The AC Wien effect:
non-linear non-equilibrium susceptibility of spin ice

P.C.W. Holdsworth
Ecole Normale Supérieure de Lyon

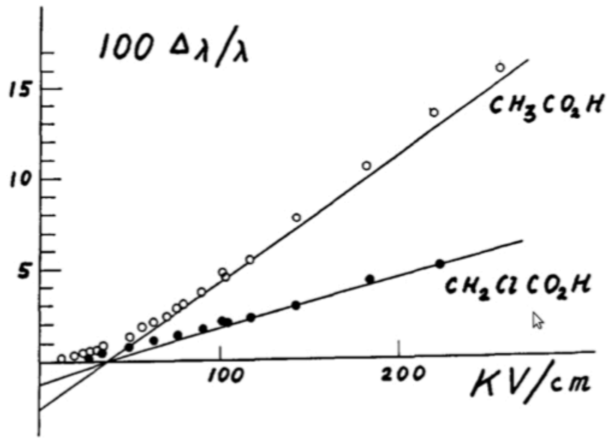
1. **The Wien effect**
2. **The dumbbell model of spin ice.**
3. **The Wien effect in a magnetic Coulomb gas**

Vojtech Kaiser, Steven Bramwell, Roderich Moessner,

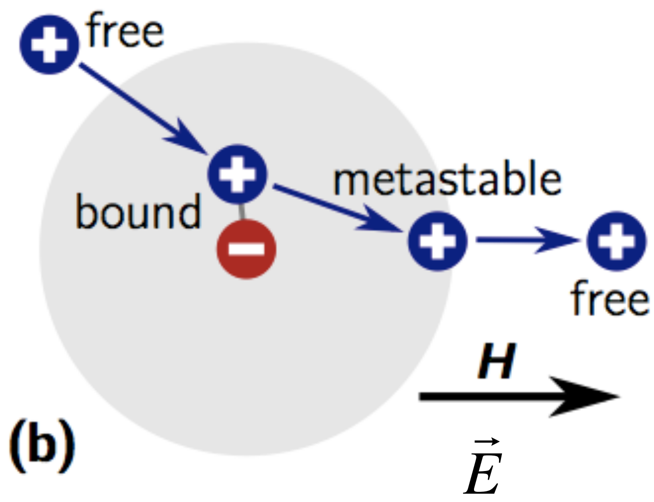
The Wien effect:

L. Onsager, "Deviations from Ohm's law in weak electrolytes". J. Chem. Phys. 2, 599,615 (1934)

Non-Ohmic conduction in low density charged fluids



Acetic acid

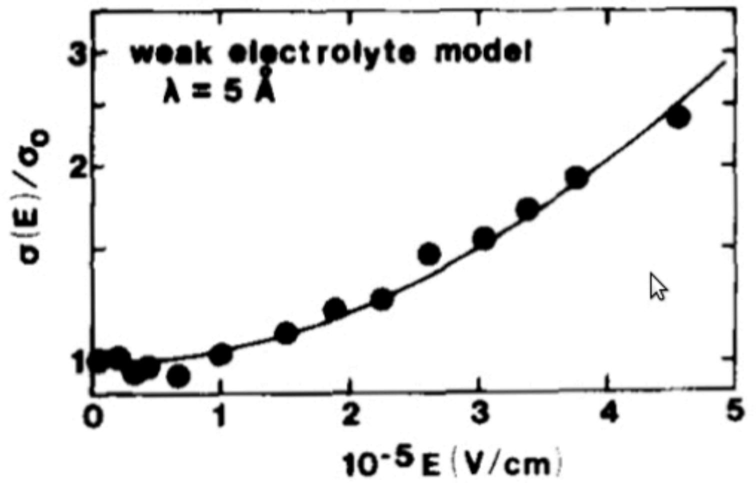


$$\boxed{n = n_f + n_b}$$

Shift in chemical equilibrium:

$$\text{charge source} \leftrightharpoons \text{bound pairs } (n_\mathrm{b}) \overset{K_\mathrm{A}(E)}{\leftrightharpoons} \text{free charges } (n_\mathrm{f})$$

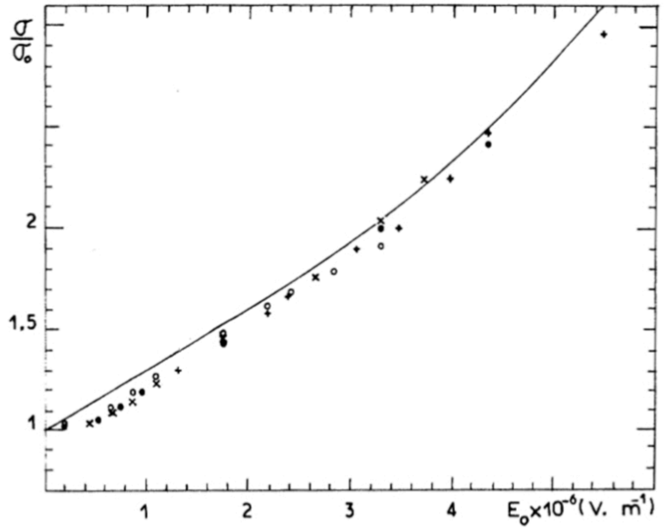
Ion-hole conduction



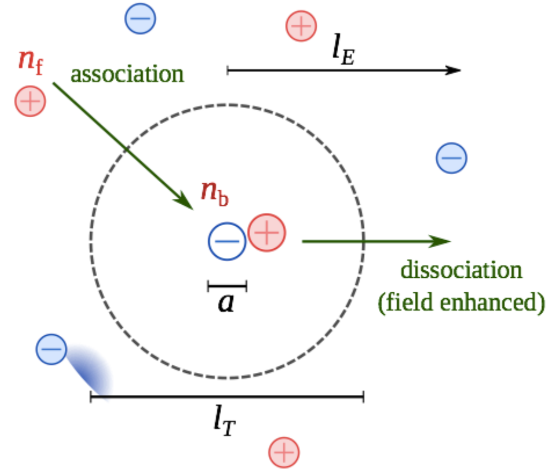
NaCaSiO glass

Example:
AOT in cyclohexane

Randriamalala et al. *IEEE TEI* (1985).



Length scales



Three length scales appear naturally:

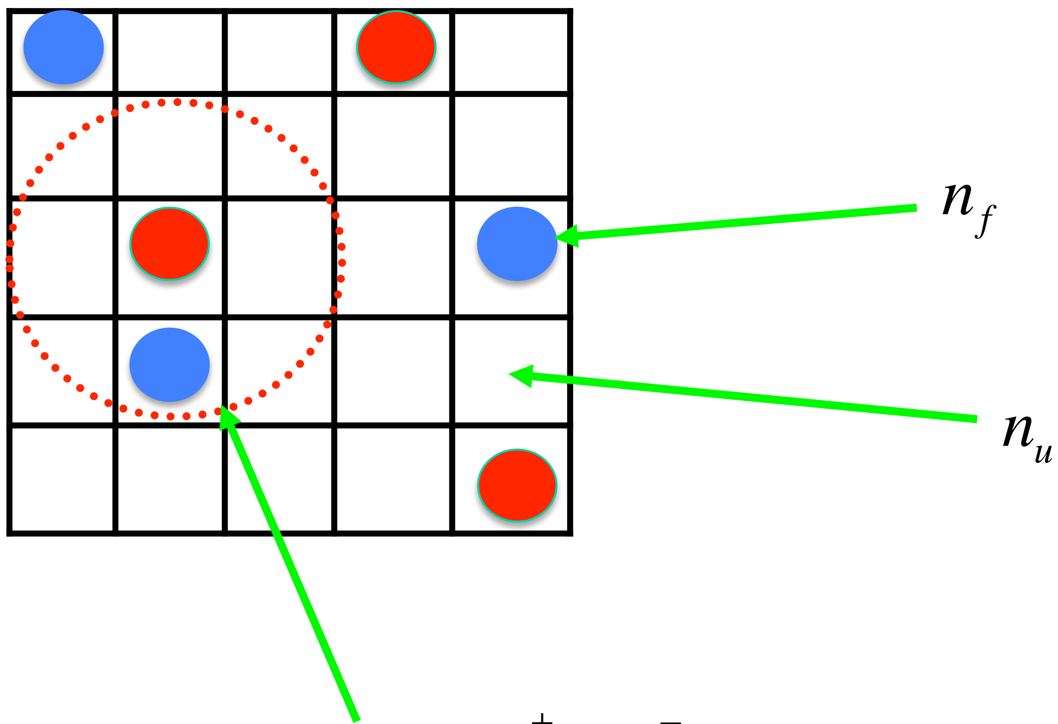
The Bjerrum length : $$l_T = \frac{q^2}{8\pi\varepsilon_0 k_B T}$$

Particles separated by $r < l_T$ are bound

Field drift length: $$l_E = \frac{k_B T}{qE}$$

Debye screening length $$l_D = \left(\frac{2\pi\varepsilon_0 k_B T a^3}{q^2 n_f}\right)^{1/2}$$

Lattice Coulomb gas:



Three species - bound particles, $n_b = n_b^+ + n_b^-$

- free particles $n_f = n_f^+ + n_f^-$

- unoccupied sites n_u

The Wien effect

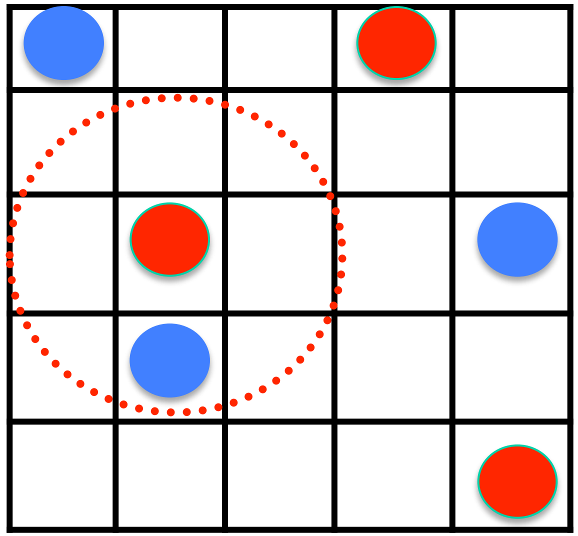
L. Onsager, "Deviations from Ohm's law in weak electrolytes". J. Chem. Phys. 2, 599,615 (1934)

$$n_b = n_b^+ + n_b^- \qquad n_f = n_f^+ + n_f^- \qquad n_u \qquad \boxed{n_u + n_b + n_f = 1}$$

$$\boxed{[n_u] \Leftrightarrow [n_b^+, n_b^-] \Leftrightarrow [n_f^+] + [n_f^-]}$$

$$\boxed{\frac{dn_f}{dt} = k^{\Rightarrow} n_b - k^{\Leftarrow} n_f^2 = 0}$$

$$K = \frac{k^{\Rightarrow}}{k^{\Leftarrow}} = \frac{n_f^2}{n_b}$$



The Wien effect

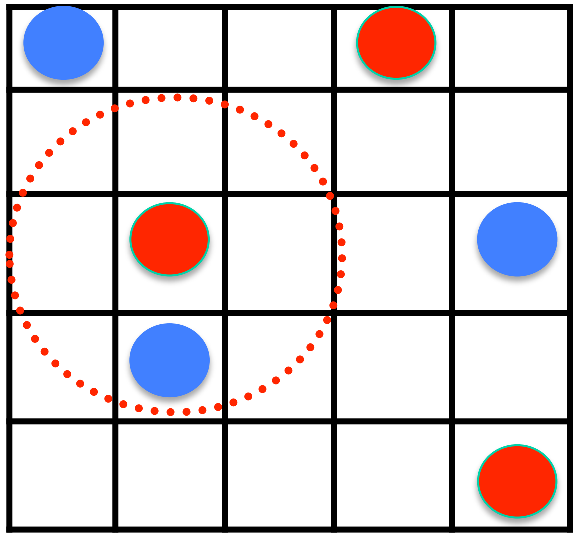
L. Onsager, "Deviations from Ohm's law in weak electrolytes". J. Chem. Phys. 2, 599,615 (1934)

$$n_b = n_b^+ + n_b^- \qquad n_f = n_f^+ + n_f^- \qquad n_u \qquad \boxed{n_u + n_b + n_f = 1}$$

$$\boxed{[n_u] \Leftrightarrow [n_b^+, n_b^-] \Leftrightarrow [n_f^+] + [n_f^-]}$$

$$\boxed{K_0 = \frac{k_0^{\Rightarrow}}{k_0^{\Leftarrow}} = \frac{n_b}{n_u}}$$

$$\boxed{K = \frac{k^{\Rightarrow}}{k^{\Leftarrow}} = \frac{n_f^2}{n_b}}$$



The Wien effect

L. Onsager, "Deviations from Ohm's law in weak electrolytes". J. Chem. Phys. 2, 599,615 (1934)

$$\boxed{K_0(E) = \frac{k_0^{\Rightarrow}}{k_0^{\Leftarrow}} = \frac{n_b}{n_u} \approx K_0(0)}$$

$$\boxed{K = \frac{k^{\Rightarrow}}{k^{\Leftarrow}} = \frac{n_f^2}{n_b} \approx K(0) + O(E)}$$

$$\boxed{\frac{K(E)}{K(0)} = \frac{I_2(2\sqrt{b})}{\sqrt{2b}} = 1 + b + O(b^2)}$$

for $l_D >> l_E, l_T$

$$\boxed{b = \frac{l_T}{l_E} \propto \frac{q^3 E}{T^2}}$$

Linear in $|\vec{E}|$ For small field

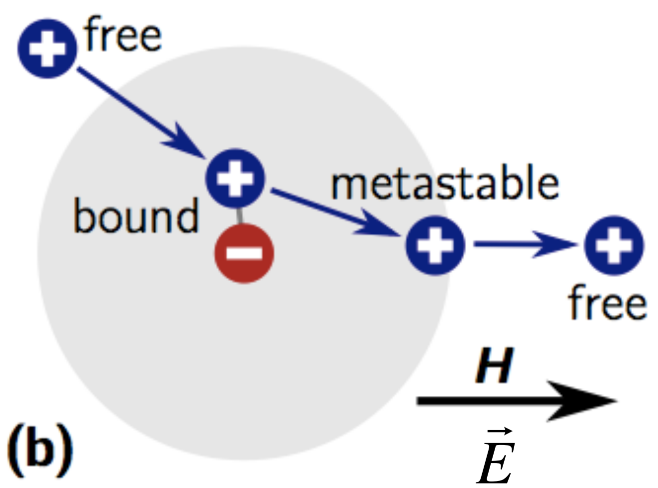
The Wien effect

L. Onsager, "Deviations from Ohm's law in weak electrolytes". J. Chem. Phys. 2, 599,615 (1934)

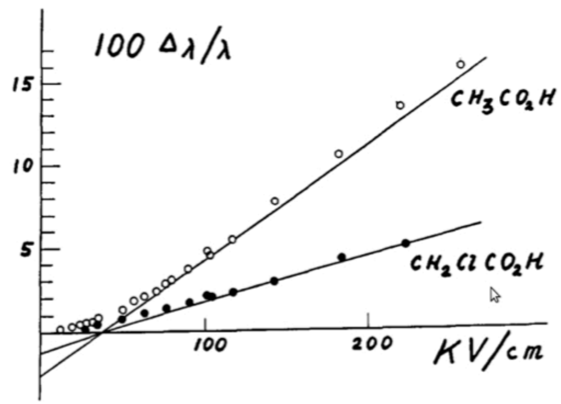
$$\boxed{n = n_f + n_b}$$

$$\boxed{\frac{n_f(E)}{n_f(0)} \approx \sqrt{\frac{I_2(2\sqrt{b})}{\sqrt{2b}}} = 1 + \frac{b}{2} + O(b^2)}$$

$$\boxed{b \propto \frac{q^3 E}{T^2}}$$



$\vec{E}$



Acetic acid

Linear in $|\vec{E}|$ For small field – this is a non-equilibrium effect

The linear field dependence => A non-equilibrium effect
=> Compare with Blume-Capel paramagnet.

$$\boxed{\mathrm{H} = -H\sum_i S_i + \Delta\sum\left(S_i\right)^2,\ S_i = 0, \pm 1}$$

$$\boxed{n(0) = n_\uparrow + n_\downarrow = \frac{2\exp(-\beta\Delta)}{1+2\exp(-\beta\Delta)}}$$

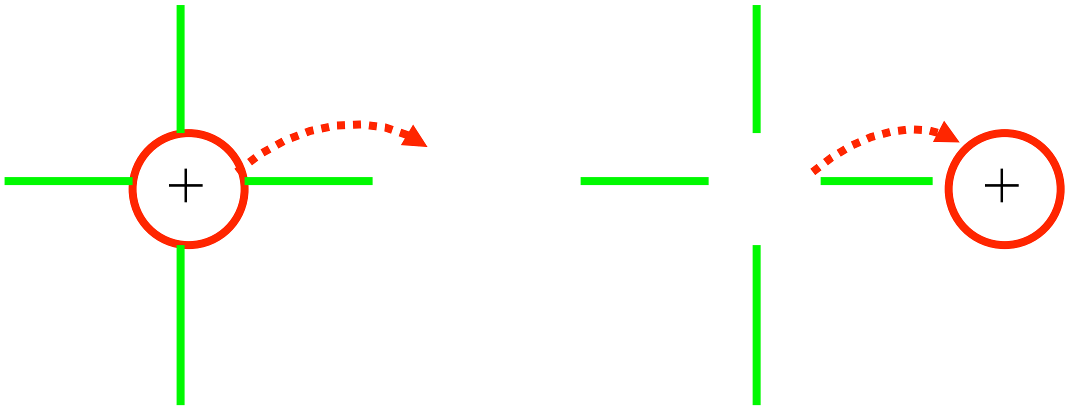
$$\boxed{\begin{aligned} n(H) &= n_\uparrow(H) + n_\downarrow(H) = \frac{n(0)}{2}(\exp(\beta H) + \exp(-\beta H)) \\ &= n(0) + O(H^2) \end{aligned}}$$

This scalar quantity changes quadratically with applied field

Lattice Electrolyte - Coulomb gase

1. Electrolyte

$\boxed{\vec{E}}$



Hopping on a diamond lattice

A grand canonical Coulomb gas.

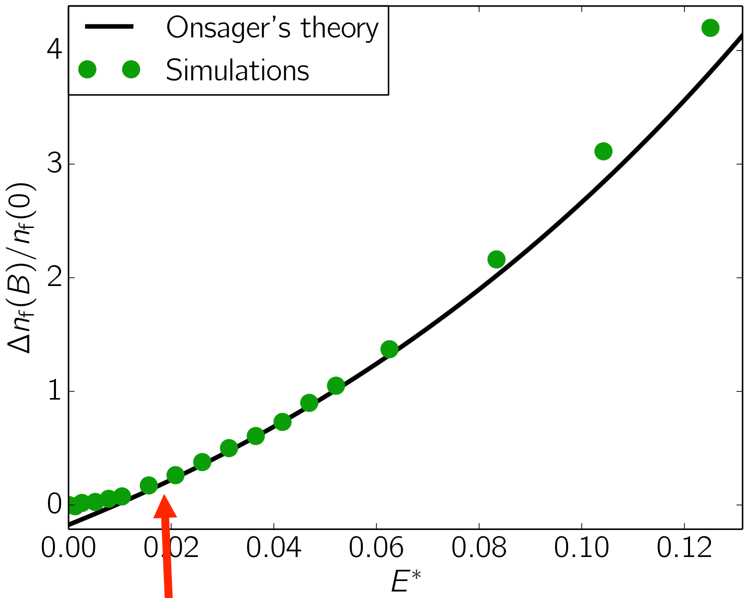
$$\boxed{H \approx \sum_{i>j} U(r_{ij}) - \mu \hat{N}}$$

Weak electrolyte limit: $\boxed{|\mu| > k_B T}$ $\boxed{n = \frac{N}{N_0} << k_B T}$

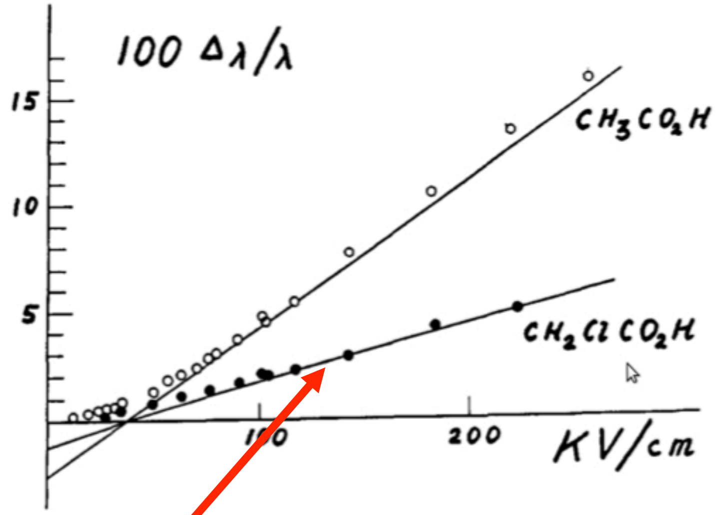
Results:

Kaiser, Bramwell, PCWH, Moessner, Nature Materials, **12**, 1033-1037, (2013)

- Parameters: T^*, μ^*, E^*.
- Reduced by Coulomb energy at contact: $q^2/4\pi\epsilon a$.



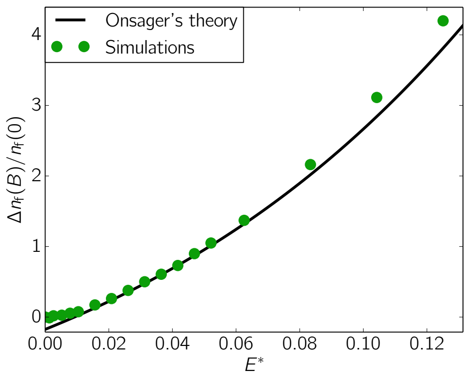
Lattice Electrolyte

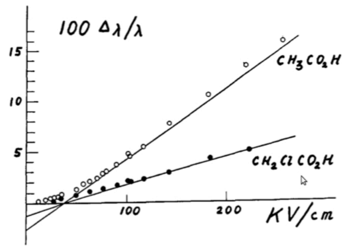


Acetic acid

Linear in $\left|\vec{E}\right|$ to lowest order

Linear term is renormalized away by Debye screening:

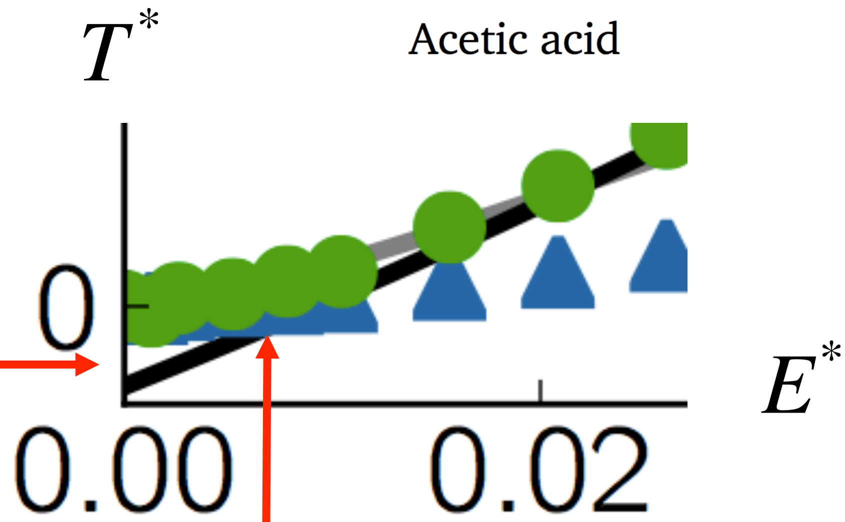




Acetic acid

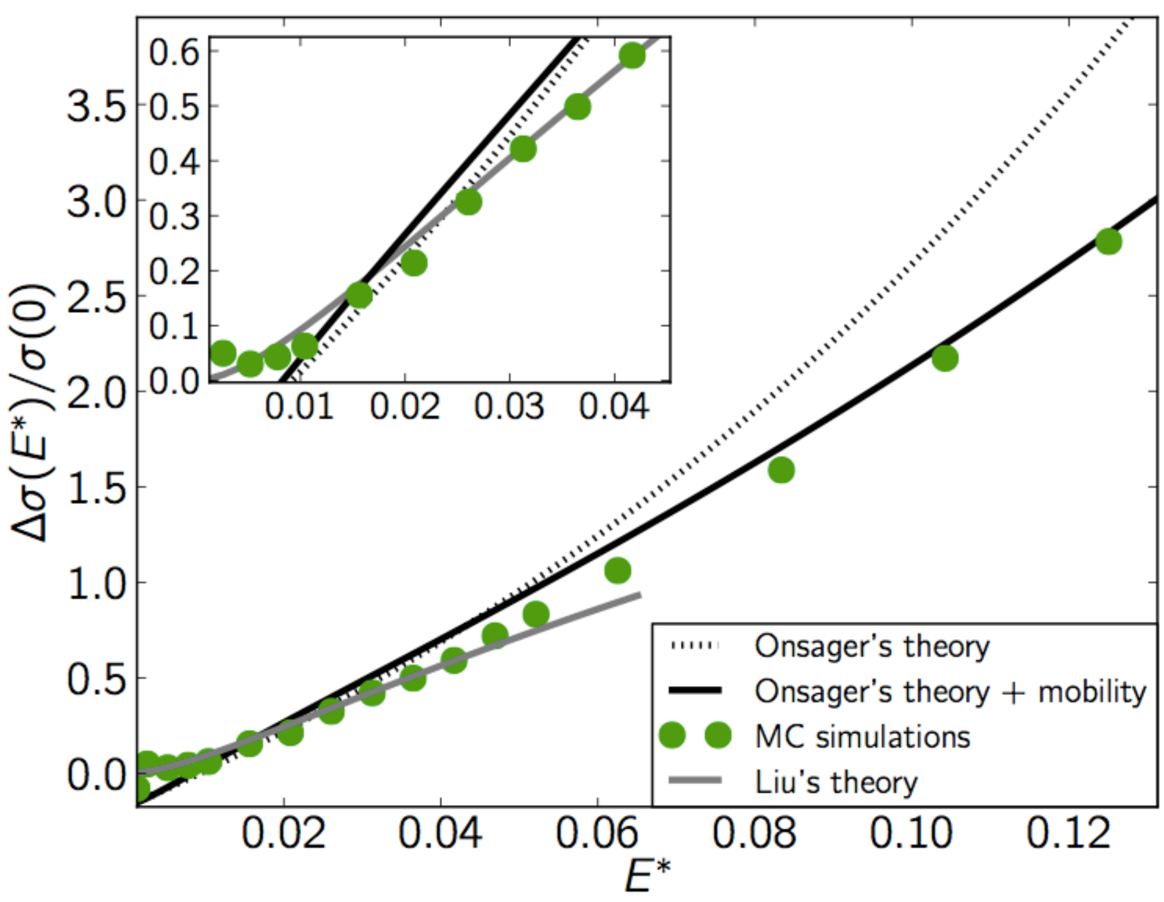
Negative offset

$$\frac{\Delta n_f}{n_f} = -(1-\gamma\ \ )$$



Crossover $\quad \ell_E > \ell_D$

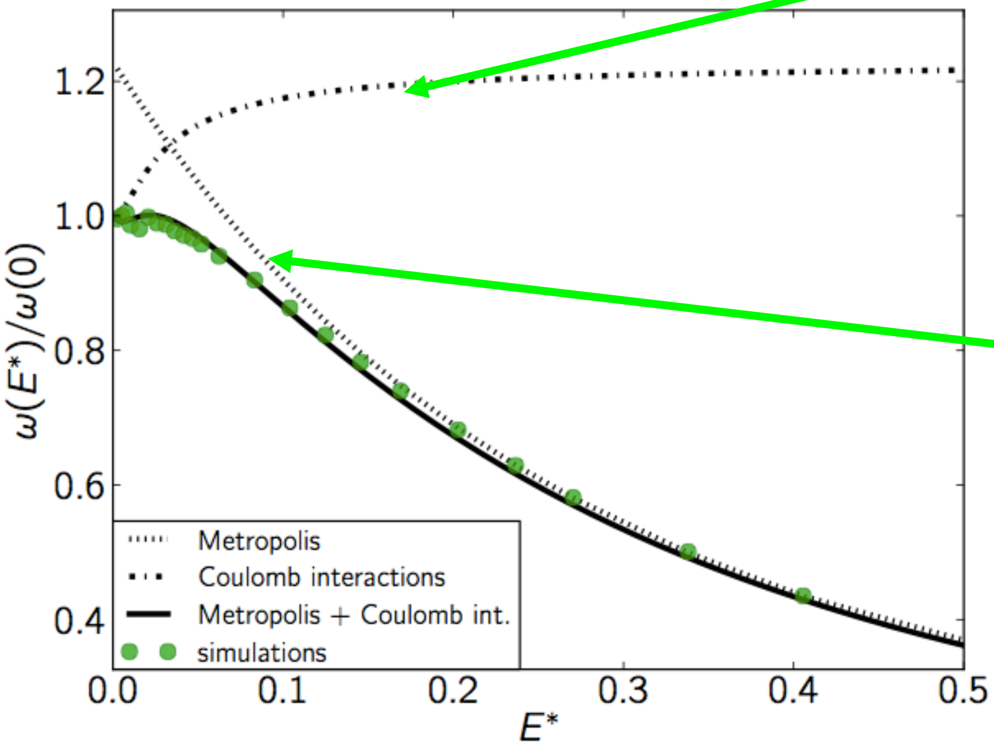
Relative conductivity falls below prediction



$$\boxed{\sigma = q^2 \kappa n_f}$$

Theory relies on mobility, κ being field independent

Field dependent mobility:



Blowing away of Debye screening cloud (1^{st} Wien effect)

Velocity max for Metropolis

$$\frac{\omega(E^*)}{\omega(0)} = \frac{1 - h\left(\ell_T, \ell_D\right) g\left(\ell_D/\ell_E\right)}{1 - h\left(\ell_T, \ell_D\right)} \frac{1 - \exp(-E^*/T^*)}{E^*/T^*}.$$

Fuoss-Onsager theory + Metropolis

Spin ice- a magnetic Coulomb gas

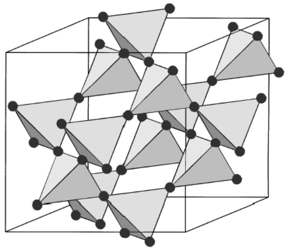


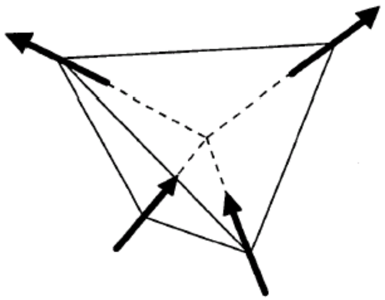
FIG. 1. The pyrochlore lattice.

Spin Ice – a dipolar magnet

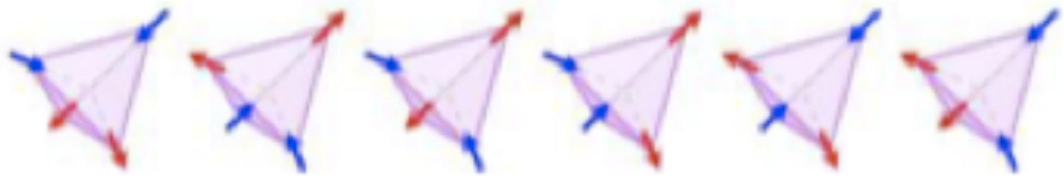
$$H = J\sum_{ij} \vec{S}_i.\vec{S}_j + D\sum_{ij}\left[\frac{\vec{S}_i.\vec{S}_j}{|\vec{r}_{ij}|^3} - \frac{3(\vec{S}_i.\vec{r}_{ij})(\vec{S}_j.\vec{r}_{ij})}{|\vec{r}_{ij}|^5}\right]$$

Long range interactions are almost but not quite screened

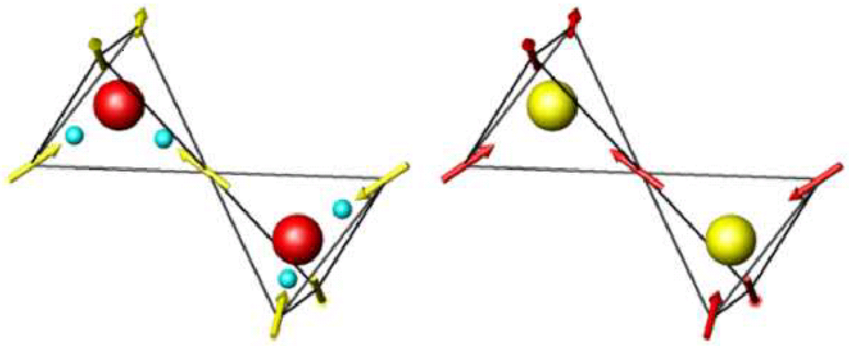
den Hertog and Gingras, PRL.84, 3430 (2000), Isakov, Moessner and Sondhi, PRL 95, 217201, 2005



Six equivalent configs for each tetrehedron

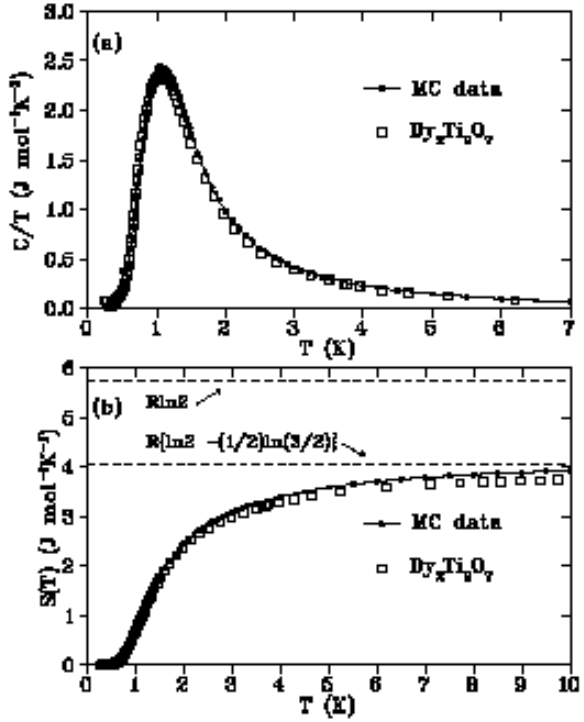


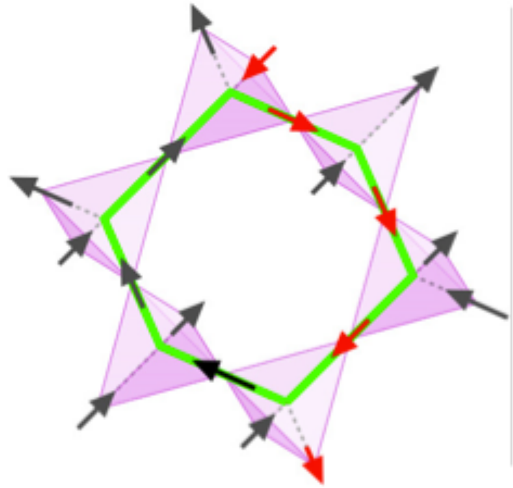
Magnetic ice rules =>
Pauling entropy.



$$\boxed{S_P = Nk_B \frac{1}{2}\ln\frac{3}{2}}$$

Magnetic
« Giauque and Stout »
experiment:





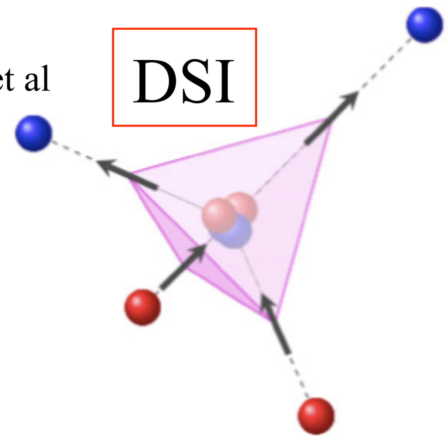
Ramirez et al, Nature
399,333, (1999)

Glassy behaviour:
Schiffer et al,
Castelnovo Moessner Sondhi,
Cugliandolo et al,
Davis et al,

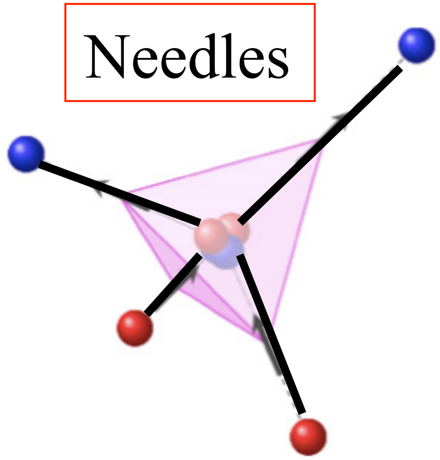
Extension of the point dipoles into magnetic needles/dumbbelles

Möller and Moessner PRL. 96, 237202, 2006, Castelnovo, Moessner, Sondhi, Nature, 451, 42, 2008

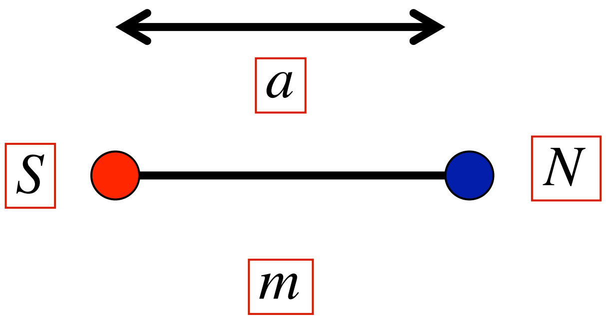
Gingras et al



$\Rightarrow$



Configurations of
magnetic charge
at tetrehedron centres



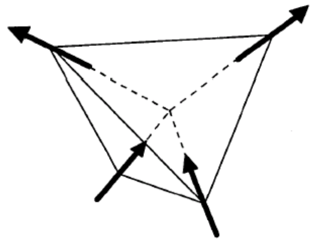
Magnetic ice rules
two-in two-out

An extensive degeneracy of states satisfy these rules – Monopole vacuum

Ice rules, topological constraints

S. V. Isakov, K. Gregor, R. Moessner, and S. L. Sondhi PRL 93, 167204, 2004

M= divergence free field

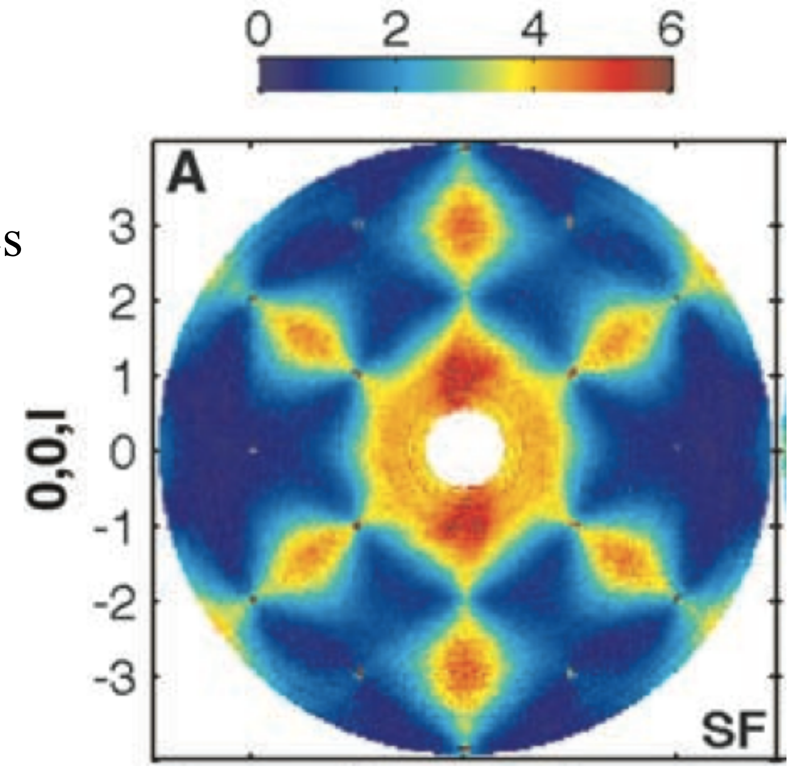


$$\vec{\nabla}.\vec{M} = 0$$

Emergent gauge field

$$\vec{M} = \vec{\nabla} \wedge \vec{A} \ = \vec{M}_d$$

Monopole vacuum has divergence free configurations - « Coulomb phase » Physics



Pinch Points:

T. Fennell *et. al*., Magnetic Coulomb Phase in the Spin Ice $Ho_7O_2Ti_2$ *Science*, 326, 415, 2009.

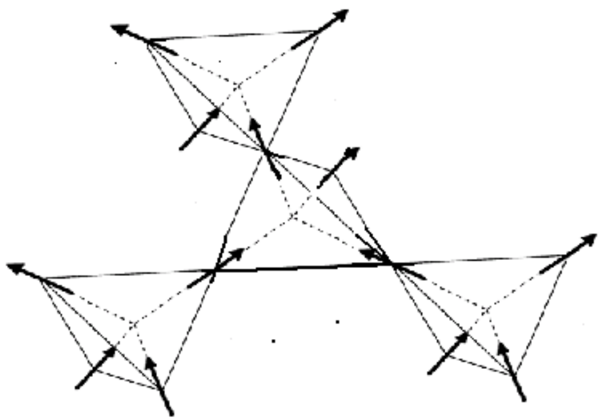
Topological sector fluctuations:

Jaubert et. al. Phys. Rev. X, 3, 011014, (2013)

Topological excitations back to paramagnetic phase space

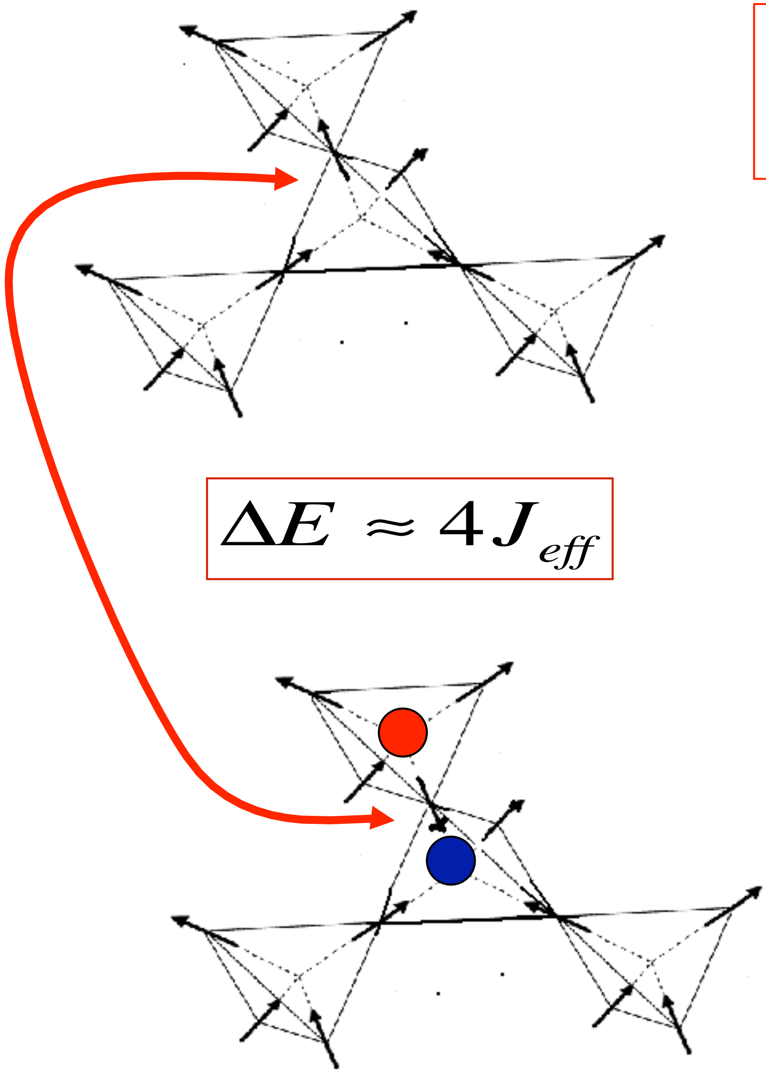
Castelnovo, Moessner, Sondhi, Nature, 451, 42, 2008 -CMS, Ryzhkin JETP, **101**, 481, 2005.

Extensive phase space of topologically constrained states = Vacuum for quasi-particle excitations



Topological constraints
Excitations back to paramagnet….

Topological constraints
Spin flip creates two defects

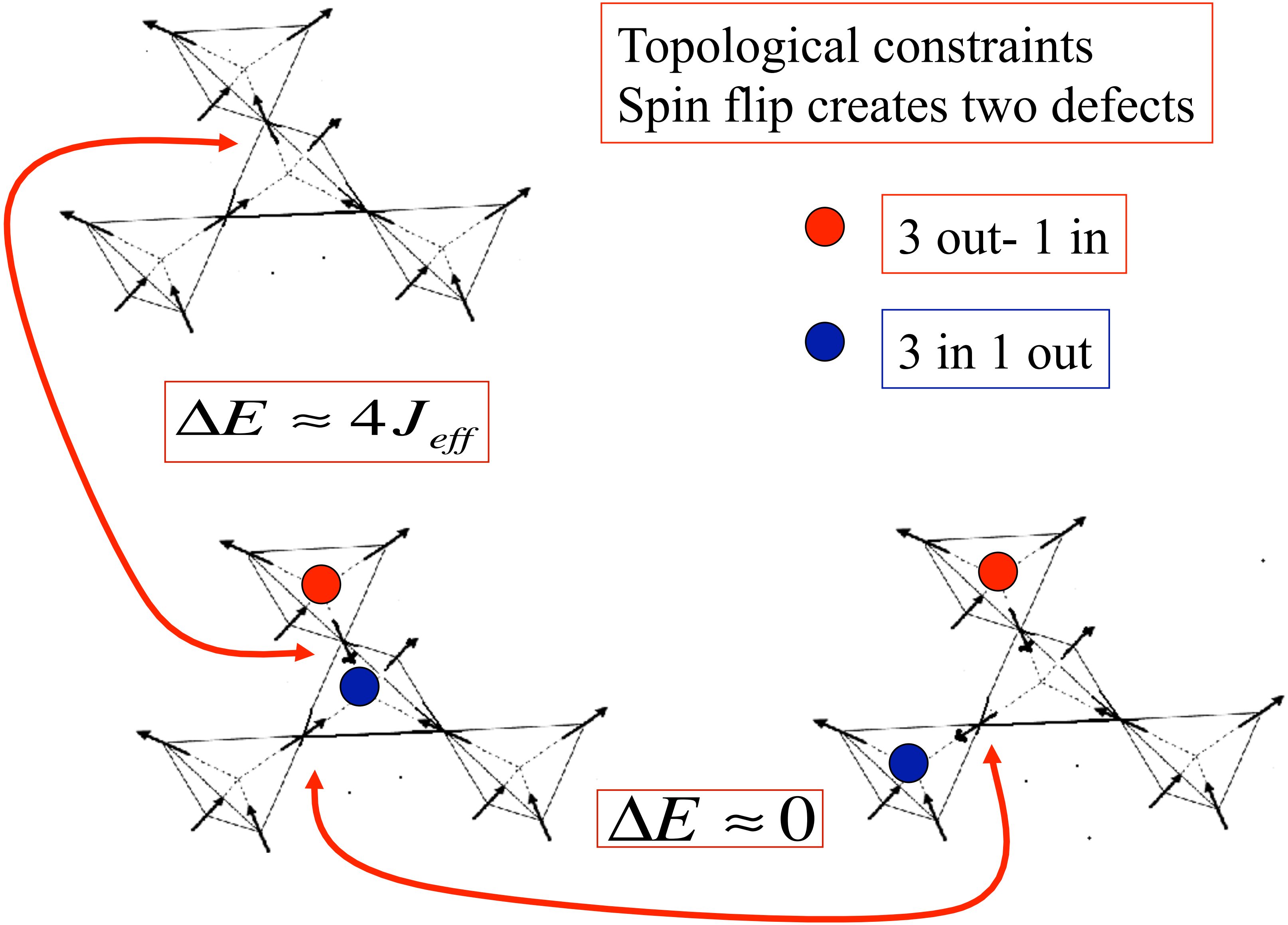


$$\boxed{\Delta E \approx 4J_{eff}}$$

3 out- 1 in

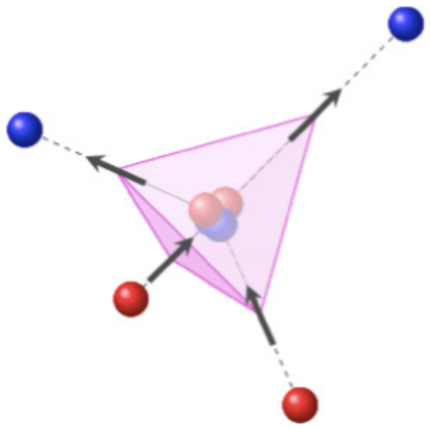
3 in 1 out

Topological constraints
Spin flip creates two defects

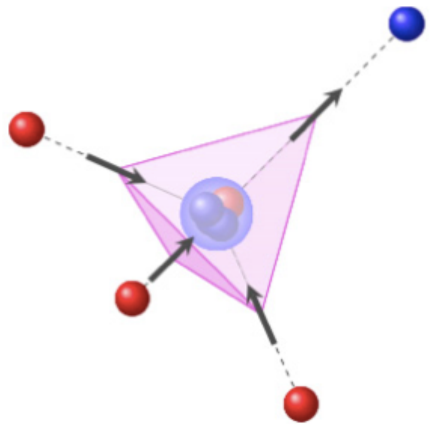


Castelnovo, Moessner, Sondhi, Nature, 451, 42, 2008 (Ryzhkin JETP, **101**, 481, 2005)

Topological defects carry magnetic charge – magnetic monopoles



$\Rightarrow$



$$\Delta M = 2m \quad \Rightarrow \quad U(r) = \frac{\mu_0}{4\pi}\frac{Q_i Q_j}{r}; \; Q_i = \pm\frac{2m}{a}$$

A grand canonical Coulomb gas of quasi particles.

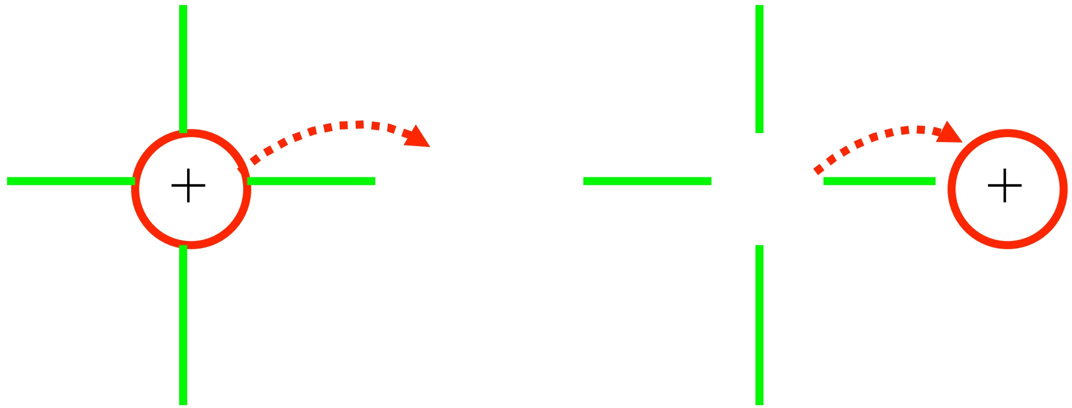
$$H \approx \sum_{i>j} U(r_{ij}) - \mu\hat{N} \qquad \mu(J,m,a)$$

In which case one should expect « electrolyte » physics + constraints - magnetolyte (Castelnovo)

Electrolyte and Magnetolyte Coulomb gases

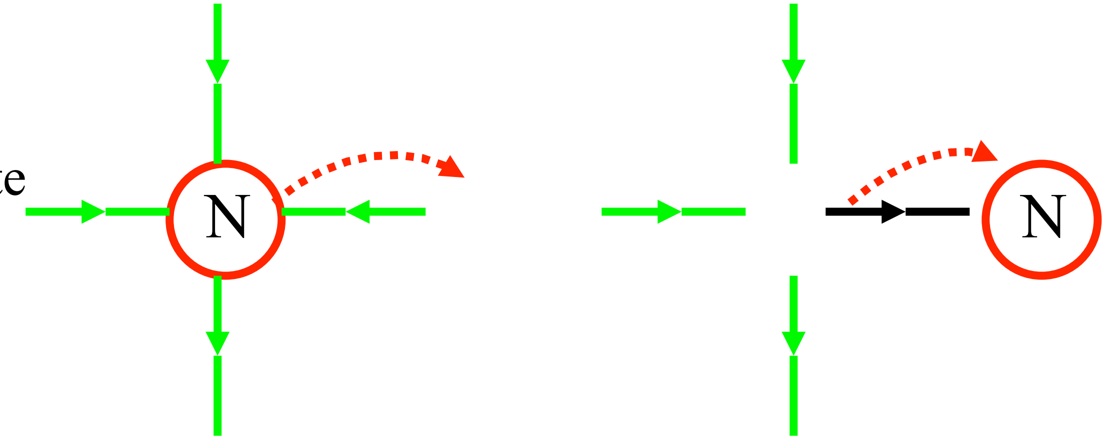
1. Electrolyte

$\vec{E}$



2. Magnetolyte

$\vec{H}$

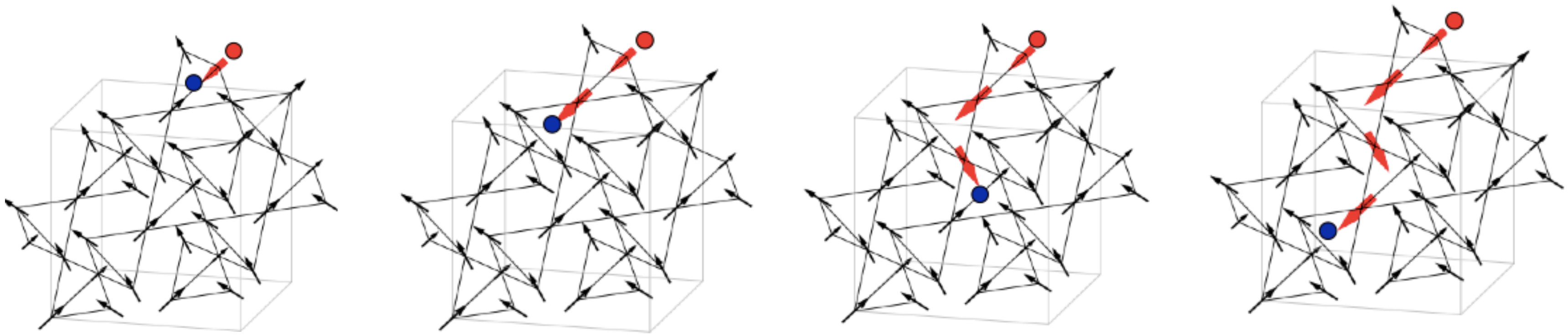


Chemical potential $\mu_1 = -4.35K$ /particle for $Dy_2Ti_2O_7$

$\mu_1 = -5.7K$ /particle for $Ho_2Ti_2O_7$

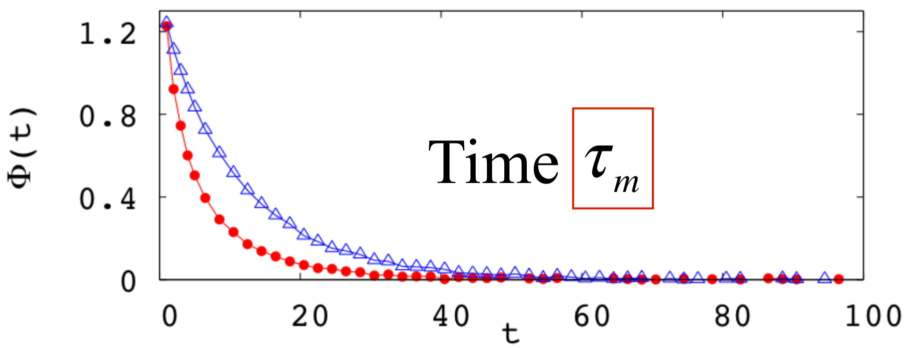
CMS, Phys. Rev. B 84, 144435, 2011, Melko, Gingras JPCM, 16 (43) R1277–R1319 (2004)

Monopole dynamics polarizes the medium



Coulomb gas physics with transient currents

Ryzhkin JETP, **101**, 481, 2005. Jaubert and Holdsworth, Nature Physics, 5, 258, 2009

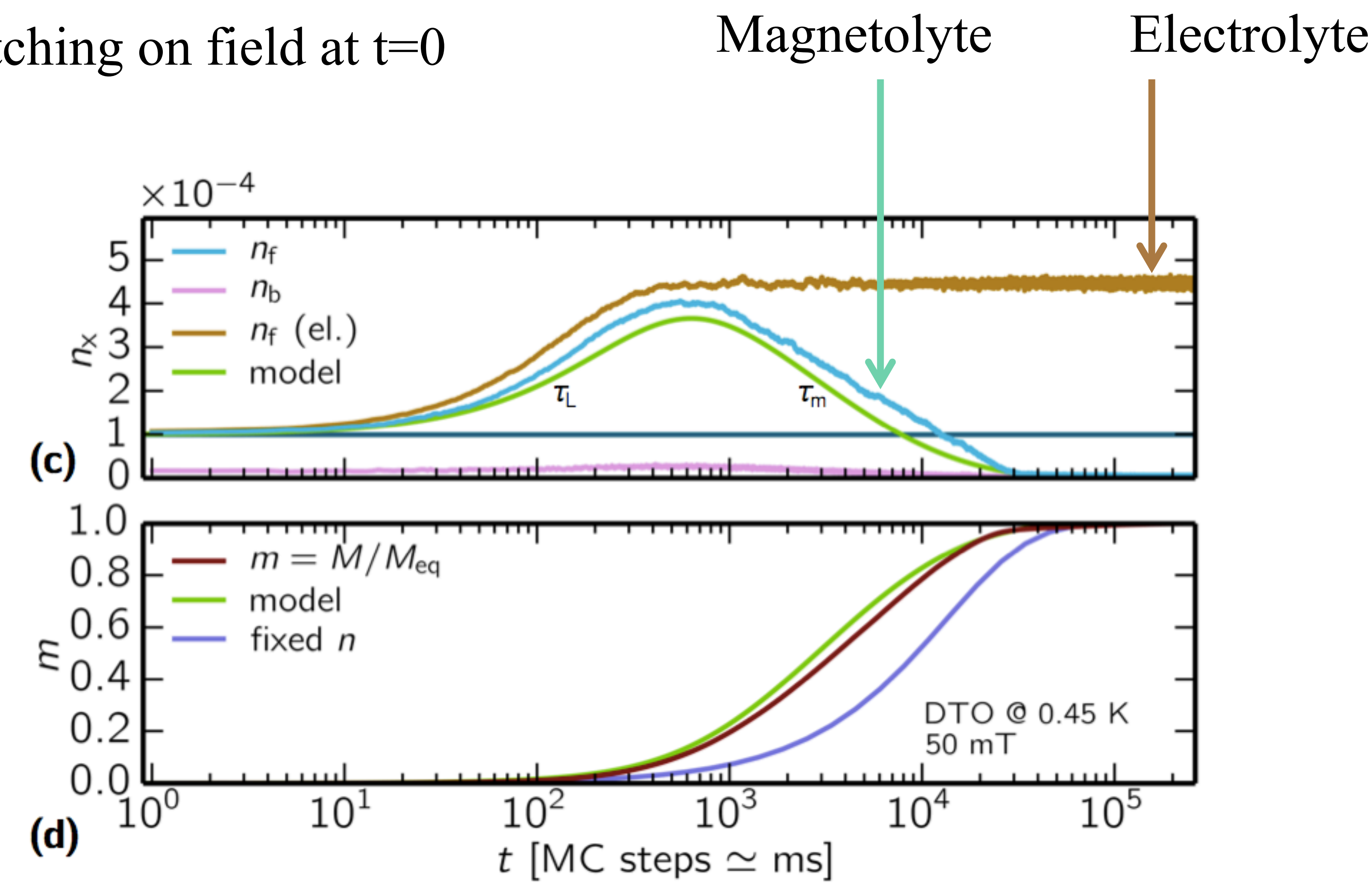


$$\boxed{\vec{j} = \frac{d\vec{M}}{dt} = \frac{1}{\tau_m}\left(\vec{H} - \frac{\vec{M}}{\chi_T}\right)}$$

Wien effect in the magnetolyte:

Kaiser et al, to appear in Phys Rev Lett.

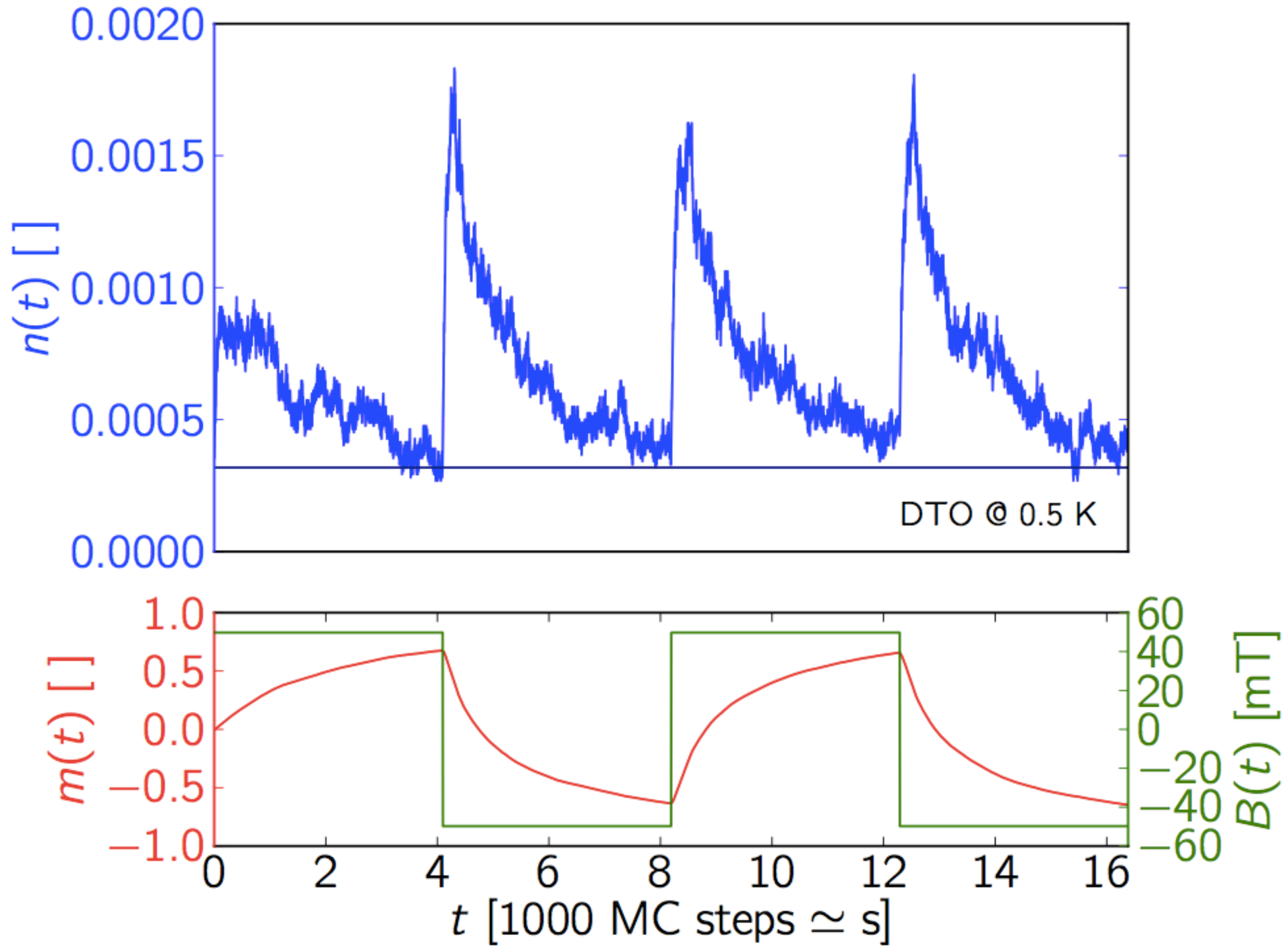
Switching on field at t=0



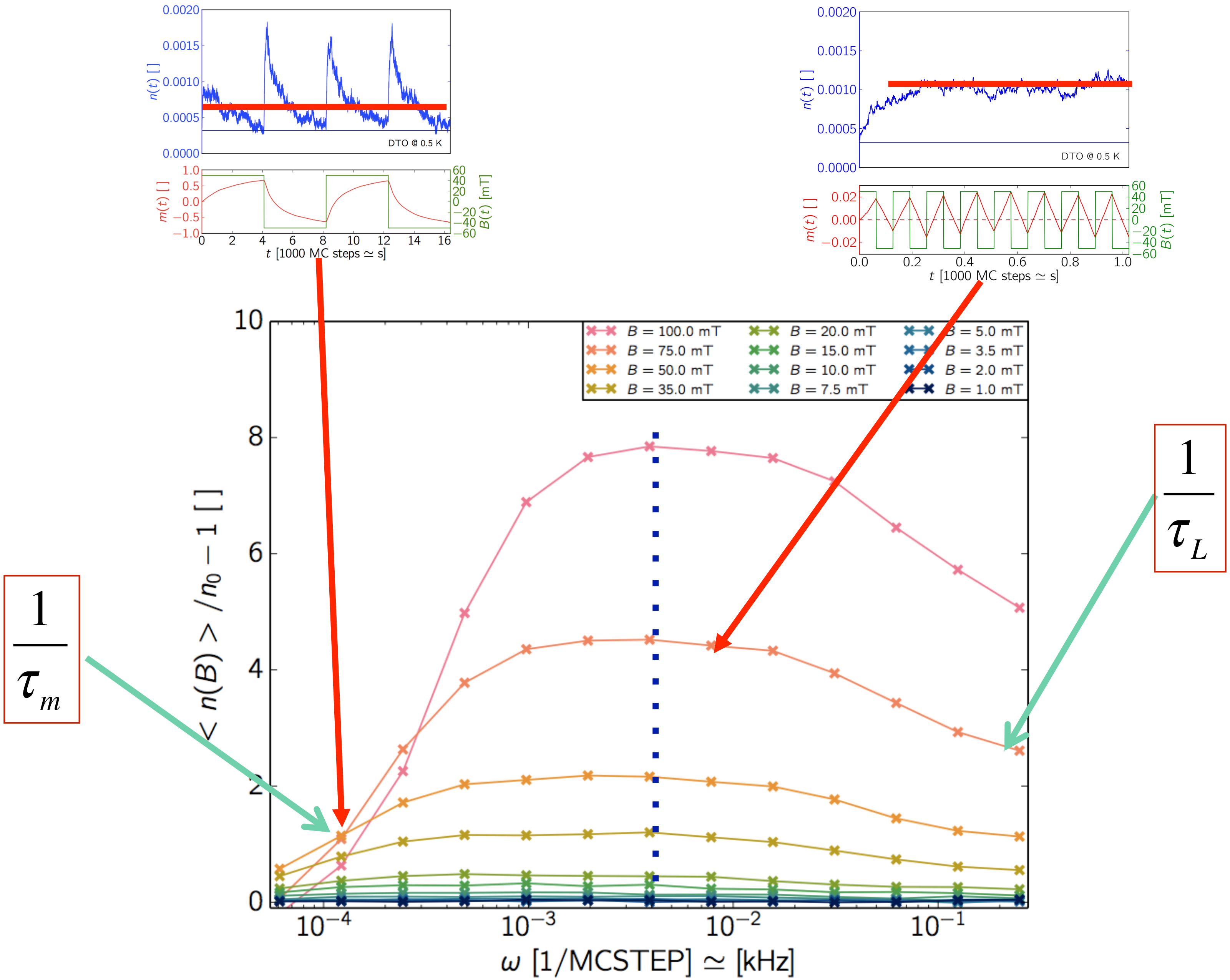
Time scale: 1 MCS = 1 ms for DTO

Jaubert and Holdsworth, Nature Phyiscs, 5, 258, 2009

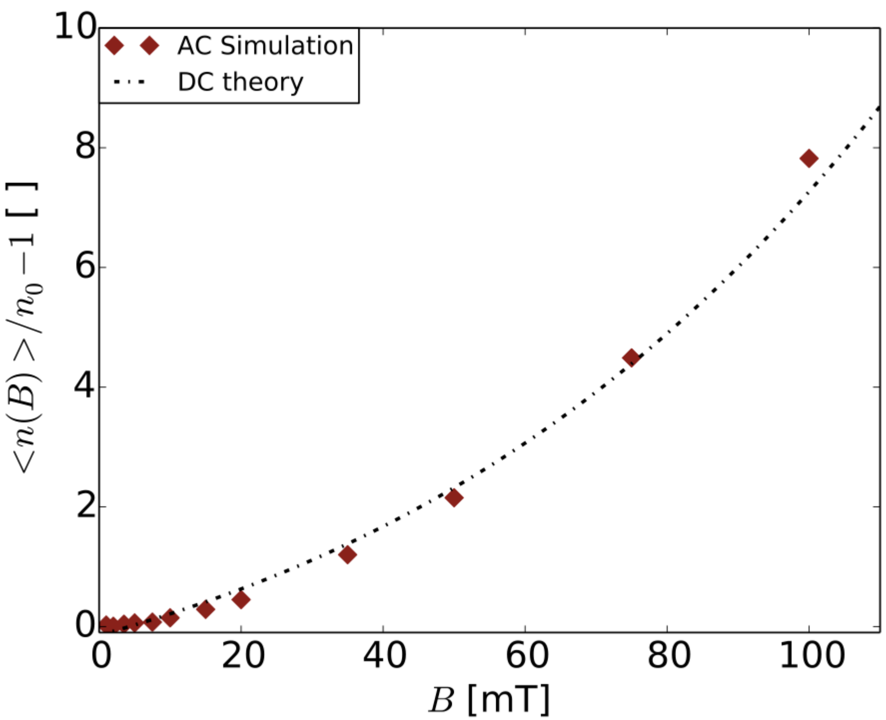
Square AC field – 8.2 secs



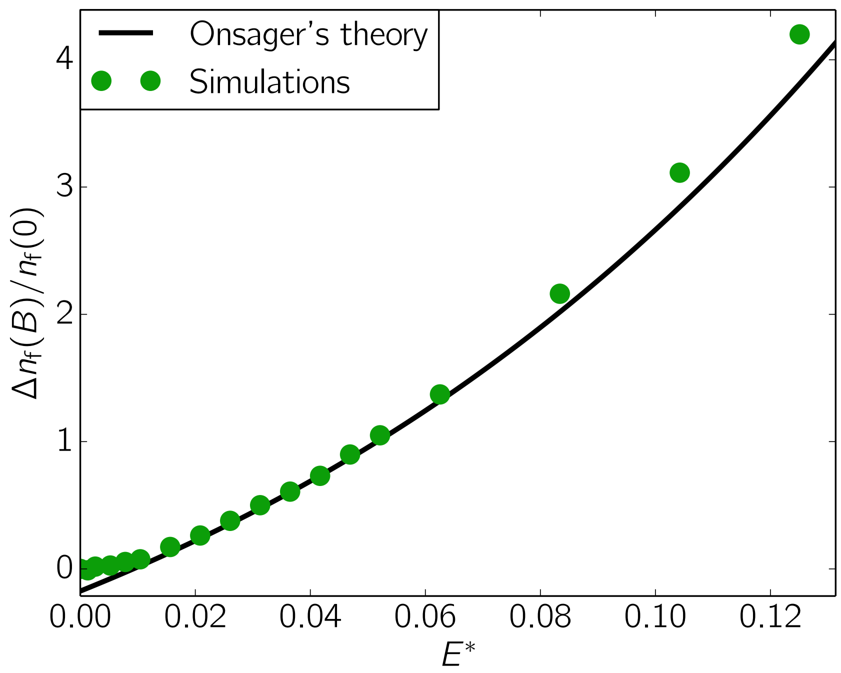
Monopole concentration with time



Magnetolyte DTO 0.5 K



Electrolyte DTO 0.43 K

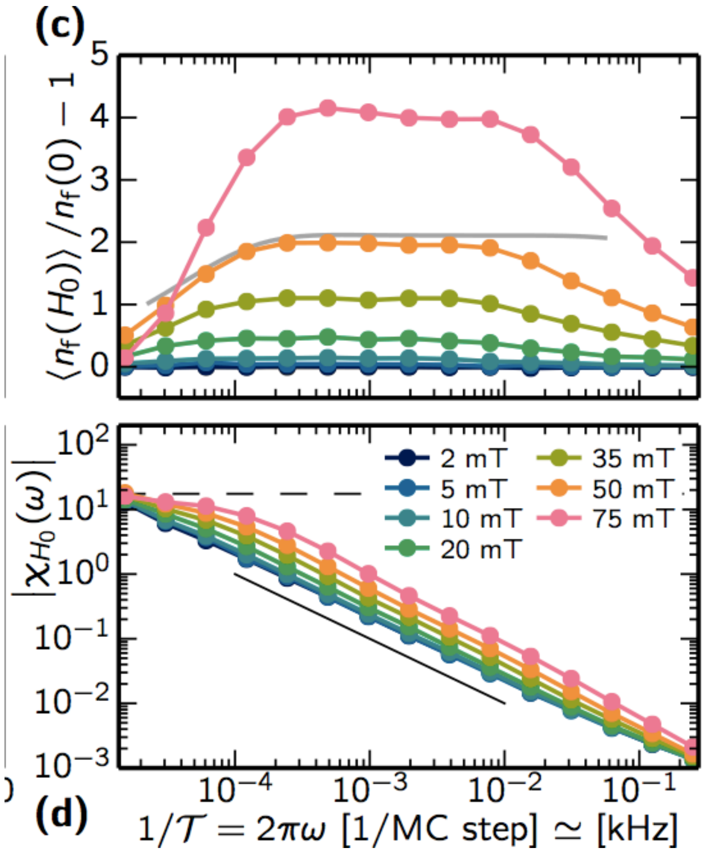


An experimental signal ?

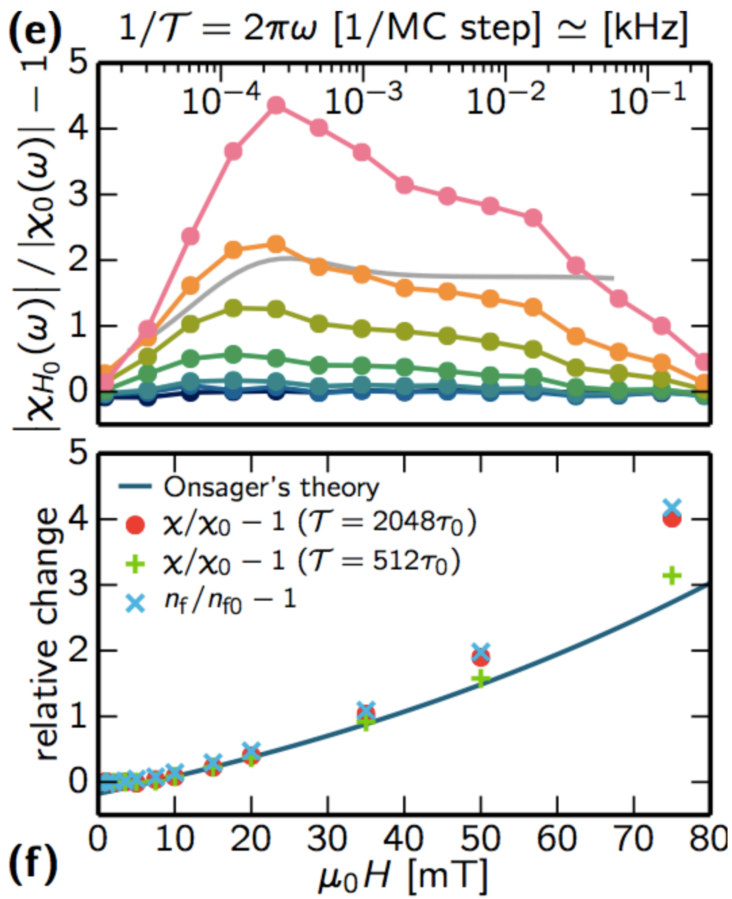
$$\boxed{H = H_0 \sin(\omega t)}$$

In equilibrium $$\chi(H,\omega) = \chi_0(\omega) + \chi_1(\omega) H^2 + \ldots\ldots$$

Wien contribution $$\chi(H,\omega) = \chi_0(\omega) + \tilde{\chi}_1(\omega)\left|\vec{H}\right| + \ldots\ldots$$



$$\frac{\left|\chi_B(\omega_0)\right|}{\left|\chi_0(\omega_0)\right|} \approx \frac{n_f(<\left|\vec{B}\right|>)}{n_f(0)}$$



Analytic approach –two coupled equations

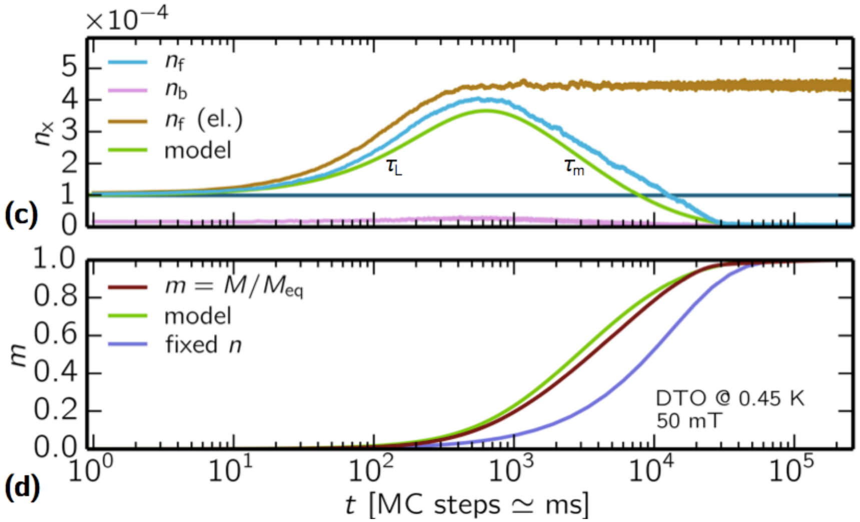
$$\vec{j} = \frac{d\vec{M}}{dt}\frac{1}{\tau_m}\left(\vec{H} - \frac{\vec{M}}{\chi_T}\right)$$

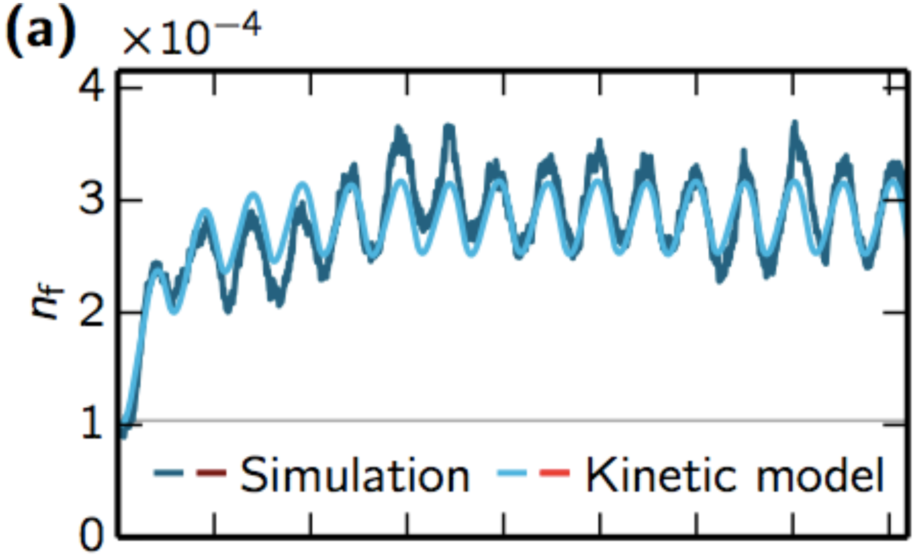
$$\tau_m \propto \frac{\tau_0}{n_f(H)}$$

$$\frac{dn_f}{dt} = k_{\Rightarrow} n_b - \frac{1}{2} k_{\Leftarrow} n_f^2$$

$$\Rightarrow$$

$$\frac{1}{n_f^0}\frac{d\Delta n_f}{dt} \propto \left(|h - m(t)| - \frac{\Delta n_f^0}{n_f^0}\right)$$





Deconfined monopole charge via **The Wien effect**

Bramwell et al, Nature, 461, 956, 2009

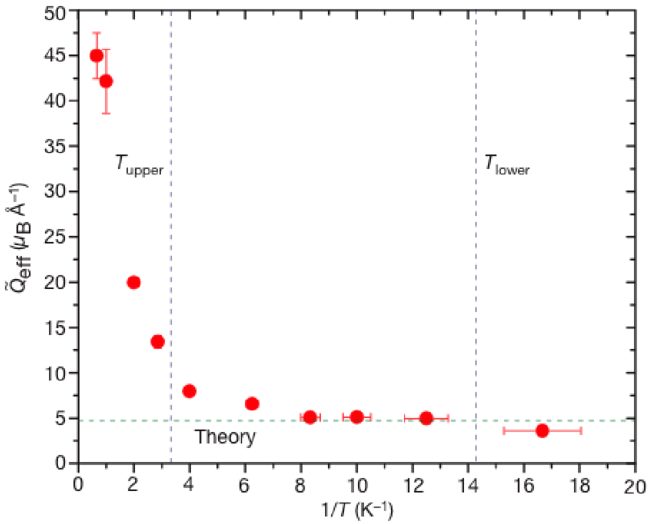


Figure 5 | Experimentally measured 'elementary' magnetic charge $\tilde{Q}_{\rm eff}$ in $Dy_2Ti_2O_7$. Onsager's theory is valid in the regime $T_{\rm lower} < T < T_{\rm upper}$ where the magnetic charges are unscreened. Experimental error bars are e.s.d. The horizontal green line marks the theoretical prediction of ref. 3. Note that $1\,\mu_B$ Å^{-1} $= 9.274 \times 10^{-14}$ J T^{-1} m^{-1}.

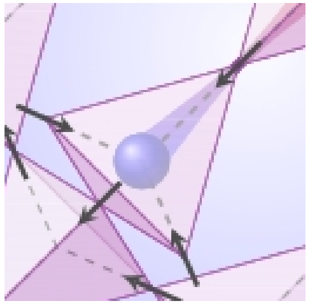
Muon relaxation

$$\boxed{\frac{\delta\sigma(E)}{\sigma} \Rightarrow \frac{\delta\nu(B)}{\nu} = \frac{BQ^3\mu_0}{16\pi k_B^2 T^2}}$$

Highly controversial !

Dunsiger et al, Phys Rev. Lett, 107, 207207, 2011
Sala *et al,* Phys. Rev. Lett. 108, 217203, 2012
Blundell, Phys. Rev. Lett. 108, 147601, 2012

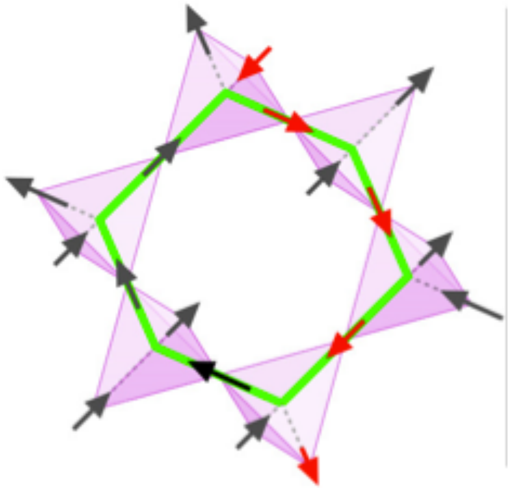
Large internal fields even in the absence of charges



$$\boxed{\left(\vec{\nabla}.\vec{H} = -\vec{\nabla}.\vec{M} = \rho\right)}$$

$$\boxed{\vec{M} = \nabla\psi + \vec{\nabla}\wedge\vec{A} \;\; = \vec{M}_m + \vec{M}_d}$$

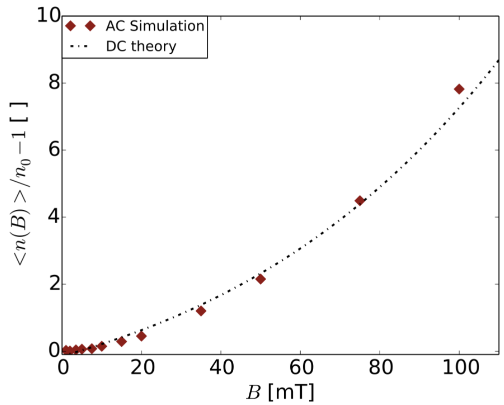
When $\rho = 0, \quad \vec{M} = \vec{M}_d$



Monopolar and dipolar parts (largely) decoupled and dynamics is from monopole movement

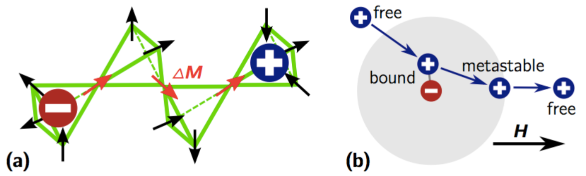
Perfect Coulomb gas within frequency window

$$\boxed{\frac{1}{\tau_L} < \omega < \frac{1}{\tau_m}}$$

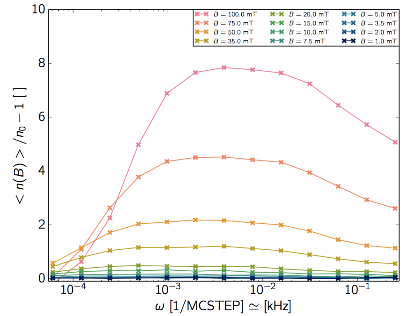


Conclusions

1. The Wien effect is a model non-equilibrium process.



2. The Wien effect emerges from the magnetic Coulomb gas.



3. Spin ice proves to be a perfect, symmetric Coulombic system.