The quantum diffusion of muons in the spin ice $Dy_2Ti_2O_7$

Among the magnetic probes available for studying condensed matter, muon Spin Relaxation (mu-SR) occupies an important place. This technique is highly sensitive to local magnetic fields and can probe spin fluctuations over time scales from roughly 10^{-5} to 10^{-12} seconds. However, the interpretation of mu-SR spectra is not always straightforward owing to the multitude of different interactions that the muons experience in matter. While mu-SR spectra are well understood in many compounds, including some magnetic oxides, their interpretation for the "magnetically frustrated", pyrochlore-lattice compounds remains challenging.

The mu-SR spectrum of the archetypal spin ice Dysprosium Titanate ($Dy_2Ti_2O_7$ or "DTO") appears however to have revealed an unexpected spin dynamics (i.e. large fluctuations of the spins). The muon data suggest that the Dy atom spins fluctuate at a constant time scale of the order of 10^{-6} s from the lowest temperatures yet investigated (a few mK) up to about 7K. This phenomenon has often been called the "persistent spin dynamics" but is problematic for two reasons. First, other magnetic probes do not reveal such a fast, temperature-independent dynamics. For example, recent A.C. susceptibility measurements indicate spin fluctuation rates up to 10 orders of magnitude slower at the lowest temperatures than those suggested by mu-SR studies. Secondly, the theory of spin-ices is now relatively well-understood and it is quite difficult to justify such fast spin dynamics theoretically. In theory, the spins should freeze almost completely below about 0.5 K in DTO.

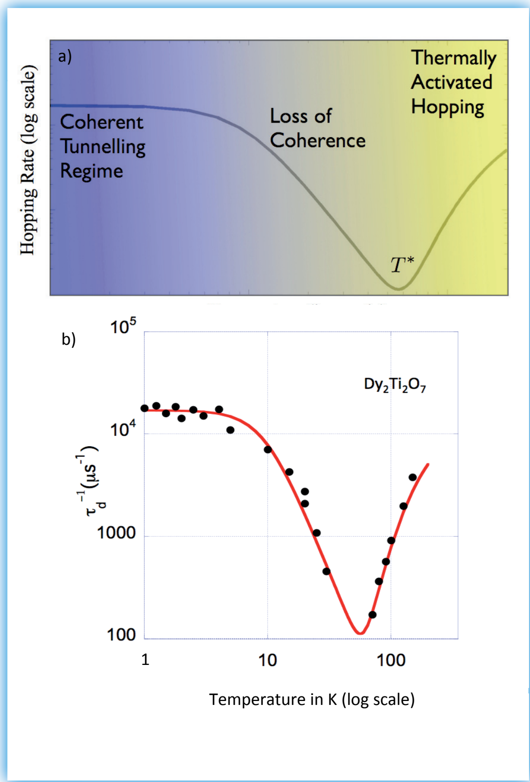

Fig. 1: In (a), the motion of a polaron is characterized by three regimes. At low temperatures, the diffusion rate (or hopping rate) is almost temperature independent. This indicates coherent quantum tunnelling of the particle. At higher temperatures, interaction with phonons progressively destroys the coherent motion up to a temperature T, where the diffusion rate is minimum. Above T*, an incoherent and thermally activated regime takes place.*
In (b): muon spin relaxation times (data points) extracted from the mu-Spin Relaxation experiment on $Dy_2Ti_2O_7$ (Dunsiger et al 2011) compared with our theoretical curve for polaronic muon diffusion.

The crystal structure of a spin-ice consists of atomic spins (the magnetic Disprosium ions in the case of DTO) disposed at the corners of tetrahedra. The strong Ising magnetic anisotropy imposes that the spins at the corners are directed either towards the centre of the tetrahedron or in the opposite direction. That leads to a "frustrated" nature of the magnetism (no given spin can couple anti-parallel with its three neighbours simultaneously) and to the so-called "ice-rule", or "two-in, two-out" rule : Two spins must point towards the centre, and two in the opposite direction.

A standard experiment with muons consists of sending positively charged, *spin-polarized* muons (charge *e+*, spin ½) along a given direction in the studied sample. What is measured is then the depolarization of the muon spins with time. The relaxation rate thus obtained gives us information about the magnetic fluctuations that the muons have felt during their 2.2 microsecond lifetime.

Clearly, there are two main possibilities. The first one is that the muons have become localized in the sample. The only sources of magnetic fluctuations then come from the temporal fluctuations of the spin structure itself. The muons would act here as local probes and it is under this hypothesis that the existence of a "persistent spin dynamics" in DTO has been claimed.

The second possibility is the reverse one : the spins in the structure are assumed frozen at low temperature and the muons are moving. We have adopted the latter hypothesis. Owing to its charge and large mass (207 times that of the electron), a muon interacts very strongly with the phonons of a dielectric such as DTO. That results in a composite particle called a polaron, a muon "dressed" with the crystal's phonons. This object moves as a whole, and calculations show that there are three regimes of diffusion as function of the temperature (see Fig. 1(a)). In the diffusion scenario, the muon spin relaxation rate is proportional to its diffusion time. Our interpretation faithfully reproduces the experimental data of S. R. Dunsiger et al (PRL 2011) over a very wide temperature range (see Fig. 1(b)) and there is so far no other competitive theory.

Our work gives the first scenario that can rule out the "persistent spin dynamics" hypothesis for systems like DTO. To go further, we have also measured mu-SR in an equivalent but non-magnetic compound: $Y_2Ti_2O_7$. Here, the muons are found to be sensitive to the spin nuclei and to magnetic defects like Ti^{3+} ions.

CONTACT

Pascal QUEMERAIS
pascal.quemerais@neel.cnrs.fr

FURTHER READING

Muon diffusion and electronic magnetism in $Y_2Ti_2O_7$
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Possible quantum diffusion of polaronic muons in $DY_2Ti_2O_7$ spin ice
P. Quémerais, P. McClarty and R. Moessner
Phys. Rev. Lett. 109, 127601 (2012)