Emergent Electrochemistry in Spin Ice: Debye–Hückel Theory and Beyond

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The low-temperature picture of dipolar spin ice in terms of the Coulomb fluid of its fractionalised magnetic monopole excitations has allowed analytic and conceptual progress far beyond its original microscopic spin description. Here we develop its thermodynamic treatment as a 'magnetolyte', a fluid of singly and doubly charged monopoles, an analogue of the electrochemical system $2\mathrm{H_2O} = \mathrm{H_3O^+} + \mathrm{OH^-} = \mathrm{H_4O^{2+}} + \mathrm{O^{2-}}$, but with perfect symmetry between oppositely charged ions. For this lattice magnetolyte, we present an analysis based on Debye–Hückel theory, which is accurate at all temperatures and incorporates 'Dirac strings' imposed by the microscopic ice rule constraints at the level of Pauling's approximation. Our results are in close agreement with the specific heat from numerical simulations as well as new experimental measurements with an improved lattice correction, which we present here, on the spin ice materials $Ho_2Ti_2O_7$ and Dy_2,Ti_2O_7. Our study of the magnetolyte shows how electrochemistry can emerge in non-electrical systems. We also provide new experimental tests of Debye–Hückel theory and its extensions. The application of our results also yields insights into the electrochemical behaviour of water ice and liquid water, which are closely related to the spin ice magnetolyte.

[1] Vojtech Kaiser, Jonathan Bloxsom, Laura Bovo, Steven T. Bramwell, Peter C.W. Holdsworth, Roderich Moessner " Emergent Electrochemistry in Spin Ice: Debye-Huckel Theory and Beyond", arXiv:1803.04668.

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