Coulomb physics in spin ice
from magnetic monopoles to magnetic currents

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In the second half of the past century it became apparent that the low temperature
behaviour of condensed matter systems can often be described by modeling their ex-
citations as quasiparticles immersed in an effective vacuum, whose properties derive
directly from those of the low temperature phase of the system. Whereas in the search
for the fundamental constituents in our universe we are bound to look for what is al-
ready there, the combinatorial nature of the periodic table could then be harvested to
realise an endless variety of new vacua, with relative exotic excitations. For instance,
particles that carry a fraction of the electronic charge were found in polyacetylene and
fractional quantum Hall systems; or electron-like quasiparticles that carry charge but
no spin (spin-charge separation) in SrCuO$_2$.

In late 2007, it was suggested that the unconventional low temperature behaviour
of a class of rare earth titanates (namely, Dy$_2$Ti$_2$O$_7$ and Ho$_2$Ti$_2$O$_7$), dubbed spin ice,
can be understood in terms of point-like quasiparticle excitations, with the exceptional
property of carrying a net magnetic charge\cite{1}.

These materials are localised spin systems where the magnetic degrees of freedom
(the rare earth ions) form a corner sharing tetrahedral lattice, with two distinctive fea-
tures: (i) a strong single ion anisotropy causes the spins to be uniaxial, with an energy
barrier in excess of 100 K; and (ii) the interactions between the large rare earth spins
are dominated by the magnetic dipolar coupling.

The combination of these properties results in a frustrated ferromagnet, whose low-
energy configurations are characterised by spin-spin correlations akin to the proton-
proton correlations in water ice – hence the name spin-ice. Every tetrahedron satisfies
the so-called ice rules: two spins point in and two point out (see Fig. 1 left panel).
An exponentially large number of spin configurations satisfy these rules, leading to an
extensively degenerate low-temperature phase, customary of frustrated systems.

The peculiar structure of the low-temperature phase in spin ice manifests itself in
the nature of its excitations. An excited state is generated when the orientation of a
spin is reversed with respect to its lowest energy state, and the two adjacent tetrahedra
no longer fulfill the ice rules (see Fig. 1 middle panel). Defective tetrahedra can then
be separated by means of spin rearrangements that do not introduce further violations
Figure 1: Pictorial representation of a spin configuration in rare earth titanates. Left panel: configuration that satisfies the two-in, two-out ice rules, whereby each tetrahedron has a vanishing net magnetic charge. From the point of view of the Wien effect, this is analogous to undissociated H$_2$O molecules in water. Middle panel: the reversal of a spin introduces two adjacent defective tetrahedra (red and blue sphere), which in the same analogy correspond to a bound [H$_3$O$^+$ OH$^-$] pair. Right panel: an appropriate rearrangement of spins can lead to the separation of two defects without introducing other violations of the ice rules. If the distance between the defects becomes larger than the screening length, they dissociate and behave as free charged ions in water.

of the ice rules (Fig. 1, right panel), at a free energy cost inversely proportional to the distance between the defects. The barrier to separate two adjacent defects to infinity is therefore finite (they are deconfined), and a single defective tetrahedron represents an elementary excitation in spin ice – a rare example of fractionalisation in three dimensions.

This phenomenon is even more striking if we consider the nature of these defects. Spins carry magnetic moment, and the ice rules imply that the spins are oriented with two north poles and two south poles close to the centre of each tetrahedron, in a locally neutral arrangement. Defective tetrahedra with three spins pointing in and one pointing out (or vice versa) are local excesses of north (south) poles. The elementary excitations in spin ice are therefore fractions of the underlying spin degrees of freedom, in that they carry a net magnetic charge – the closest (classical) realisation of a magnetic monopole to date!

The presence of magnetic monopoles allows to explain the liquid-gas structure of the experimental phase diagram in a magnetic field (1) – a feature that was reported as “unprecedented in a localised spin system” – as well as an exceptional increase in the characteristic time scales for magnetic relaxation at low temperatures (2).

Since the theoretical proposal, a broad experimental effort materialised at lightning speed to find more direct confirmations of the existence of these monopoles. Three of the major research groups in the field, based in Germany, England, and Japan, succeeded. Using neutron scattering techniques, distinctive evidence was found for both the characteristic reversed spin chains separating pairs of monopoles, and for the dipolar correlations between the underlying spins, fathering the peculiar nature of these excitations. Together with susceptibility and heat capacity measurements,
the experimental results strongly support the idea that the low temperature behaviour of spin ice is akin to that of a gas of free magnetically charged particles, i.e., a magnetic Coulomb liquid.

What new phenomena can one expect in materials where the low energy phase exhibits magnetic monopole excitations?

To begin with, the onset of the ice rules ought to be substantially different from phase ordering kinetics in more conventional magnets. Concepts like domain growth and coarsening are replaced by diffusion and annihilation of Coulomb-interacting point-like defects. [6]

O. Tchernyshyov, writing in Nature, [7] further speculated that “learning how to move magnetic monopoles around would be a step towards technologies such as magnetic analogues of electric circuits and magnetic memories operating on the atomic scale”.

One important difference between spin ice monopoles and free magnetic charges is that a steady flow (direct current) is forbidden: a monopole moving through the lattice orients the spins along its path in a way that does not allow another monopole with the same charge to follow the same path. However, there are no reasons of principle that prevent alternating currents. A concrete step in this direction was cleverly accomplished by Steve Bramwell and collaborators, combining a 1934 theory by Onsager on the behaviour of electrolytes, with state of the art muon spin rotation measurements. [8]

Weak electrolytes are known to exhibit a non-linear increase in dissociation constant $K$ in presence of an applied electric field – known as the second Wien effect. Consider for simplicity the familiar case of autoionisation in water. While most of the molecules have no net charge, a small fraction of them is dissociated into $\text{H}_2\text{O}^+$ and $\text{OH}^-$ ions. Opposite ions attract each other via Coulomb interactions, and free charges appear only at the cost of overcoming the Coulomb energy barrier to separate them beyond the screening length. The system is therefore governed by two successive thermal equilibria,

$$2\text{H}_2\text{O} \rightleftharpoons [\text{H}_3\text{O}^+ \text{OH}^-] \rightleftharpoons \text{H}_3\text{O}^+ + \text{OH}^-.$$  \hspace{1cm} (1)

An applied electric field $E$ reduces the barrier for bound pairs to become free charges, which affects the dissociation constant $K$ of the second process in Eq. (1). The central result in Onsager’s theory quantifies this change perturbatively in the applied field strength. [9]

$$K(E) = K(0) \left[ 1 + b + \frac{b^2}{3} + \ldots \right]$$ \hspace{1cm} (2)

$$b = \frac{e^3E}{8\pi\varepsilon_0 k_B T^2},$$ \hspace{1cm} (3)

where $e$ is the ionic charge, $\varepsilon_0$ the electric permittivity of the vacuum, $k_B$ Boltzmann’s constant, and $T$ the temperature. A remarkable feature of this thermodynamic result is that it allows to determine experimentally the value $e$ of the carrier charge.

After a sudden change in the applied field, the dissociation constant $K$ relaxes to its equilibrium value exponentially, with a decay rate $\nu_K$ proportional to the conductivity of the system. Onsager showed that in the limit of small free charge density, the
conductivity is in turn proportional to the square root of \( K \), so that
\[
\nu_K(E) \approx \frac{\sqrt{K(E)}}{K(0)} \approx 1 + \frac{b}{2}.
\]

Onsager theory successfully applies to several electrolytes, solid or liquid. If the low-temperature description of spin ice in terms of magnetic charges is correct, then the theory should bear relevance in that context as well, provided we replace \( e \) with the magnetic monopole charge \( Q \), \( E \) with the applied magnetic field \( B \), and \( \varepsilon_0 \) with the permeability of the vacuum \( \mu_0 \). There is however an important difference: the motion of monopoles under the influence of the field leads to a change in the magnetisation of the underlying spin configuration. In the weak field limit, Bramwell and co-workers argue that the magnetisation change per unit forward reaction is constant, independent of the magnetic field, and therefore the relaxation rate \( \nu_\mu \) of the magnetisation decay after a field quench is proportional to \( \nu_K \),
\[
\frac{\nu_\mu(B)}{\nu_\mu(0)} = \frac{\nu_K(B)}{\nu_K(0)} = \sqrt{\frac{K(B)}{K(0)}} \approx 1 + \frac{b}{2}
\]
\[
b = \frac{\mu_0 Q^3 B}{8\pi k_B^2 T^2}.
\]

Eq. (6) allows then to obtain the value of \( Q \) from relative experimental measurements sensitive to the relaxation of the magnetisation of the sample.

The experimental probe of choice to this purpose is transverse field muon spin rotation. The spins of muons implanted in a sample undergo a characteristic oscillatory relaxation behaviour as they precess about the applied field, subject to dephasing caused by the fluctuating local field due to the magnetisation of the sample. At low temperature, when the fluctuations in magnetisation are sufficiently slow, dephasing leads to an exponential decay envelope for the oscillatory behaviour, with a characteristic decay rate proportional to \( \nu_\mu \). The results by Bramwell and co-workers on \( \text{Dy}_2\text{Ti}_2\text{O}_7 \) show clear evidence of the scaling behaviour in Eqs. (5-6) in the temperature range \( 0.07 < T < 0.3 \text{ K} \), and the measured value of the magnetic charge \( Q \approx 5 \text{ } \mu_B\text{Å}^{-1} \) (see Fig. 2) is in good agreement with the one predicted by the theory.

Not only do these measurements provide further compelling evidence of the presence of magnetic monopole excitations in spin ice materials, and of their magnetic Coulomb interactions. They also show that spin ice monopoles respond to external magnetic fields (to leading order) in the same way as electric charges do for instance in water, making \( \text{Dy}_2\text{Ti}_2\text{O}_7 \) the first material of a class that one might rightfully call magnetolytes.

What Bramwell and co-workers have accomplished is the first step towards determining whether macroscopic alternating currents are ultimately achievable in spin ice. This gives new emphasis to the study of magnetic charges in condensed matter system, as well as a concrete perspective to potential technological applications. Further experimental and theoretical efforts are needed to fill the gap from magnetolytes to magnetrecticity – time will tell how far these monopoles can travel.
Figure 2: Experimental values of the magnetic carrier charge in spin ice, [8] compared to the theoretical prediction (horizontal line). [1]

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It was proposed that spin ice, a class of rare earth titanates, hosts magnetic monopoles as elementary excitations. Recently, Bramwell and co-workers measured the Wien effect in Dy$_2$Ti$_2$O$_7$, directly probing the nature of these monopoles and making Dysprosium titanate the first example of a magnetolyte.

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