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Enhanced dynamics in disordered non-Kramers spin ice Ho2(Ti1–x-Hfx)2O7 : toward the Coulomb quantum spin liquid state

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Dy2Ti2O7 and Ho2Ti2O7 classical spin ice compounds have been extensively studied over the past 30 years. Their excitations, described as emergent magnetic monopoles, exhibit very slow dynamics at low temperature, which manifests by a freezing and a strong irreversibility in Zero Field Cooled - Field Cooled (ZFC-FC) magnetization measurements. It has been proposed that in spin ices made of non-Kramers magnetic ions, i.e. where J is an integer so that the ground-state crystal electric field (CEF) is a non-protected doublet, such as Ho2Ti2O7, non-magnetic disorder induces quantum fluctuations that can push the system toward a Quantum Spin Liquid phase (QSL). This phase is characterized, inter alia, by a much faster dynamics.
We present a study performed on Ho2Ti2O7 where non magnetic disorder is introduced through a controlled substitution of Ti4+ ions by Hf4+ ions, with substitution rate from 0 to 40%. X-ray and neutron diffraction (HRPT - SINQ) show that the crystal structure and the spin ice correlations are preserved up to at least 30% and that the introduction of disorder results in an oxygen depletion of the Ho3+ coordination shells, so-called Frenkel pair defects, that locally break the CEF symmetry. As a result, a broadening of the CEF levels is observed in inelastic neutron scattering (EIGER - SINQ). Nevertheless, DC magnetic measurements indicate that the Ising nature of the single-ion
ground state remains. Interestingly, AC susceptibility and Neutron Spin Echo (WASP - ILL) reveal a faster dynamics below 30 K as the substitution rate grows. In addition, ZFC-FC measurements show that the freezing temperature is lowered. This speeding up of the dynamics strongly suggests that disorder indeed enables quantum fluctuations in the system. This opens the way to the stabilization of a Coulomb QSL phase at lower temperature.

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