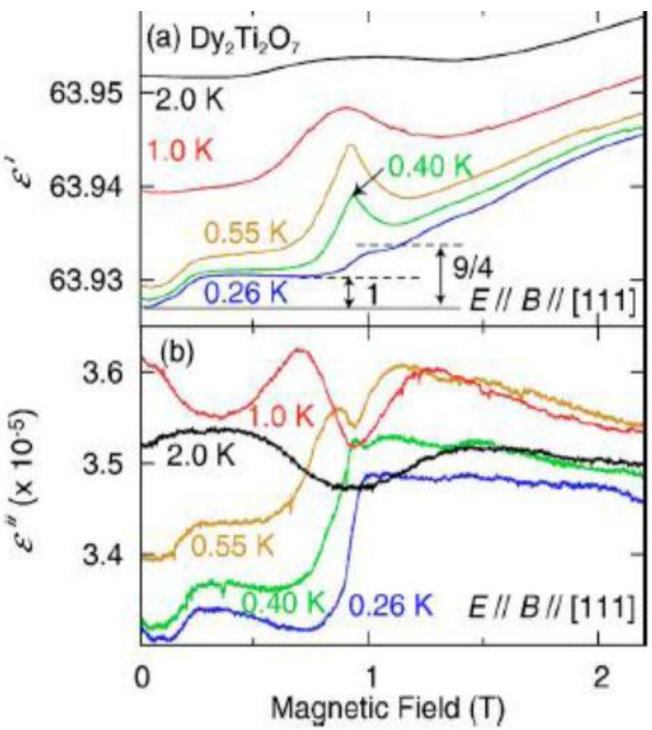
D.I. Khomskii, ***Nature Comm.***, **3**, 1 (2012)

Experimental evidence:

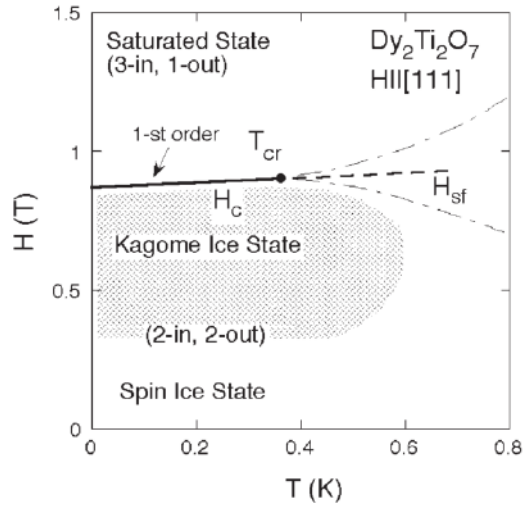
PHYSICAL REVIEW B 72, 144422 (2005)

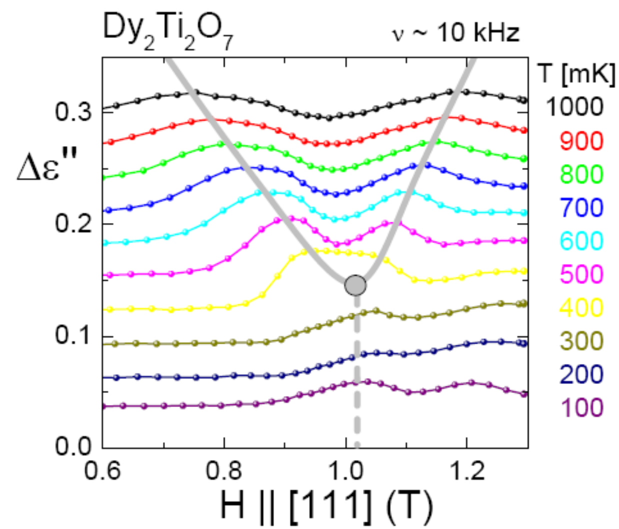
Magnetodielectric response of the spin-ice $Dy_2Ti_2O_7$

Masafumi Saito,[1] Ryuji Higashinaka,[1] and Yoshiteru Maeno[1,2,*]



[Aoki et al., JPSJ (2004)]





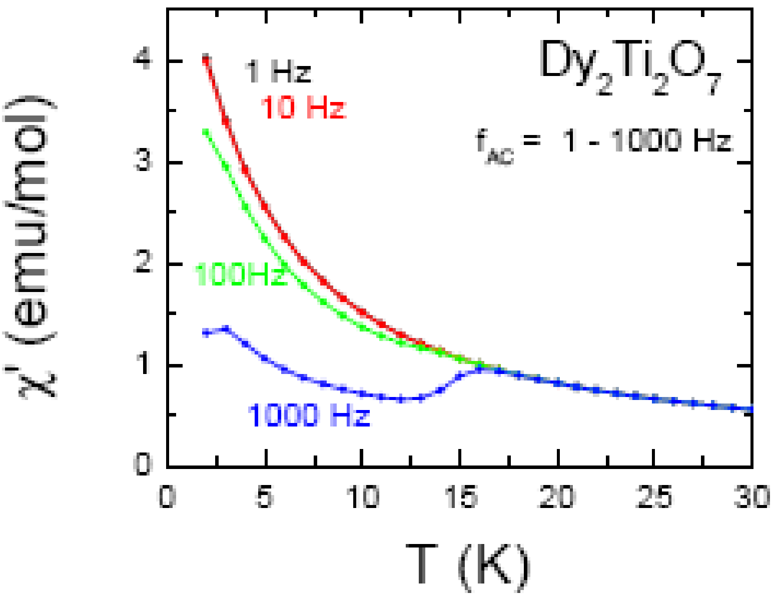
-> Anomalies at and above transition into monopole order

But: frequency dependent !

(large offset in ε' due to titanium sublattice)

-> dynamics ...

Magnetic AC-susceptibility

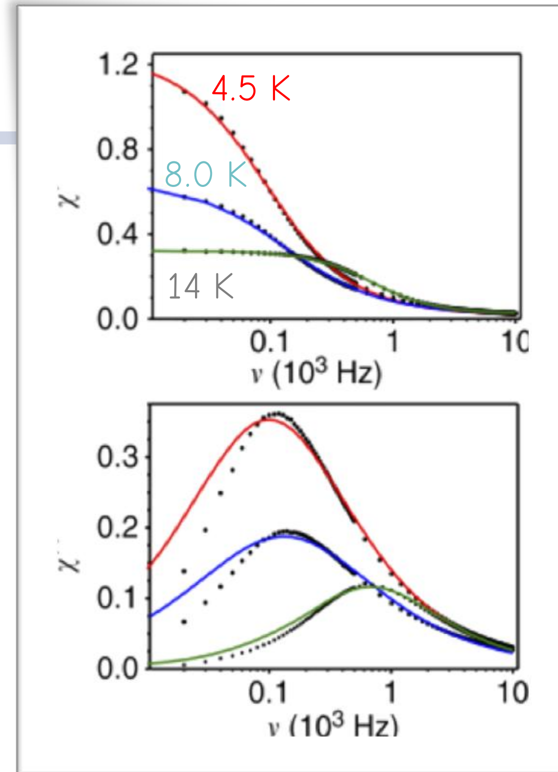


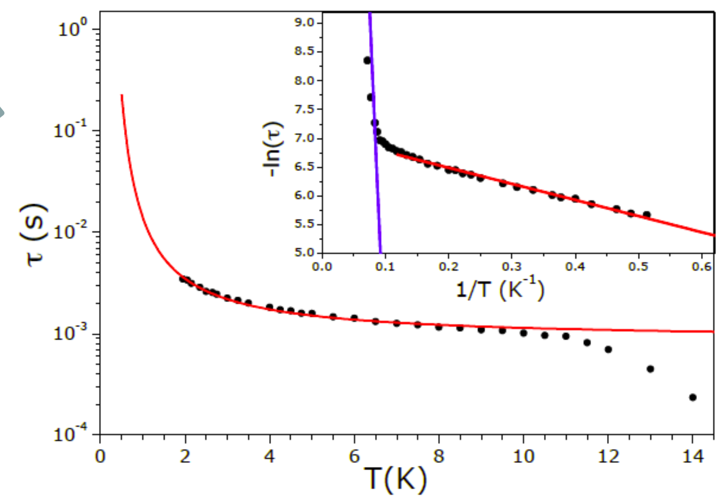
-> "freezing" below 15K
(dispersion, Debye-like relaxation)
-> χ stays finite at HF
-> two contributions: slow and fast,
"thermodynamic" and "adiabatic"

$$\frac{\chi(\omega)-\chi_S}{\chi_T-\chi_S}=\frac{1}{1+(i\omega\tau)^{1-\alpha}}$$

-> relaxation time of the slow process
stays relatively fast down to low temperatures
(Quantum tunneling ?)

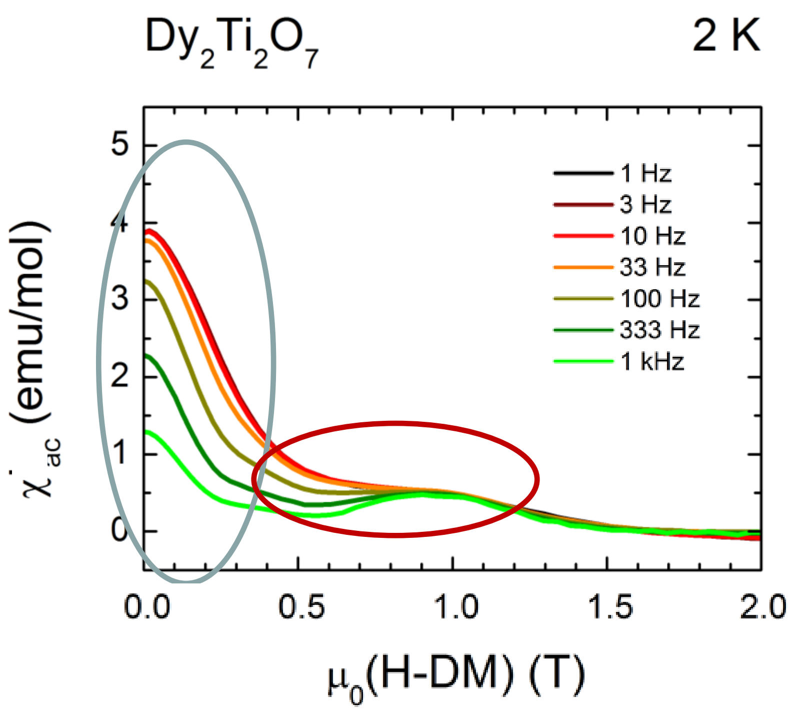
$$\tau = 1/2\pi\nu_p$$





Bovo, Bramwell, et al.,
Nature com. 4, 1535. (2013)

Magnetic field dependence susceptibility of χ_{AC} at low T

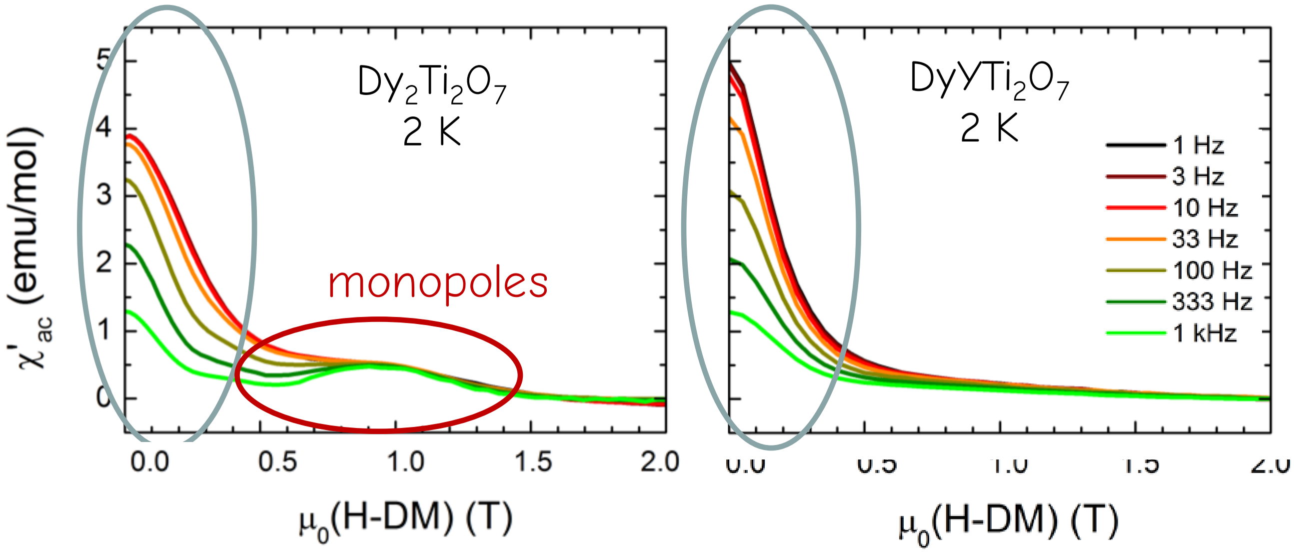


Two processes:

- slow dynamics dominant at low fields
- fast dynamics above approx. 1 kHz

Magnetic field dependence susceptibility of χ_{AC} at low T

collective spin reorientation



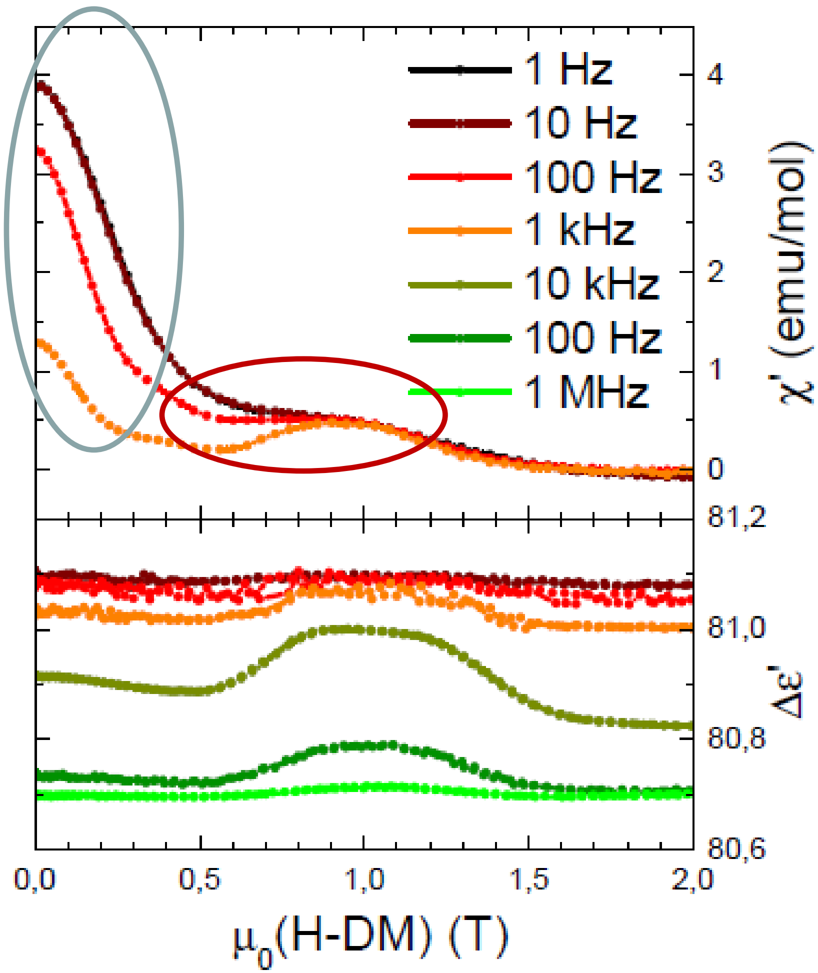
$DyYTi_2O_7$:
every tetrahedron
has only 2 spins
-> no broken ice-rule
-> no monopoles

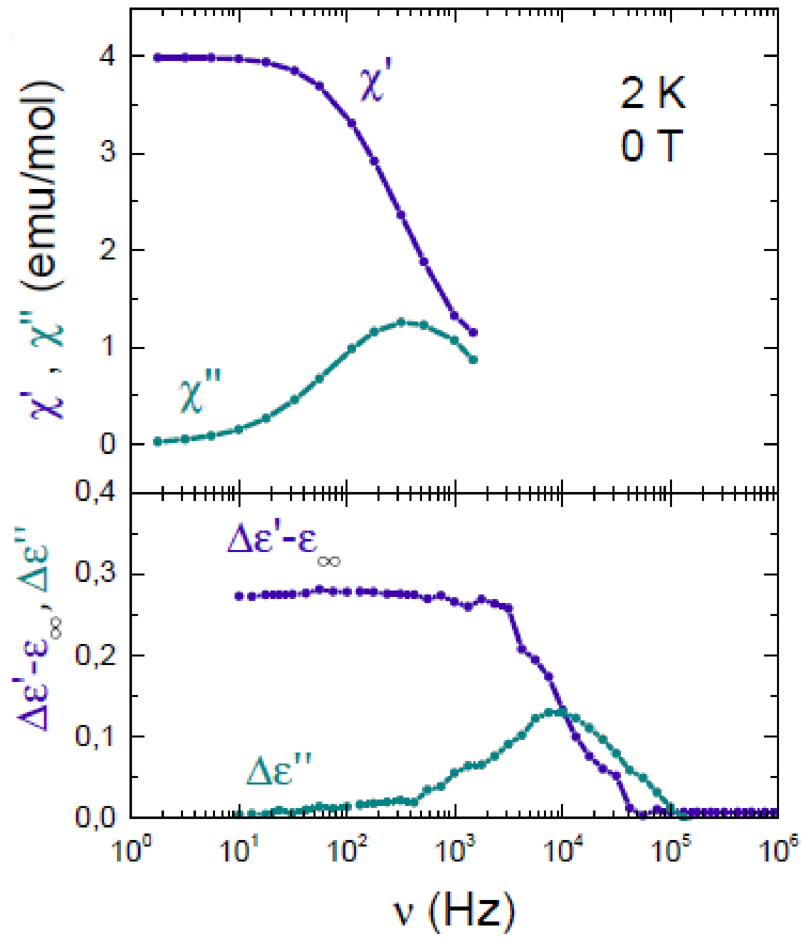
-> fast process due to monopole hopping !

- contribution to **ε** and χ_s ?
- larger frequency range needed ...

-> dielectric spectroscopy

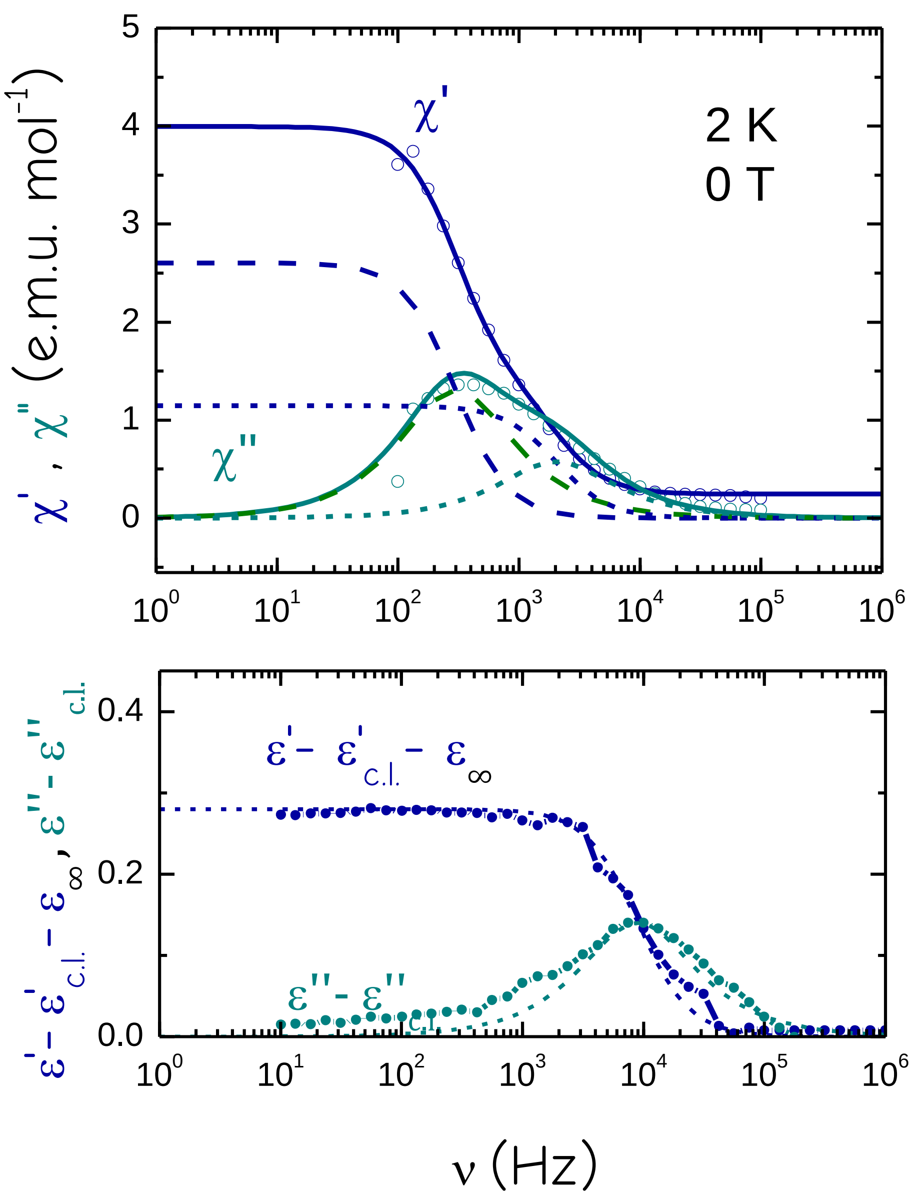
Comparison between χ_{AC} and ε





-> (only) fast dynamics are visible in dielectric spectroscopy !?

Comparison between "high frequency" χ_{AC} and ε

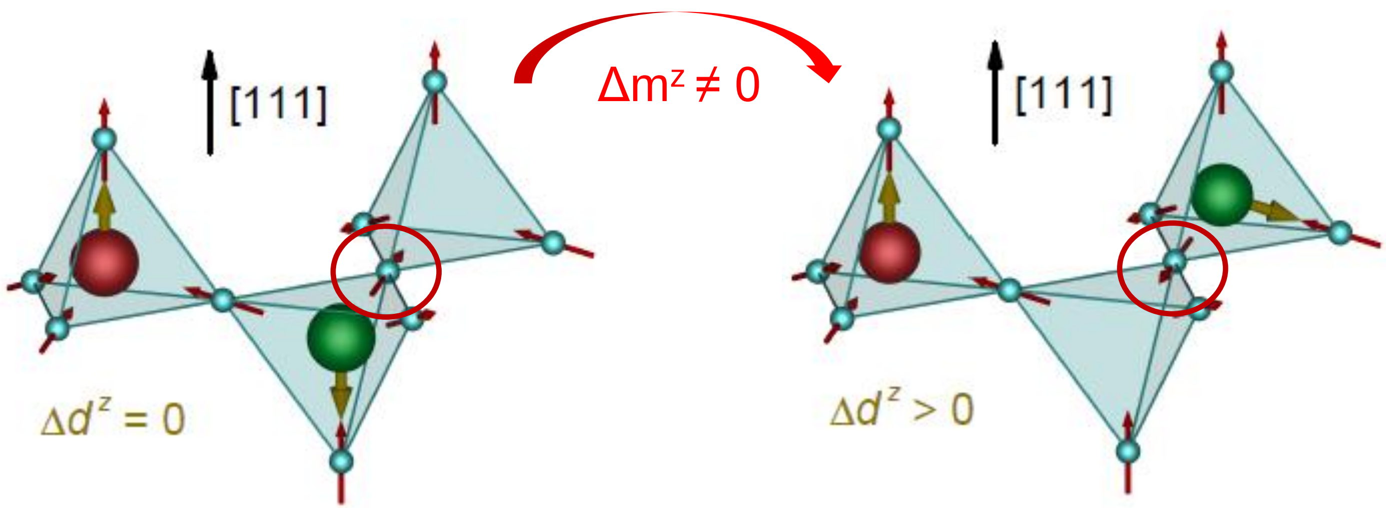


-> broadened spectra in χ_{AC}
containing **slow** and
fast processes

-> broadened spectra in ε
containing only
fast processes

Monopole contribution to permittivity -> microscopic origin

Hopping of monopoles
(for H || [111] mainly confined within Kagome plane):



-> spin flip -> magnetic response

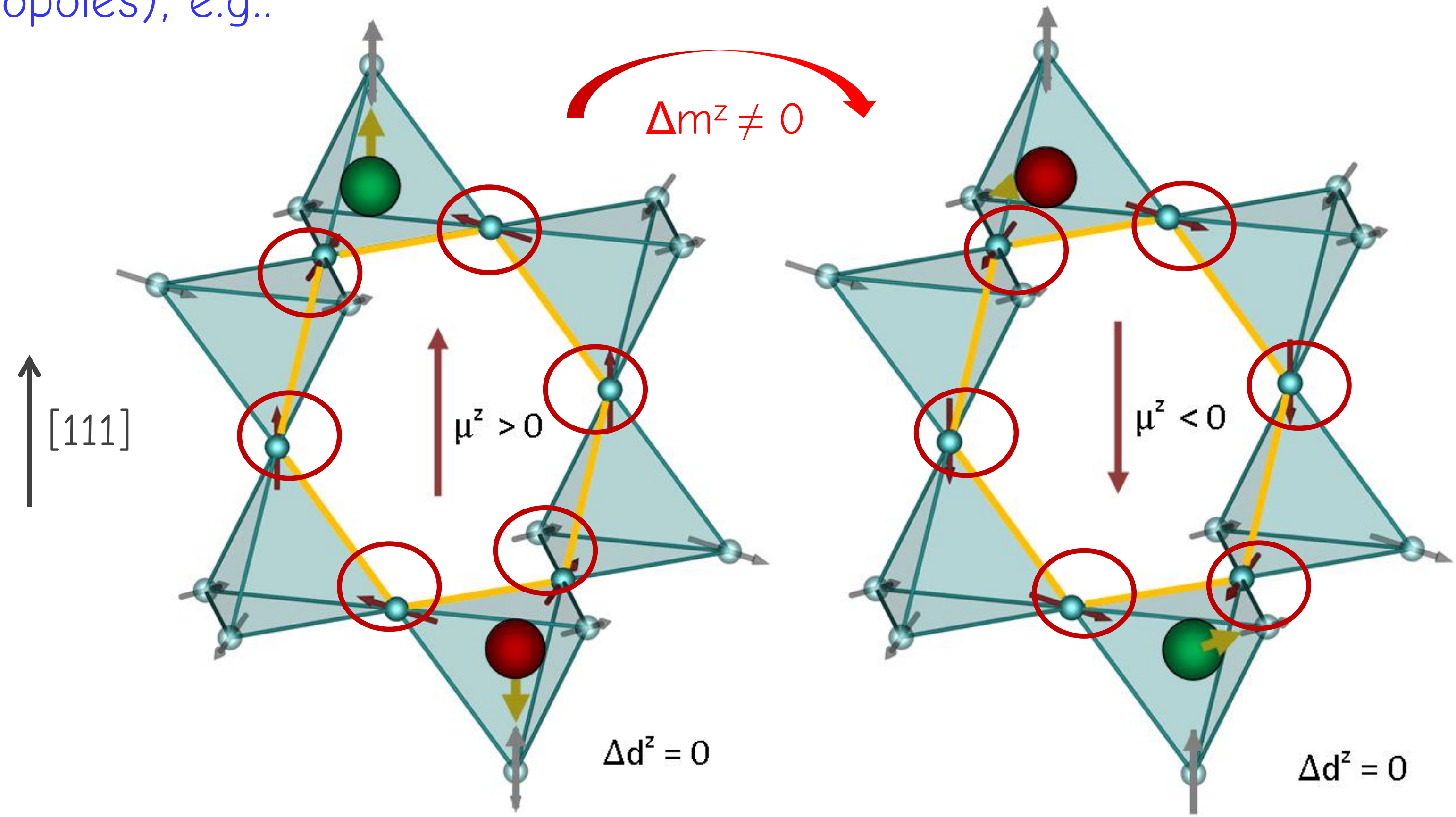
-> change of el. dipole direction -> dielectric response

(-> fast process due to spin-lattice coupling)

(Adjacent monopole/anti-monopole pairs have Δd^z=0
-> not excitable via electric (microwave) electric field !)

Comparison between χ_{AC} and ε -> microscopic origin

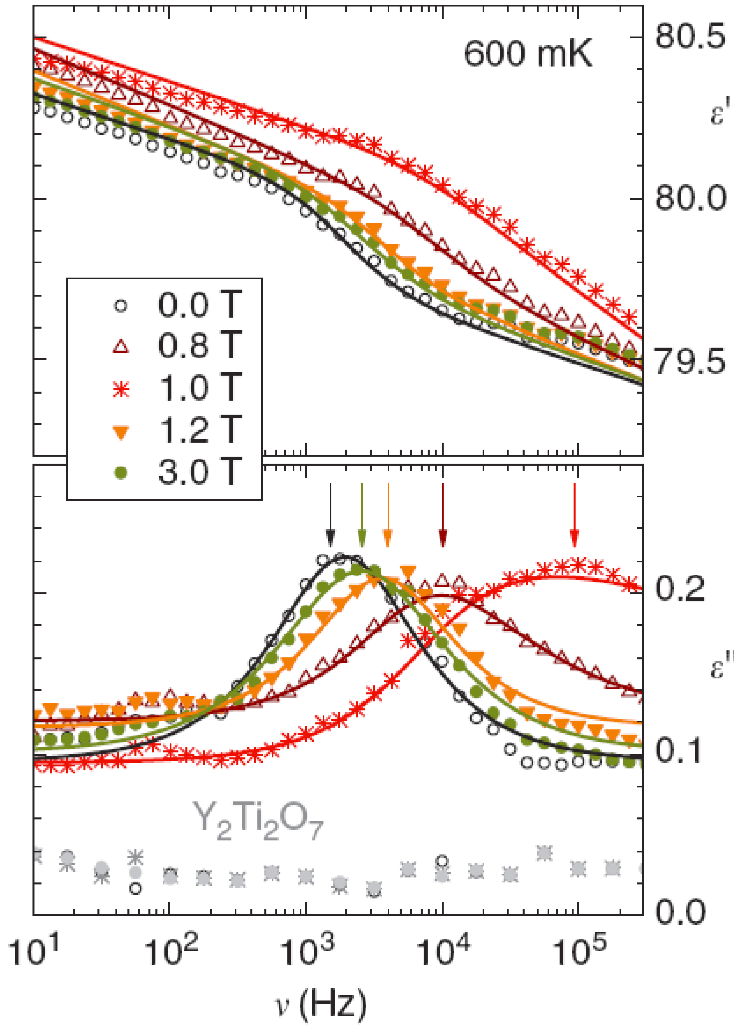
Collective spin reorientations without breaking of ice-rule (no monopoles), e.g.:

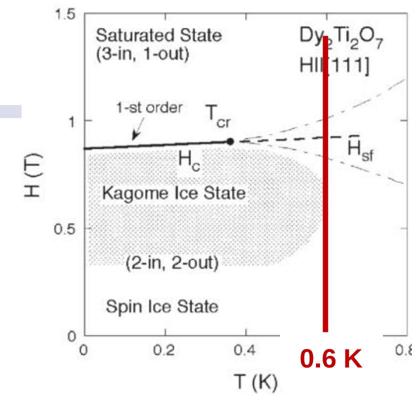


ring-like spin flips with monopole-pair -> magnetic response

monopole exchange -> no dielectric response

Dielectric response of monopole hopping





Step in **ε**' and peak in **ε**''
define relaxation time τ

$$\tau = 1/2\pi\nu_p$$

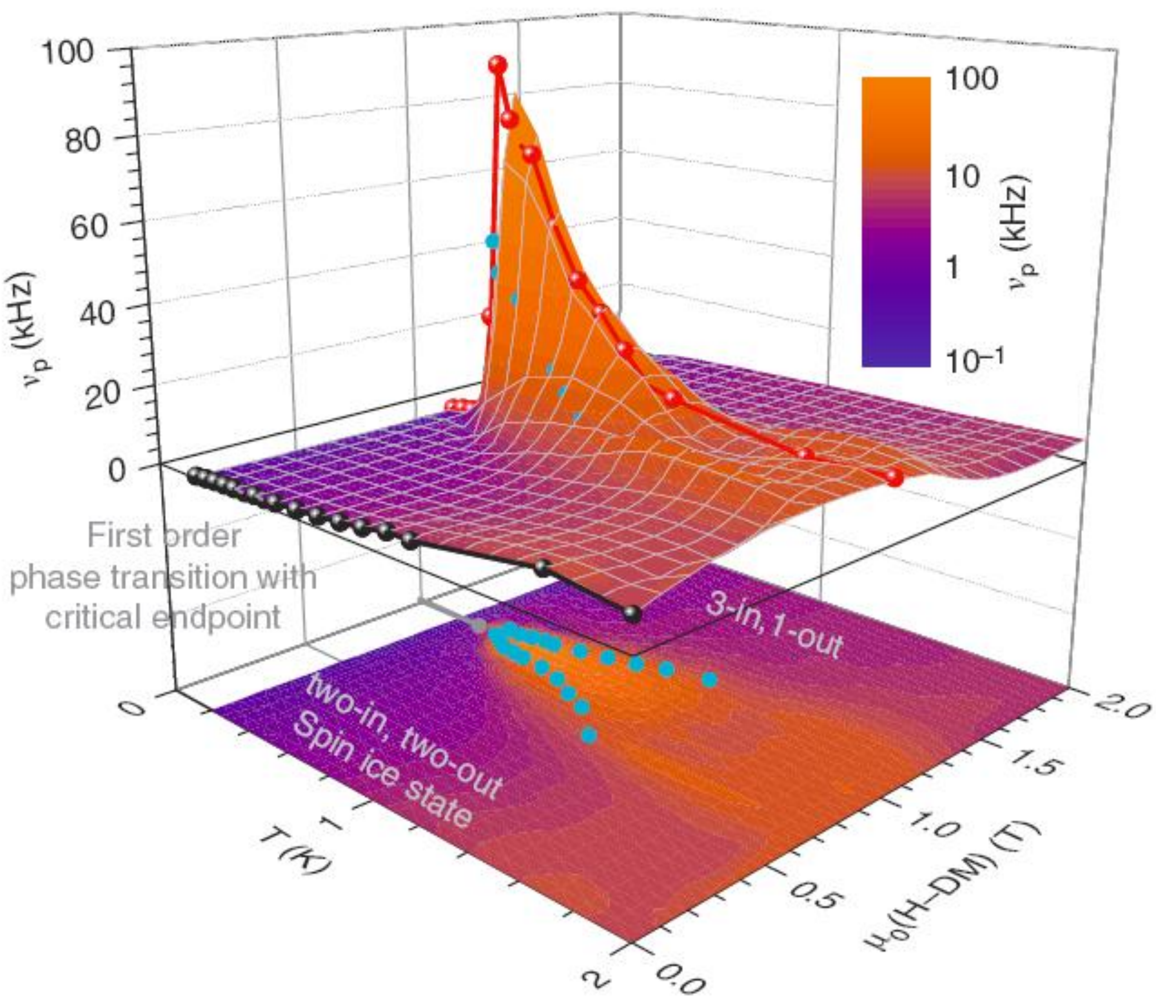
Spectra can be fitted using a Havriliak/Negami function
(Distribution of Debye-like relaxators):

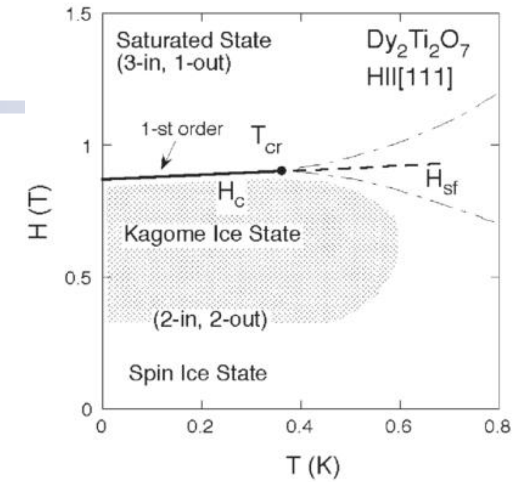
$$\varepsilon_{HN} = \varepsilon_\infty + \frac{\Delta\varepsilon}{(1+(i\omega\tau_{HN})^\beta)^\gamma}$$

-> Relaxation time shows magnetic field dependence above critical end-point !

[C.P.Grams, M. Valldor, M. Garst, J.H., Nature Commun. 5:4853 (2014)]

Monopole dynamics in $Dy_2Ti_2O_7$



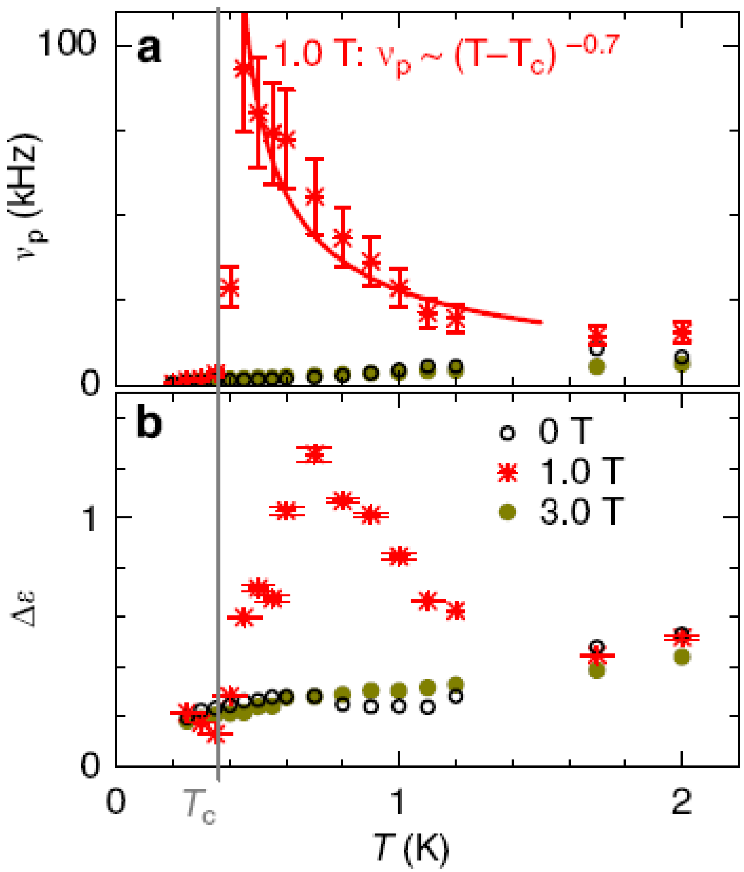


-> $\nu_p = 1/2\pi\tau$
recaptures features of
"monopole condensation"
-> slowing down outside
crossover lines

-> Critical **speeding up** above critical end-point !!!

[C.P.Grams, M. Valldor, M. Garst, J.H., Nature Commun. 5:4853 (2014)]

Relaxation time



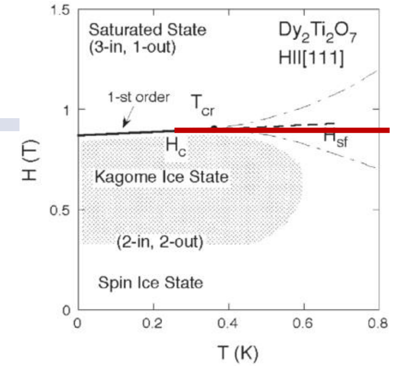
Expected behavior:

$$2\pi\nu_p = 1/\tau \sim (T - T_c)$$

(quantum-)critical slowing down
[Sachdev/Keimer, PhysicsToday 2011]

-> can only be seen at low and high magnetic fields

-> sign-change of dynamical critical exponent at crossover lines:
-> negative z !



Why?
- coherent monopole movement ?
-> reduction of damping
reduction of τ

Relaxation strength:

-> decrease with temperature due to reduction of monopole density

-> increase above due to clustering/ spatial fluctuations

$$\boldsymbol{\Delta\varepsilon} \sim np^2$$

... **qualitatively reproduced by Monte-Carlo Simulations**
[J. Attig, M. Garst, S. Trebst]
[Takatsu et al. , JPSJ 82, 073707 (2013)]

Again: Analogy to supercritical fluids

VOLUME 65, NUMBER 21 PHYSICAL REVIEW LETTERS 19 NOVEMBER 1990

Critical Speeding Up Observed

-> "piston effect"
(like second sound)

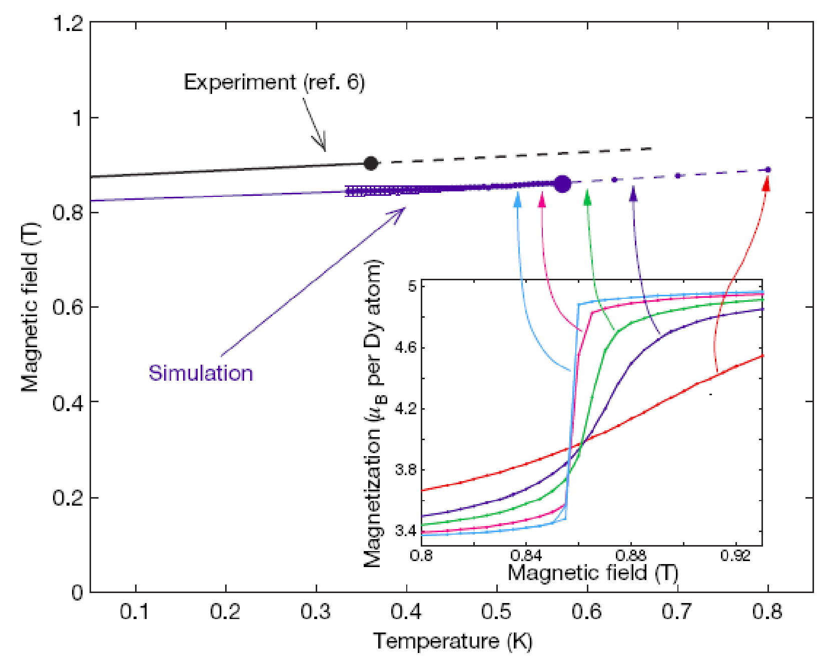
Hacène Boukari, Matthew E. Briggs, J. N. Shaumeyer, and Robert W. Gammon
Institute for Physical Science and Technology, University of Maryland, College Park, Maryland 20742
(Received 3 August 1990)

The extreme compressibility of a fluid near its liquid-vapor critical point significantly alters its dynamic response to temperature changes. Adiabatic processes allow a fluid of constant volume to thermalize in a matter of seconds, compared with the hours or days that thermal diffusion would require. Moreover,

Fluid:
-> pressure
-> compressibility

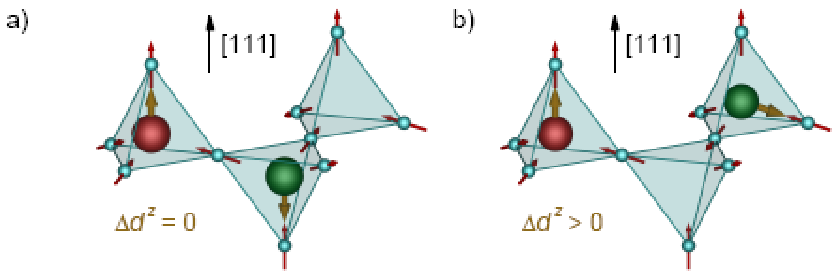
?

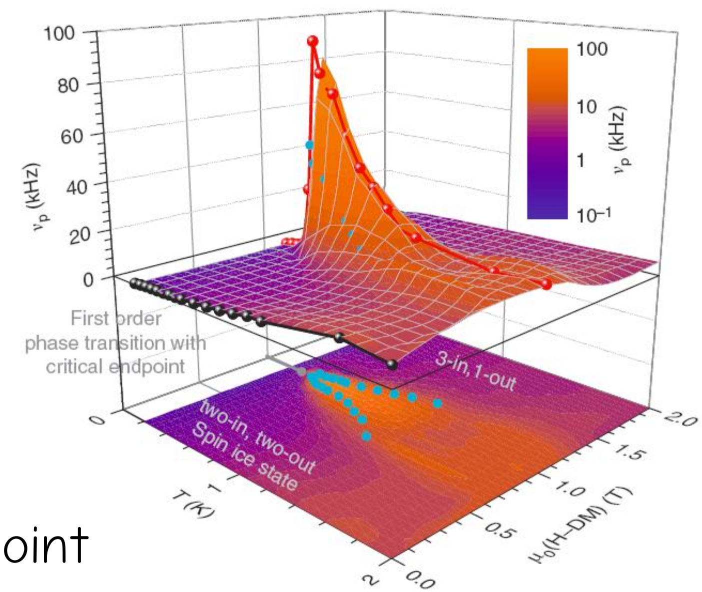
Spin-Ice:
-> magnetic field
-> susceptibility



Castelnovo, Moessner, Sondhi, ***Nature***, **451**, 42 (2008)

Summary: Critical Dynamics of Complex Order





- Spin-Ice $Dy_2Ti_2O_7$

Magnetoelectric coupling of
magnetic monopoles to **electric dipoles**
Critical speeding-up above the critical end-point
of monopole condensation

[Nature Commun. 5:4853 (2014)]

What else ? (Outlook)

Monopoles in
Quantum Spin-Ice e.g. $Ho_2Ti_2O_7$, $Yb_2Ti_2O_7$, $Pr_2Zr_2O_7$ ...