Debye-Hückel Theory and Internal Fields in Spin Ice

Claudio Castelnovo

Hubbard Theory Consortium

Department of Physics

Royal Holloway University of London

15-12-2011 – Workshop on Geometrically Frustrated Magnets: From Spin Ice to Kagome Planes

International Institute of Physics, Natal, Brazil.

Collaborators

G. Sala
Royal Holloway

R. Moessner
MPI-PKS Dresden

S. L. Sondhi
Princeton University

K. Kitagawa
M. Takigawa
ISSP Tokyo

R. Higashinaka(*)
Y. Maeno
Kyoto University
(*)Tokyo Metropolitan Univ.

D. A. Tennant
B. Klemke
HZB

R. S. Perry
Edinburgh

and several discussions with:

S. T. Bramwell UCL
S. Giblin, P. Baker ISIS
J. Quintanilla Kent
S. J. Blundell Oxford

EPSRC
Engineering and Physical Sciences Research Council

Hubbard Theory Consortium

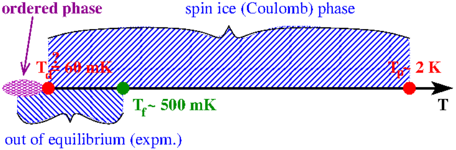
SEPnet
South East Physics Network
Working together to promote excellence in Physics

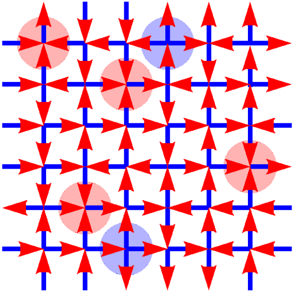
Outline

- Debye-Hückel theory for spin ice:
  - 'non-interacting' low temperature limit
  - Coulomb interactions important in the temperature range 0.1–1 K: heat capacity and magnetic susceptibility
  - corrections: mono-antimono pairing and entropic interaction
- internal field distribution:
  - monopole fields can be observed (in principle)
  - field strength distribution akin to disordered magnet but opposite temperature dependence
  - NMR, μSR, avalanches, and surface probes

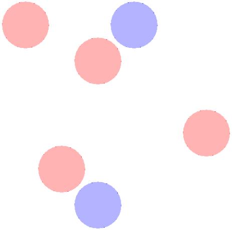
Spin ice as a Coulomb liquid

CC, RM, SLS 2008





$\Leftrightarrow$



+ Coulomb interactions
+ entropic interactions
+ kinematic constraints

Free energy: (Δ = bare monopole cost; ρ = monopole density)

$$F \sim \Delta\rho + T\rho\ln\rho + F_{\rm int}$$

Debye-Hückel (DH) Theory

approximation for the electrostatic energy:

$$F_{\rm el} \sim T\left[\frac{\kappa^2}{2} - \kappa + \ln(1+\kappa)\right], \qquad \kappa \propto \sqrt{\frac{E_{\rm nn}\rho}{T}}$$

inverse screening length

where $E_{\rm nn}$ = nearest-neighbour Coulomb energy (dependent on magnetic as well as entropic charge)

$$F[\rho] \sim \Delta\rho + T\rho\ln\rho + F_{\rm el}$$

$$\frac{\delta F[\rho]}{\delta\rho} = 0 \qquad \Rightarrow \qquad \rho_{\rm eq}(E_{\rm nn}; T)$$

(numerical recursive solution)

From $F[\rho_{\rm eq}(E_{\rm nn}; T)]$ one obtains straighforwardly several thermodynamic quantities (e.g., heat capacity, monopole density)

Debye-Hückel (DH) Theory

CC, RM, SLS '09-11

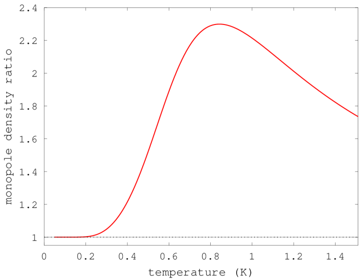
approximation for the electrostatic energy:

$$F_{\mathrm{el}} \sim T\left[\frac{\kappa^2}{2} - \kappa + \ln(1+\kappa)\right], \qquad \kappa \propto \sqrt{\frac{E_{\mathrm{nn}}\rho}{T}}$$

inverse screening length

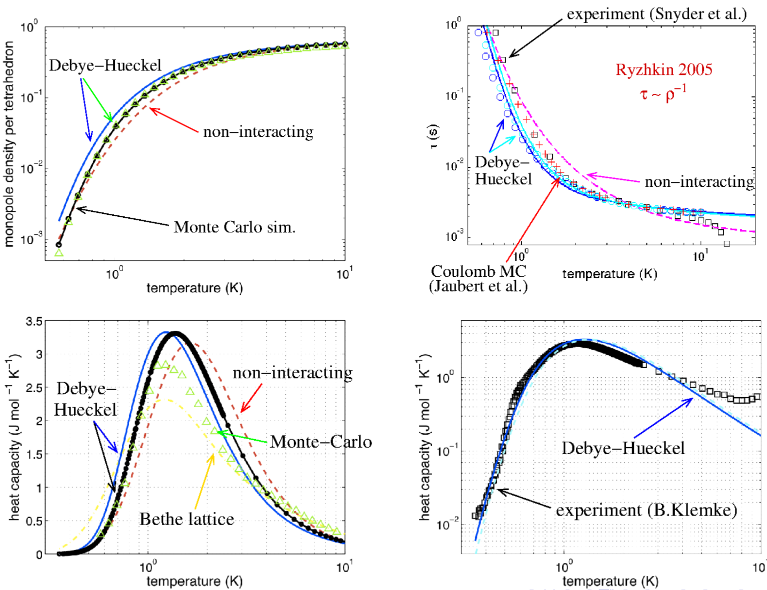
where E_{nn} = nearest-neighbour Coulomb energy (dependent on magnetic as well as entropic charge)

Cf. ratio of DH density vs. non-interacting case $\rho_0(T) \sim \exp(-\Delta/T)$



'non-interacting' at low T but up to twice the density in the regime of experimental interest

Density, Heat Capacity, and Susceptibility CC et al. '09-11



Possible Extensions

CC, RM, SLS 2011

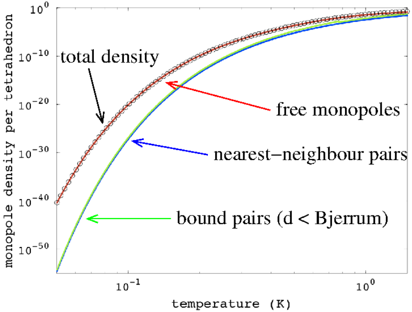
entropic contribution:

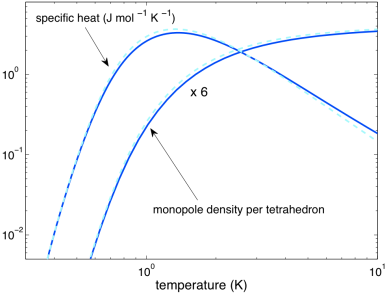
$$\mathcal{P} \propto \exp\left[-\frac{\mathcal{K}}{2} v_{\text{cell}}^{-1} \int \left|\vec{B}^{\text{ent}}(r)\right|^2 d^3 r\right]$$

$$\times \exp\left[-\frac{\mu_0}{2k_B T} v_{\text{cell}}^{-1} \int \left|\vec{H}^{\text{mag}}(r)\right|^2 d^3 r\right]$$

first term $\longrightarrow$ $E_{\text{nn}}^{\text{ent}} = \frac{2T}{\sqrt{3}\pi}$

modify DH free en.: $E_{\text{nn}} + E_{\text{nn}}^{\text{ent}}$





bound pair formation:

free energy F_d for pairs of monopoles at fixed distance d

bound: $d <$ Bjerrum length ℓ_B

$$F_{\text{tot}} = F_{\text{free}} + 2\sum_{d=1}^{\ell_B} F_d$$

Conclusions I

Debye-Hückel theory appears to capture reasonably well the low-tempeature properties of spin ice materials (heat capacity, magnetic susceptibility, monopole density at equil.)

- better than conventional approaches for localised spin systems $\Rightarrow$ evidence of the Coulomb liquid nature of the monopole excitations
- entropic and pairing corrections appear to be unimportant
- DH is a promising starting point for further microscopic modelling of response and relaxation processes

  $\longrightarrow$ understanding slow dynamics and freezing in spin ice

Internal fields in spin ice

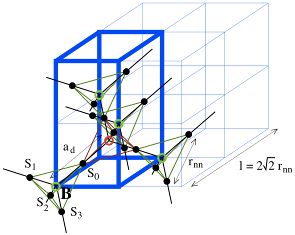
why do we care?

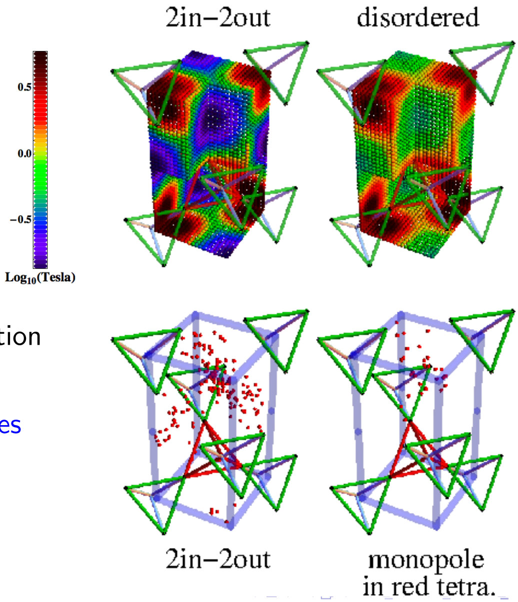
- key to understand local probes (μSR, NMR, SQUID)
- measuring the Coulombic field is the ultimate signature of the presence of monopoles
- further insight on distinctive properties of spin ice vs. conventional magnets

challenges: (from a theoretical perspective)

- several contributions difficult to de-convolve: overall magnetisation, dipolar fields from nearby spins, emergent Coulombic contribution
- internal monopole field small compared to nearby spin fields
- $\nabla \cdot \vec{M} + \nabla \cdot \vec{H} = \nabla \cdot \vec{B} = 0$: 'Dirac string' flux compensates for sources and sinks of $\vec{H}$

Spatial distribution of average field strength

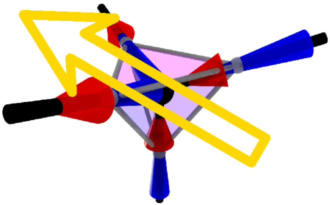




spatially resolved distribution of field strengths:

- very few low-field sites ($\lesssim$ 10 mTesla)
- disorder increases the local fields!

Spin ice vs. conventional ferromagnet

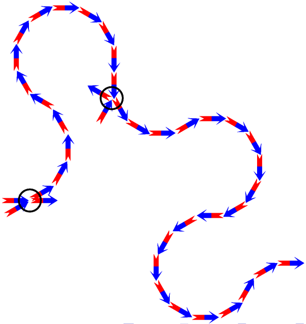


spin ice is locally a ferromagnet
(positive Curie-Weiss temp.)

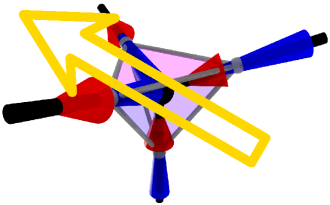
$\Rightarrow$ field strength should increase with order (low-T)

2in-2out ice rules
$\Updownarrow$
divergenceless constraint

spins form closed "flux tubes" with vanishing overall dipole moment



Spin ice vs. conventional ferromagnet

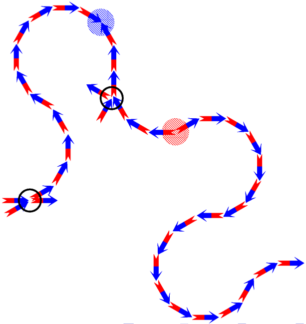


spin ice is locally a ferromagnet
(positive Curie-Weiss temp.)

$\Rightarrow$ field strength should increase with order (low-T)

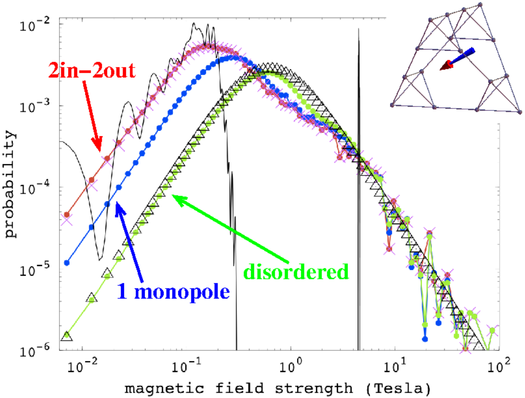
2in-2out ice rules
$\Updownarrow$
divergenceless constraint

spins form closed "flux tubes" with vanishing overall dipole moment



Distribution of field strengths

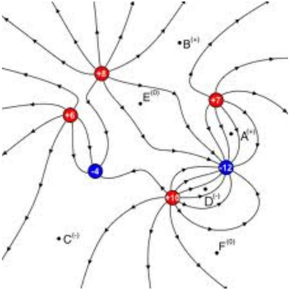
- fields increase as monopole number (disorder) increases
- low-field sites away from spins (super-tetrahedra)
- large fields at centres of tetrahedra



overall random field distribution:

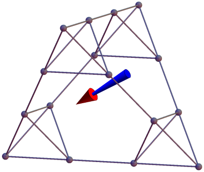
$$P(h) \sim \frac{h^2}{\left(h^2 + H_0^2\right)^2}$$

Probing the Coulombic field



strategy:

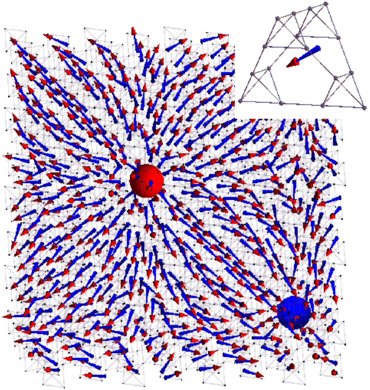
- reduce single spin contributions by keeping as far as possible
- vector-average the fields over configurations with fixed monopole positions



(super-tetrahedra centres are most isolated)

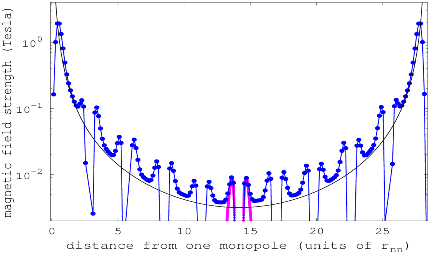
Probing the Coulombic field

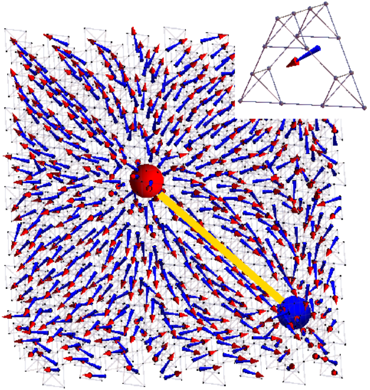
strategy: vector-field average at super-tetrahedra centres over spin ice configurations with fixed monopole positions



Probing the Coulombic field

strategy: vector-field average at super-tetrahedra centres over spin ice configurations with fixed monopole positions



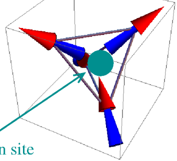


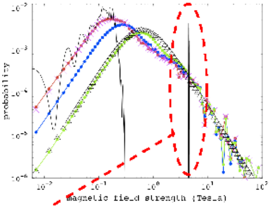
clear signature of magnetic Coulomb field

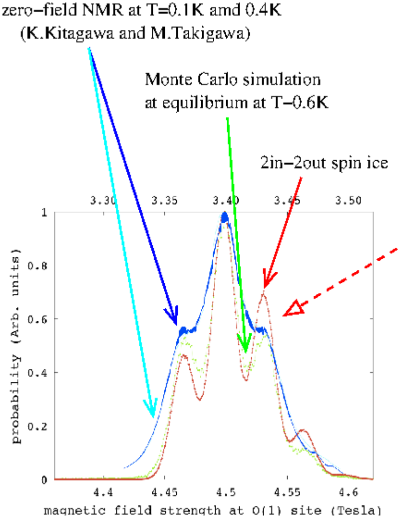
quantitative agreement with theoretical prediction of monopole charge

residual periodic deviations due to nearby spins and 'Dirac strings'

Zero-field NMR

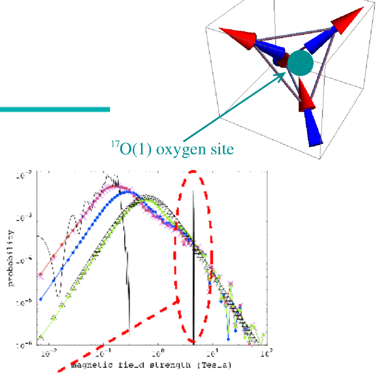


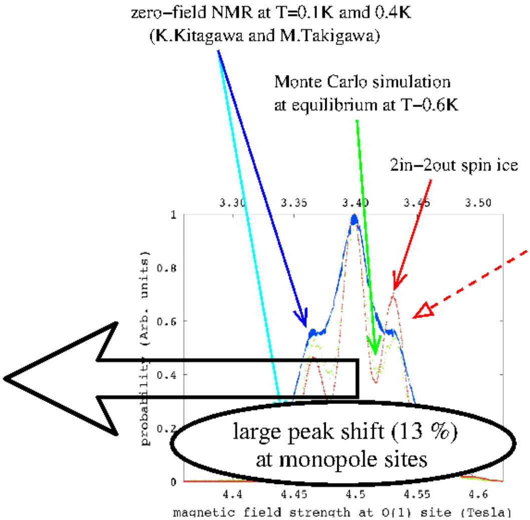




- NMR provides info about local spin correlations

Zero-field NMR





- NMR provides info about local spin correlations
- the O(1) field is reduced by 13% in presence of a monopole ⇒ potential for direct measure of monopole density

Considerations regarding μSR measurements

- large internal fields are inconsistent with muon spin precession doubling when $B_{\rm ext} = 1$ mTesla $\rightarrow 2$ mTesla
  $\Rightarrow$ unlikely that muons are sitting inside *pristine* bulk

- increase in monopole density $\rightarrow$ increase in internal fields $\rightarrow$ faster decay in μSR signal
  $\Rightarrow$ confirmation of one mechanism at the basis of the Wien interpretation of μSR

- what about muons close to but outside the sample? Can a possible monopole contribution $\sim 1/R^2$ be isolated in the stray magnetisation? Temperature dependence may be used to discriminate

further work needed — perhaps other surface probes? (e.g., spatially-resolved magnetisation measurements)

Conclusions II

- internal field distribution akin to random spins **but** spin ice correlations lead to stark constrast with conventional ferromagnets
- more defects ⇒ larger internal fields: temperature dependence can be used to discriminate a magnetic signal from monopole vs. overall magnetisation
- Coulombic magnetic fields from monopoles can be measured inside spin ice (but may require appropriate time averaging)
- internal fields at O(1) sites can be accessed via zero-field NMR: (i) local spin correlations; (ii) potential avenue for direct measure of monopole density