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QUANTUM PHENOMENA IN SMALL ANTIFERROMAGNETS*

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We show that the most characteristic properties of mesoscopic antiferromagnets can be explained in terms of the response on a spatially *inhomogeneous* perturbation. This concept allows to explain the dynamic properties (quantum resonance, coherence and tunnelling rates) as well as static perturbations which, for increasing size of an antiferromagnet, leads to a transition from the quantum mechanical oscillating system to a classical antiferromagnet with well defined Néel vectors.

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A classical antiferromagnet (AFM) can be characterised by a doubly degenerate ground state corresponding to different spin orientation in two sublattices. Within this model, the transition probability between the two degenerate Néel states induced by external perturbations in small AFM, has been estimated by Barbara and Chudnowski [1]. It is not obvious whether this transition rate is equivalent to "macroscopic quantum coherence" (MQC), i.e. the coherent transition between states which originate from coupling of the AFM grain to its environment [2]. Experimental results, and here in particular the resonance observed by Awschalom et al. [3] and also a puzzling interlayer coupling [4], indicate rather oscillatory character of small AFM. In the original paper [3], the authors interpreted the experimental data within the tunnelling formalism but Garg [5] argues that this resonance cannot correspond to a transition between the Néel states but rather to a magnetic resonance of uncompensated magnetic moments.

To clarify this problem a systematic quantum-mechanical (QM) analysis is needed. Some calculation has been recently done by Levine and Howard [6]. They discuss a rigorous solution of Heisenberg AFM clusters but they do not consider

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any coupling of the cluster to the environment. As a consequence, their quantum resonances, which they call MQC, cannot be related to MQC as defined by Leggett [2].

In this paper we discuss the QM solution for a simple AFM cluster and we consider possible perturbations. Time dependent fields lead to resonant transitions while the static perturbations, which correspond to a coupling to the environment, lead to the formation of static staggered magnetisation with two possible equivalent orientations, reflecting the transition from QM to the classical limit. Such an analysis allows us to distinguish simple QM transitions between the ground- and excited states, which we relate to Awschalom's resonance, and MQC within the doubly degenerate ground state.

The crucial points of our considerations are: (i) only spatially inhomogeneous fields can perturb the ground state of the AFM cluster and (ii) the induced magnetisation, $\mu(r_i)$, has different spatial distribution as compared to the perturbing field $H_i = H(r_i)$.

Our considerations are based on the analysis of 1D AFM strings with an even number of spins. This is a simplest AFM object where the influence of perturbations can be analysed. The following Hamiltonian describes the exchange coupling, J, and the magnetic anisotropy, D, caused by a non-Heisenberg interspin coupling:

$$\mathcal{H} = \sum_{i,j} J_{i,j} \boldsymbol{S}_i \boldsymbol{S}_j + \sum_{i,j} D_{i,j} \left(3 S_{i,z} S_{j,z} - \boldsymbol{S}_i \boldsymbol{S}_j\right). \tag{1}$$

As an example, the resulting quantum energetic structure for $N = 10$ particles, and its dependence on the anisotropy parameter D is shown in the inset to Fig. 1. It is characterised by a non-magnetic singlet ground state, $\langle S_i \rangle = 0$. The magnetic anisotropy splits the first excited triplet state and, for axial anisotropy (positive D), two singlets are the lowest states of the AFM grain. With increasing magnetic anisotropy, the energetic distance, Δ, between the singlets decreases approaching zero for Ising anisotropy ($D/J = 1$). Simultaneously, the energy gap above these quasi-degenerate singlets increases. The dependence Δ, for two different anisotropy parameters, D, is shown in Fig. 1. With increasing particle size, N, the energy splitting, Δ, decreases as $1/N$ for Heisenberg coupling and much faster for an axial anisotropy.

We consider the perturbations which are linear in the spin components

$$\mathcal{H}_\mathrm{p} = g\mu_\mathrm{B} \sum_i \boldsymbol{H}_i \boldsymbol{S}_i = g\mu_\mathrm{B} \boldsymbol{H} \circ \boldsymbol{S}. \tag{2}$$

They do not lead to any first-order perturbation of the $M = 0$ ground state. Thus a formation of local magnetic moments can originate from a second-order perturbation only. The dominant effect is caused by an admixture of the first excited triplet. The z-component of the spin operators $S_{i,z}$ mixes the ground state with the excited $M = 0$ state while the operators $S_{i,+}$ and $S_{i,-}$ lead to an admixture of $M^2 = 1$ states. The difference of the energy splittings leads to strong anisotropy of the generalised magnetic susceptibility (see below).

For small perturbations the ground state energy changes with the square of the matrix element

$$p = \langle \psi_\mathrm{a} | \mathcal{H}_\mathrm{p} | \psi_\mathrm{s} \rangle = g\mu_\mathrm{B} \sum_i \boldsymbol{H}_i \langle \psi_\mathrm{a} | \boldsymbol{S}_i | \psi_\mathrm{s} \rangle \equiv g\mu_\mathrm{B} S_\mathrm{ch} (\boldsymbol{H} \circ \boldsymbol{\sigma}). \tag{3}$$

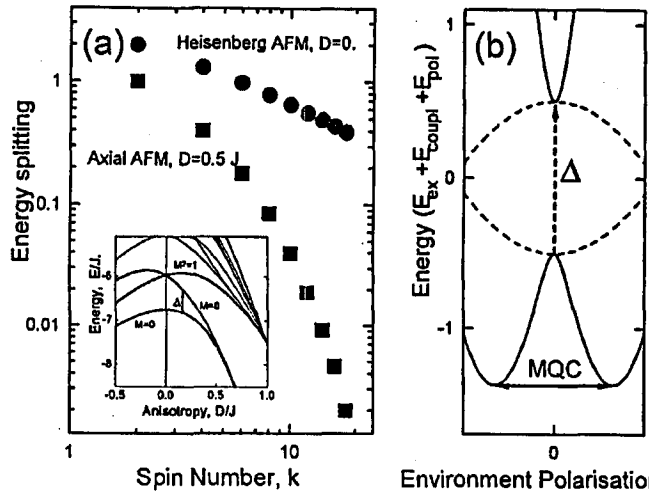

Fig. 1. (a) The dependence of the excitation energy of AFM chain on the cluster size. The inset shows variation of the energy structure of an AFM chain $N = 10$, $S = 1/2$ on the anisotropy parameter D. (b) Total energy of AFM cluster (sum of exchange, coupling to environment and environment polarisation energies) as a function of an environment polarisation.

Simultaneously, the local moments, μ_{ch}, are induced. Their characteristic spatial distribution, is determined by a normalised vector σ as defined by Eq. (3) to be proportional to the matrix elements between the singlet ground and the excited states. The induced magnetisation

$$\mu_{ch} = -g\mu_B\langle S_i\rangle = \chi_g(\sigma \circ H)\sigma = \chi_g H_p \sigma \tag{4}$$

is independent on the spatial distribution of perturbing fields but on the projection of the local fields on the characteristic vector $H_p = H \circ \sigma$ only. Since the ground singlet state differs from the first excited triplet by the parity symmetry (due to spatial cluster symmetry) the characteristic vector, σ, is antisymmetric. Thus only an antisymmetric component of the local field leads to the formation of μ_{ch}. For homogeneous fields H_p vanishes. The generalised susceptibility, χ_g, is determined by the second derivative of the energy gain $\Delta E = p^2/\Delta$, over the local field amplitude, H_p. It increases with an increase in the cluster size thus the big AFM cluster becomes unstable. A small fluctuation can lead to the formation of a static staggered magnetisation, μ_{ch}, i.e. to transition from QM to classical AFM.

Most interesting is the case if there is a coupling of magnetic moments, μ_{ch}, to an environment and $\mu_{ch}(\pi)$ can induce the environment polarisation, $\pi(\mu_{ch})$ (e.g. the hyperfine interaction or a spin coupling to another local moments). In such a case, the total energy of the system (see Fig. 1b) consists of (i) the energy of QM states, (ii) the energy needed for formation of the staggered magnetisation, (iii) the coupling energy, and (iv) energy needed for polarisation of the environment. The formation energy (ii) is determined by $\chi_g\mu_{ch}^2 \sim 1/N$. It dominates for small AFM cluster and is shown in Fig. 1b by the dashed line. The coupling

energy (iii) is proportional to $\pi\mu_{ch}(\pi) \sim N$. For big clusters, it leads to the symmetry breakdown, to an induction of the static Néel states and to the formation of a barrier between them. The temperature dependent increase in the ground state energy for big π originates from (iv) the increase in the energy needed for polarisation of the environment.

The probability of the transition between two equivalent minima of the total energy is strongly temperature and size dependent since both the thickness and the height of the barrier increases with cluster size and with temperature decrease. Without any detailed discussion of the type of perturbations which leads to the transition, one can notice that the tunnelling rate (caused by time dependent fluctuations) and the coherence rate (cased by an additional static perturbation which does not commute with the staggered magnetisation) decreases exponentially with the cluster size, i.e., the number of individual spin flips needed for the transition between the minima. Analysing the transition probability one has to consider not only the number and the probability of individual spin flips, but, in general, also the probability of a simultaneous change of the environment polarisation. In the case when the formation of the static staggered magnetisation is caused by a effective hyperfine coupling [5], the transition probabilities are predominantly limited by a small probability of the nuclear spin flips. Then the MQC probably cannot be experimentally observed.

We suppose that the resonance observed by Awschalom [3] corresponds to the energy distance Δ. As is shown in Fig. 1a, Δ decreases exponentially with the size of the cluster which is the most characteristic feature of Awschalom's resonance [1, 6]. The nature of the resonance does not correspond, however, to any precession of a rotator but rather to an oscillation μ_{ch}. Therefore, the excitation of such a resonance is possible by a spatially inhomogeneous field only. Strictly speaking, the transition probability is proportional to the square of the projection of the oscillating field on the characteristic vector.

The oscillating character of small AFM magnetic clusters helps us also to understand other puzzling effects. In particular, neutron diffraction investigations [4] brought evidence of an interlayer correlation between ultrathin AFM layers across intercalated diamagnetic layers of several ML thickness. There is no coupling mechanism which is strong enough to provide a static coupling, but a much weaker interaction between similar AFM layers may provide a dynamic correlation among oscillating resonators. Thermal neutrons are so fast that they monitor only the momentary spin correlations.

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