Spin ice and magnetricity

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Outline

Introduction to spin ice:
Spin ice analogous to water ice.
Effective interaction.
Canonical Ho₂Ti₂O₇ and Dy₂Ti₂O₇ spin-ice materials.
Spin ice and emergent quasi particles (magnetic monopoles).
Magnetricity.
Quantum ices.

- Ultrasound experiment: Ultrasound set-up. Acoustic modes in cubic crystals.
- Results:

Intrinsic and extrinsic nonstationary field-driven processes. Phase transition in the spin-ice materials $Dy_2Ti_2O_7$ and $Yb_2Ti_2O_7$.



- Geometrical frustrated magnets and emergent phenomena.
- Topological excitations which can be experimentally observed.
- The prominent example: fractionalized magnetic dipoles in spin ice.
- Some remarkable phenomena: in presence of magnetic monopoles.
- Lattice degrees of freedom and crystal-electric-field effects in the spin ice.

Spin ice analogous to the water ice.

- Ice rule means 2 spins are directed inward and 2 spins are directed outward.
- The ground state has macroscopic degeneracy.



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Spin configurations in 2D

Spin ice analogous to the water ice.

- Ice rule means 2 spins are directed inward and 2 spins are directed outward.
- The ground state has macroscopic degeneracy.
- Pauling entropy can be derived for spin ice.







Spin configurations in 2D

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Effective interaction

• Effective interaction is made up of nearest neighbor exchange and dipolar interactions.

$$J_{eff} = J_{nn} + D_{nn}$$

• Dipole-dipole interaction is dominated

 $J_{nn}(HTO) = -0.52 \, K$

 $J_{nn}(DTO) = -1.24 K$

 $D_{nn}(DTO \& HTO) = 2.35 K$

 Phase diagram indicates that for J_{nn}/D_{nn}> - 0.91 a spin-ice state is stable.



• spin-ice properties can be seen even for antiferromagnetic nearest-neighbor exchange interactions, $J_{nn} < 0$, as long as $J_{eff} > 0$.

B. C. den Hertog and Michel J. P. Gingras, Dipolar interactions and origin of spin ice in Ising pyrochlore magnets, Phys. Rev. Lett. 84, 3430 (2000).

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Spin ice and magnetic monopoles



$$q_m = \pm 2\mu/a_d$$

• This kind of magnetic monopoles obeys the magnetic Coulomb law.

$$V(r_{\alpha\beta}) = \begin{cases} \frac{\mu_0}{4\pi} \frac{Q_\alpha Q_\beta}{r_{\alpha\beta}} & \alpha \neq \beta \\ \frac{1}{2} v_0 Q_\alpha^2 & \alpha = \beta \end{cases}$$

$$\mu = qa = \uparrow = \uparrow a$$

·q



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

Spin ice and magnetic monopoles



• Monopole is connected to antimonopole via Dirac string.

$$q_m = \pm 2\mu/a_d$$

• This kind of magnetic monopoles obeys the magnetic Coulomb law.

$$V(r_{\alpha\beta}) = \begin{cases} \frac{\mu_0}{4\pi} \frac{Q_{\alpha}Q_{\beta}}{r_{\alpha\beta}} & \alpha \neq \beta \\ \frac{1}{2}v_0 Q_{\alpha}^2 & \alpha = \beta \end{cases}$$

$$\mu = qa = \uparrow = \uparrow a$$

-a

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



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Magnetic monopoles and Dirac string

- Effects of Neutron-Dirac string can be seen as broadening of pinch-point
 features in the Brillouin zone.
- diffuse scattering occurs when the magnetic spins on the same site in different unit cells are not the same.
- Required energy to separate the monopole-antimonople approaches to zero.



T. Fennell, P. P. Deen, A. R. Wildes, K. Schmalzl, D. Prabhakaran, A. T. Boothroyd, R. J. Aldus, D. F. McMorrow, and S. T. Bramwell, Magnetic Coulomb Phase in the Spin Ice Ho2Ti2O7, Science 415, 326 (2009).

Quantum effects in spin ice

- In these materials, there is a quantum tunneling between different configurations (2 spins in and 2 spins out) of the spin ice ground state.
- In quantum ice materials J_{nn} is comparable or very close to D_{nn}.
- Neutron scattering and μSR provide evidence that Tb₂Ti₂O₇ and Yb₂Ti₂O₇ are examples for quantum spin-ice materials.
- A Bose-Einstein condensation of monopoles via a Higgs mechanism during the transition.





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T. Fennell, P. P. Deen, A. R. Wildes, K. Schmalzl, D. Prabhakaran, A. T. Boothroyd, Magnetic Coulomb Phase in the Spin Ice Ho 2 Ti 2 O 7, Science 415, 326 (2009).

Monopoles and magnetricity

- Consideration of monopoles by mapping to electrolyte.
- Bound defects (monopoles) are equivalent of associated ion pair and lead to uncharged solvent.
- The unbound defects (free monopoles) are equivalent to dissociated ionic defects.
- Two relaxation times related to the bound pair and unbound monopoles.
- MuSR can provide some signature for the relaxations or effective charge and conductivity of magnetic monopoles.
- The field energy $-QBr_z$ competes with the coulomb potential $-\mu_0 Q^2 4\pi r$

S. T. Bramwell, S. R. Giblin, S. Calder, R. Aldus, D. PrabhakaranT. Fennell, Measurement of the charge and current of magnetic monopoles in spin ice, Nat. Phys. 46, 461 (2009).



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Structure and phase diagram of the spin-ice materials

- Transitions from spin ice to Kagome ice and from Kagome ice to saturated state.
- Phase diagrams can be interpreted in context of magnetic monopoles.

A first-order phase transition is observed in both spin-ice compounds which can be interpreted as transition from low concentration to high concentration of monopoles.



H. Aoki, et al., J. Phys. Soc. Jpn. 73, 2851 (2004).

C. Krey et al., Phys. Rev. Lett. 108, 257204 (2012).

Ultrasound set-up

- A proper probe of the phononic degrees of freedom and Phase transitions.
- This experiment can be performed in a wide range temperatures and magnetic fields.



Ultrasound set-up

B(T)

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- A proper probe of the phononic degrees of freedom and Phase transitions.
- temperatures and magnetic fields.





- The elastic constant c_{ij} is related to the sound velocity $c = \rho v^2$, where ρ is the mass density.
- The cubic system requires only three independent elastic constants of c_{11} , c_{12} , c_{44}

B. Lüthi, Physical Acoustics in the Solid State, (Springer, Berlin, 2005).

Calculation of elastic constants using sound velocity measurements



- The elastic constant c_{ij} is related to the sound velocity $c = \rho v^2$, where ρ is the mass density.
- The cubic system requires only three independent elastic constants of c_{11} , c_{12} , c_{44} .

• A more general relationship between the stress tensor, σ_{ik} , and the strain tensor \mathcal{E}_{jl} can be established by $\sigma_{ik} = \sum_{jl} c_{ikjl} \mathcal{E}_{jl}$ in which the elastic constant, c_{ikjl} , is a four–index tensor.

$c_L = \frac{c_{11} + 2c_{12} + 4c_{44}}{3}$	$k \parallel u \parallel [111]$
$c_T = \frac{c_{11} + c_{44} - c_{12}}{3}$	$k \parallel [111] \bot u$
$c'_L = \frac{3c_L + 3c_{11} - 12c_T}{6}$	$k \parallel u \parallel [112]$
$c_{44} = \frac{6c_T - 3c_{11} + 3c_L}{6}$	$k \parallel u \parallel [001]$

 Another elastic constants along various crystallographic directions of a cubic systems are a combination of these independent constants.

B. Lüthi, Physical Acoustics in the Solid State, (Springer, Berlin, 2005).

Temperature and sweep-rate effects



Different temperatures

- First order gas-liquid transition.
- Different regimes within a single peak.

S. Erfanifam et al., Phys. Rev. B 84, 220404(R) (2011)

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Temperature and sweep-rate effects



Different temperatures

- First order gas-liquid transition.
- Different regimes within a single peak.



Different sweep-rates

- In-situ thermometer
- Peak to peak distance versus sweep rate.
- S. Erfanifam et al., Phys. Rev. B 84, 220404(R) (2011)

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Intrinsic and extrinsic nonstationary field-driven processes





- Magnetic field drives the system out of equilibrium. Re-equilibration can occur in the form of avalanches with the temperature rising up to T_f~ 600 mk.
- Nonstationary processes: The release of (Zeeman) energy from spins, and transfer of the energy out of the sample.

S. Erfanifam et al., Phys. Rev. B 84, 220404(R) (2011)

Additional evidence for Intrinsic and extrinsic processes



- Different heat- bath coupling conditions significantly affect the nonequilibrium states.
- Demagnetization factor affects the transition field.

S. Erfanifam et al., Phys. Rev. B 84, 220404(R) (2011)

Dynamical steady state configuration of spins



- A sharp and quite unique dip at the gasliquid transition.
- Transition to some dynamical steady-state configuration of spins with energy higher than the lowest energy of spins.
- In dynamical steady states two or more reversible processes occur at the same rate.
- phononic pumping and redistribution of excited (lifted) states lead to the reduction of the sound attenuation.
- simulation based on exchange-striction coupling.
- S. Erfanifam et al., Phys. Rev. B 84, 220404(R) (2011)

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 Field dependence of the sound velocity for different field and sound propagation directions at 0.29 K under weak thermal-coupling conditions.

- Symmetric quasi-periodic peaks respect to the applied field polarity.
- Along [001] direction Kasteleyn transition was predicted which not very well resolved by ultrasound.



Field-dependence acoustic characteristics in HTO

- In Ho₂Ti₂O₇, however, we do not observe any non-equilibrium peaks due to monopole avalanches.
- If present, relaxation to equilibrium could be too fast to be detected in our experiment.
- The acoustic characteristics in Ho₂Ti₂O₇, exhibit anomalous features at phase transition in both c_T and c_L acoustic modes.
- Nuclear contribution in HTO is important.



Acoustic signatures of the phases and phase transitions in Yb₂Ti₂O₇

- magneto-elastic coupling as a main mechanism of phonons to spin fluctuations.
- Quantum fluctuations can cause a softening of the sound velocity.
- Bose-Einstein condensation of magnetic monopoles via a Higgs mechanism has been predicted.
- Quantum-spin-ice state to a ferromagnet associated with the condensation of magnetic monopoles or a low-temperature Coulomb ferromagnetic phase that has finite magnetization.



Acoustic signatures of the phases and phase transitions in Yb₂Ti₂O₇

- The magnetic field suppresses quantum fluctuations.
- Longitudinal acoustic modes exhibits sharp anomalies at almost the same range of temperature.
- A very complicated phase diagram in context of magnetic monopole and ongoing open searches.



- Spin ice and quantum ice mapped from water ice.
- Some emergent-quasiparticles promising candidate for future applications in magnetricity.
- intrinsic and extrinsic nonstationary field-driven processes and the role of magneto-acoustic interactions in the (quantum)spin-ice compounds.
- Avalanches of magnetic monopoles occur to re-equilibrate the system driven out of equilibrium.
- A sharp dip in the attenuation. The experimental results agree reasonably well with the theoretical analysis, based on the exchange-striction coupling.
- A number of features that are common to all spin ice systems.
- Enigmatic number of peaks?
- Complicated ground state?

Thanks for your attention

- Thus the monopole concentration should vary between, at most, 10⁻¹⁴ and 10⁻⁴. This is indeed a typical for a weak electrolyte that obeys Onsager's theory.
- network and this almost certainly precludes a direct current.

$$n_{0} = n_{u} + n_{b} \quad \alpha = \frac{n_{u}}{n_{b}}$$
$$K = n_{0} \frac{\alpha^{2}}{1 - \alpha}$$
$$\alpha \ll 1 \rightarrow n_{0} \approx n_{b} \approx N$$

$$\frac{\nu_{\alpha}(B)}{\nu_{\alpha}(0)} = \frac{K(B)}{K(0)} = \frac{\alpha(B)}{\alpha(0)}$$

- A thermally generated spin flip out of this state corresponds to a bound pair of ice's ionic defects.
- The free magnetic monopoles have a short lifetime as annihilation takes place when two monopoles meet each other.
- The magnetic conductivity is proportional to the fluctuation rate of the magnetic moment.
- The quantity $-KT\sqrt{8b}$ is interpreted as the Coulombic barrier to ion pair dissociation.



Exchange – striction coupling

• The Hamiltonian describing exchange striction can be written as follows

$$H_{exs} = \sum_{ij} \left[J \left(\boldsymbol{\delta} + \boldsymbol{u}_i - \boldsymbol{u}_j \right) - J \left(\boldsymbol{\delta} \right) \right] \boldsymbol{S}_i \cdot \boldsymbol{S}_j \,.$$

 Exchange-striction mechanism can be arisen from an acoustic wave modulation of distance between magnetic ions, that affects the exchange interactions.



B. Lüthi, Physical Acoustics in the Solid State, (Springer, Berlin, 2005).





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And Lacknowledge support from "IMPRS"				

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- Quantum ASI and its dynamics.
- Detailed consideration of Quasi-periodic peaks in ASI.





Temperature dependences in low-temperature regime





- In DTO, the sound velocity and attenuation exhibit pronounced anomalies at $T_f \approx 0.5$ K.
- In HTO, the sound velocity shows a drastic change of the slope near 2 K, in which the spin-ice freezing regime terminates in this compound.
- Presumably due to a stronger spin-lattice interaction and coupling to spin degrees of freedom, the $c_{\rm T}$ mode begins to soften at temperatures much higher than the $c_{\rm L}$ mode.





High magnetic field results



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The sound velocity for various acoustic modes and higher fields





Acoustic-mode	d_{23}/d_{12}	d_{34}/d_{23}	d_{34}/d_{12}
c ₁₁	1.006	1.108	1.115
c'_L	1.05	1.082	1.138
c_L	1.179	1.108	1.307
c_T	1.179	1.259	1.494

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- The non-equilibrium peaks, however, are nicely observed also for these modes and field directions in the weak thermal-coupling condition (³He cryostat).
- Indeed, the same number of peaks appear in the up sweeps for all field directions.
- All the features and peaks are symmetric with respect to the applied field polarity.
- The gas-liquid transition is not as well resolved as for the H [[111].



High magnetic field results

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B-sweeps exhibit an anomaly in $Dy_2Ti_2O_7$ for the c_L ' mode (k/|u/|H/|[112]), a similar behavior is seen in $Ho_2Ti_2O_7$ for the c_L mode (k/|u/|H/|[111]).

A hysteresis is observed for both Ho₂Ti₂O₇ and Dy₂Ti₂O₇.





- In DTO, the sound velocity first grows with increasing temperature up to $T_f \approx 0.5$ K, followed by a plateau and a maximum at approximately 1.7 K.
- In HTO, the sound velocity decreases rapidly from the lowest temperatures showing a drastic change of the slope near 2 K, in which the spin-ice freezing regime terminates in this compound.
- At 2 K, that is slightly above T_f a clear change in the slope both in the sound velocity and in the sound attenuation occurs.
- Presumably due to a stronger spin-lattice interaction and coupling to spin degrees of freedom, the $c_{\rm T}$ mode begins to softer at temperatures much higher than the $c_{\rm L}$ mode.

The sound velocity as a function of field for various acoustic modes

0.0 = 0.2 K∆α(dB/cm) -0.2 0.8 K -0.4 1.5 2.5 0.5 1 2 3 $\mu_0 H(T)$ = 0.2 K 0.0 .6 K 0.8 K 1.0 K 10⁵Δv/v -2.5 -5.0 0.5 1 1.5 2 2.5 3 $H_{H}(T)$

Hysteresis in the sound velocity Below 2 T accompanied with Different anomalies.

> A pronounced peak at about 2 T in the sound attenuation can be attributed to a phase transition.

Out of equilibrium monopoles in various crystallographic orientations.

There is conservation of pick numbers in different orientations in the increasing field process.

All the features and peaks are symmetric respect to the applied field polarity.



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Temperature dependence in various acoustic modes

- Most of the crystals show hardening in temperature dependence of the sound velocity from room temperature down to low temperatures.
- There are hardening below 200 K with different slopes in various acoustic modes in spin ice.





TABLE I: Absolute values of sound velocity (elastic constants) in m/s (GPa) measured at T = 1.3 K.

Acoustic-mode	v(DTO)	v(HTO)	$c_{ij}(DTO)$	$c_{ij}(HTO)$
c_{11}	7190	6960	293	354
c_L	6980	6705	326	312
c_T	3670	4040	90	113



Field-dependence acoustic characteristics in HTO



steresis in the sound velocity Below 2 T accompanied with Different anomalies.

The pronounced peak at about 2 T in the sound attenuation can be attributed to a phase transition.

Out of equilibrium phase transition in HTO.

Extended measurements revealed that, transition point in the well-coupled condition shifts to the lower fields in the weakly-coupled condition in HTO.



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Comparison of Ho₂Ti₂O₇ and Dy₂Ti₂O₇



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Comparison in higher fields



Different phases such as: a conventional paramagnetic phase and the spin-ice state are well defined. Kagome ice or spin-liquid phase can be identified.



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Crystal Field energy-level effects in HTO



Comparison in higher fields



Different phases such as: a conventional paramagnetic phase and the spin-ice state are well defined. Kagome ice or spin-liquid phase can be identified.



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