Coherent spin quantum dynamics in antiferromagnetic rings

Florian Meier [a,1], Daniel Loss [a]

[a] *Department of Physics and Astronomy, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland*

Abstract

Molecular magnetic clusters with antiferromagnetic exchange interaction and easy axis anisotropy belong to the most promising candidate systems for the observation of coherent spin quantum tunneling on the mesoscopic scale. We point out that both nuclear magnetic resonance and electron spin resonance on doped rings are adequate experimental techniques for the detection of coherent spin quantum tunneling in antiferromagnetic molecular rings. Although challenging, the experiments are feasible with present day techniques.

Key words: molecular magnet; mesoscopic quantum phenomena; spin quantum tunneling

1. Introduction

Quantum tunneling of the magnetization has been widely studied in ferromagnetic molecular magnetic systems during the past years. In contrast to ferromagnets, where spin dynamics is induced by a transverse anisotropy or magnetic field, in antiferromagnetic systems the exchange interaction gives rise to pronounced spin quantum dynamics [1]. To date, several molecular rings with antiferromagnetic nearest neighbor exchange interaction have been synthesized [2] and well characterized [3]. In these rings, an easy axis anisotropy tends to align the spins along the z–axis perpendicular to the ring plane. Due to the spin dynamics induced by antiferromagnetic exchange, the classical Néel ordered states are not energy eigenstates. Rather, the spins are expected to tunnel jointly between the two degenerate classical ground state configurations [4–7]. Due to the small tunnel action $\mathcal{S}/\hbar = 2$–4, the tunneling rate Δ can be as large as several GHz and tunneling is expected to be coherent. In contrast to ferromagnetic molecular clusters where incoherent spin tunneling is readily observed by ESR, tunneling of the Néel vector in antiferromagnetic systems with compensated sublattice spins leaves the total spin invariant. The experimental detection of coherent Néel vector tunneling in antiferromagnetic rings hence requires probes coupling to single spins of the rings only.

[1] E-mail: Florian.Meier@unibas.ch

2. Nuclear magnetic resonance

Nuclear spins are natural candidates for probes coupling only to single electron spins. For a nuclear spin $I = 1/2$ coupled to one electron spin with spin quantum number s by a hyperfine contact interaction $\hat{H}' = A\hat{\mathbf{s}}_1 \cdot \hat{\mathbf{I}}$ [Fig. 1a], the nuclear magnetic susceptibility accessible in CW nuclear magnetic resonance (NMR) can be evaluated explicitly. In presence of a magnetic field B_x applied in the ring plane, for $\Delta \gg As$ we find that

$$\chi''_{I,yy}(\omega) \simeq \frac{\pi}{4}\Big[\tanh\left(\frac{\beta\gamma_I B_x}{2}\right)\delta(\omega - \gamma_I B_x/\hbar) + \left(\frac{As}{\Delta}\right)^2 \tanh\left(\frac{\beta\Delta}{2}\right)\delta(\omega - \Delta/\hbar)\Big] - [\omega \to -\omega] \quad (1)$$

exhibits a satellite resonance at the tunnel splitting Δ of the electron spin system [5]. The reason for this is that the coherently tunneling electron spin produces an oscillating hyperfine field $As\cos(\Delta t/\hbar)$ at the site of the nucleus which leads to a precession of $\mathbf{I}$ around the z–axis superimposed on the one around the static field B_x. The rapid change in the electron spin direc-

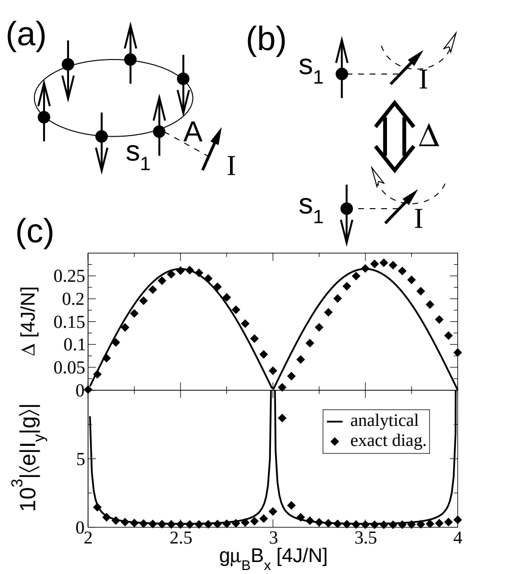

Fig. 1. (a) A nuclear spin $\mathbf{I}$ coupled to one electron spin of the ring by hyperfine interaction acts as a local probe for the electron spin dynamics. (b) The coherent tunneling leads to a rapid change of the electron spin direction from $+\mathbf{e}_z$ to $-\mathbf{e}_z$ at a rate Δ/h. The precession of $\mathbf{I}$ around the z–axis is reversed at the same rate. (c) Comparison of theoretical and analytical predictions for the tunnel splitting Δ and the spectral weight of the NMR satellite resonance for a small model systems ($N = 4$, $s = 3/2$, $k_z/J = 0.2$).

tion due to tunneling results in a reversal of the precession direction of $\mathbf{I}$ at a rate Δ/h [Fig. 1b]. Hence, the spectral weight of the corresponding peak in $\chi''_{I,yy}$ is usually small, but can be increased by reducing Δ. Our analytical results are in good agreement with exact diagonalization of small systems [Fig. 1c].

3. Doped antiferromagnetic rings

In doped antiferromagnetic rings in which one of the Fe(III) or Cr(II) ions with spin $s = 5/2$ or $s = 3/2$, respectively, is replaced by an ion with different spin $s' \neq s$, the ring acquires an excess spin which traces the dynamics of the Néel vector [Fig. 2a]. Local spin probes then are no longer required to detect quantum coherent spin tunneling. Rather, the ESR spectrum exhibits a resonance at the tunnel splitting Δ, the width of which is an upper bound for the decoherence rate of electron spin tunneling. The curves in Fig. 2b, obtained by numerical exact diagonalization, show that for doped Na:Fe_6 rings spin quantum tunneling is associated with a change in staggered magnetization by $\simeq 15\hbar$. Hence, doped antiferromagnetic rings are promising candidates for the observation of coherent spin quantum tunneling.

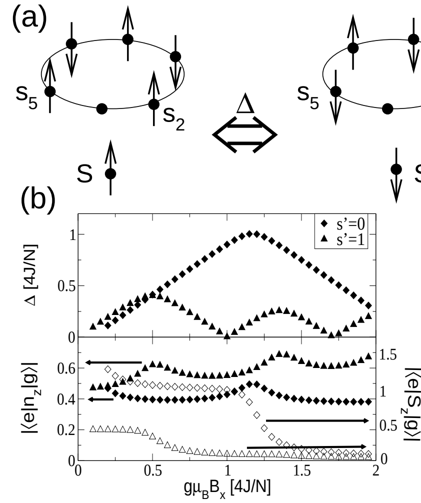

Fig. 2. (a) In doped molecular rings, in which one of the spins s in the ring is replaced by one with a different spin quantum number (here, $s' = 0$), a finite net spin S emerges which oscillates as the Néel vector tunnels. (b) Tunnel splitting Δ and transition matrix elements $|\langle e|\hat{n}_z|g\rangle|$, $|\langle e|\hat{S}_z|g\rangle|$ for Na:Fe_6 doped with $s' = 0$ (diamonds) and $s' = 2$ (triangles). $|e\rangle$ and $|g\rangle$ are the tunnel–split states. $|\langle e|\hat{n}_z|g\rangle|$ (filled symbols) is proportional to the tunneling staggered spin $\simeq 2Ns|\langle e|\hat{n}_z|g\rangle| \simeq 15\hbar$. The finite value of $|\langle e|\hat{S}_z|g\rangle|$ shows that the transition is ESR active.

Acknowledgements

We acknowledge support by the European TMR network MOLNANOMAG, the BBW Bern, the Swiss NSF, and DARPA.

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