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SMALL ANTIFERROMAGNETIC CLUSTERS*

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The analysis of the quantum mechanical model of mesoscopic size antiferromagnets shows that small antiferromagnets are characterized by a stronger exchange coupling and by an oscillating character of spins polarizations. Experimental evidence of the quantum character is discussed. We interpret the interlayer coupling in antiferromagnet superlattices and a puzzling resonance observed in nano-size antiferromagnet grains.

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Nano-scale antiferromagnetic (AF) grains, ultrathin layers of AF, or diluted magnetic semiconductors, which under some circumstances can be treated as a random set of AF clusters, exhibit properties which cannot be explained within classical models. Within the classical Néel model, the ground state of an AF is doubly degenerate: two different orientations of the magnetic moment in each of the AF sublattices are possible. The model predicts a non-zero static Néel vector, i.e., a finite magnetization of each of the two sublattices. Within the quantum mechanical (QM) model [1–3], in contrast, the AF cluster of an even number of spins is characterized by a singlet ground state which is built from a superposition of the Néel states. Thus the mean value of the net magnetic moment on each spin site vanishes. The case of a cluster with an odd number of spins is a little bit more complicated because a single spin remains uncompensated. For simplicity reasons, in this paper we will discuss the case corresponding to an even number of spins only. Generally, the classical and the QM model predict very different properties of AF and there is no simple correspondence between small and large AF. In particular, an extrapolation of the QM picture does not predict a static Néel structure of AF. A well settled Néel vector can be expected only when a coupling to an environment, e.g., to the nuclear spins by hyperfine coupling, is taken into considerations.

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We examine the rigorous QM solutions for small AF grains by discussing the spin density distribution and spin correlators. An example of the energy diagram is shown in Fig. 1a. For even spin number, N, the ground state is separated from excited states by an energy gap, Δ. The dependence of Δ on the cluster size is shown in the inset to Fig. 1a. The gap decreases strongly with increasing cluster size, N, for large clusters the gap is inversely proportional to the cluster size [1, 3].

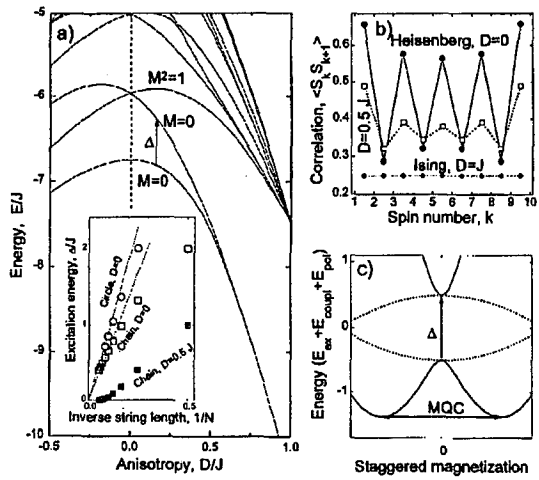

Fig. 1. (a) Energy of an AF chain $N = 8$ $S = 1/2$ spins as a function of the anisotropy parameter Δ. For $\Delta > 0$, two nonmagnetic singlets are separated by the energy Δ. The inset shows the dependence of Δ on the clusters size N. (b) Correlation between neighboring spins (i and $i + 1$) in an $N = 10$ chain of $S = 1/2$ spins for different magnetic anisotropy. A "surface pair" in an Heisenberg AF is coupled much stronger as compared to "volume couples" and to the Ising AF. (c) Total energy of an AF cluster (sum of exchange, coupling to an environment, e.g. a hyperfine coupling, and energy due to polarization of the environment) as a function of staggered magnetization. Dashed lines correspond to the oscillating limit where the mean value of magnetization vanishes and formation of a staggered magnetization is accompanied by an energy increase. We suggest that the vertical arrow corresponds to the Awschalom resonance [5] whereas the horizontal arrow to a macroscopic quantum coherence.

Calculation of the correlation for various pairs of neighboring spins allows to distinguish "the inner volume" from the edges (surface) of AF chains (grains). The distribution of the pair correlation in AF chains is shown in Fig. 1b. The magnetic structure of the "inner" part, which can be determined from an extrapolation of open chains and closed rings for increasing number of spins, is characterized by the same correlators. For Heisenberg $S = 1/2$ chains, $\langle S_i S_{i+1} \rangle = 0.443$, while for Ising chains ($D = J$) it is equal to 0.25.

At the ends of chains (surface of grains) the coupling between subsequent pairs (layers) oscillates. There is a well-evidenced tendency to form strongly coupled pairs which are weakly correlated to the next pairs. Such a tendency is ob-

served as well for even as for odd numbers of spins. Averaging of the exchange correlations leads to the conclusion that the density of exchange energy at the ends of the chain (surface of grains) is bigger as compared to the energy density in the inner volume. As a consequence, smaller systems are characterized by a stronger exchange energy. This effect originates from the fact that the "dangling" spins at the surface have a better chance for optimum antiparallel orientation of every spin component. The effect vanishes for an Ising AF ($D/J = 1$) where only z-components of spins are anticorrelated.

The rigorous QM calculations for 2D [2,3] and 3D are very difficult — only small clusters can be evaluated. The experimental data [4], however, show that in ultrathin AF layers, where spins within each monolayer are ferromagnetically correlated, an increase in the exchange energy is observed. In spite of a spin diffusion at the interface, the Néel temperature in EuTe superlattices is by factor 2 bigger than in bulk crystals. Thus we expect that the QM effect of increasing the exchange energy does not depend directly on the dimension but it is governed rather by the ratio of (dangling) surface spins to volume ones. From that point of view, the strongest effect is expected for diluted AF which can be treated as a random set of various clusters. In diluted AF with a frustrated AF structure the QM increase in the exchange energy acts together with the fact that a dilution reduces the frustration level.

The two ground singlets play a crucial role in the character of an AF. Both of them are characterized by $M = 0$ and at each of these states the mean values of every spin vanish: there is no magnetization of the Néel sublattices. They differ by parity symmetry — thus a transition is generally forbidden and no classical magnetic resonance can be observed. On the other hand, the energetic distance Δ corresponds to zero-point oscillations of the staggered magnetization — the Néel vector oscillates with the frequency Δ/h around its zero value. As a consequence, a small AF can be treated as an oscillator with a characteristic frequency which decreases with the size of AF. Excitation of these oscillations is difficult since only spatially inhomogeneous external fields lead to a finite transition probability. Nevertheless, such a resonance has been observed by Awschalom et al. in small AF grains [5] in the MHz frequency range.

Another indication of the oscillating character of quasi-2D AF layers has been found by neutron diffraction [6] which demonstrates efficient correlations between spins from AF layers (MnTe, EuTe) separated by a diamagnetic spacer (CdTe, PbTe). There is no interlayer coupling which is strong enough to provide any static interlayer correlation. But a weak interlayer, e.g. magnetic dipole, coupling is sufficient to explain a correlated phase of oscillations of two similar resonators. The expected characteristic frequency for quasi-2D layers is 10^9–10^{11} s^{-1}. Thus the characteristic period is longer than the time the neutron takes to pass through the layers which is about 10^{-12} s. Therefore, neutrons monitor a momentary picture of the orientations of the oscillating spin sublattices.

Calculations for the AF cluster show that the $M = 0$ singlet state remains the ground state of the system for any cluster size but the energy distance between two ground singlets decreases with increasing cluster size. When the cluster is big enough and the coupling energy of AF spins to their environment becomes com-

parable to the excitation energy Δ then, because of a mixing of symmetric and antisymmetric states, the oscillating character is suppressed and a stable staggered magnetization appears [7]. Anyway, to reach the classical limit of AF, where local spins are characterized by a nonvanishing magnetization, an influence of additional (staggered or, generally, spatially nonhomogeneous) local fields is necessary. The hyperfine coupling $\boldsymbol{AIS}$ of AF spins S to the nuclear spins I suppresses the quantum oscillation when the total hyperfine energy, $NAS\langle I\rangle$, exceeds the energy distance $\Delta \sim J/N$, i.e., for $N^2 > J/AS\langle I\rangle$. In Fig. 1d the energy splitting Δ is plotted as a function of the staggered polarization of nuclear spins which is induced and proportional to the static staggered magnetization of electronic spins. At zero temperature, MnTe strings reach the classical limit already for $N = 30$ spins. With increasing temperature the polarization of nuclear spins decreases and the classical limit occurs for longer strings. The open question is the estimation of the classical limit for 2D and 3D AFM. The experimental data [4, 6] suggest, however, that quasi 2D layers where one dimension is reduced to a few monolayers exhibits already a quantum character of AF.

The transition to the classical limit, i.e., the formation of a static staggered magnetization, increases the frequency of the Awschalom resonance [5] (see Fig. 1c) but simultaneously the transition probability decreases. In that sense, the observation of the resonance in AF systems reflects the quantum character of these systems. Also the dynamic interlayer coupling [6] proves the quantum character of AF layers. The stabilization of the sublattices kills the zero point motion of the staggered magnetization. In mesoscopic system there is a hypothetical possibility of a quantum transition between two minima. Macroscopic quantum tunneling (MQT) or macroscopic quantum coherence (MQC) has been discussed [3, 5, 8] as a possible interpretation of the Awschalom resonance. According to our knowledge, however, there is no experimental evidence of such a transition. Figure 1c demonstrates the difference between MQC and the resonance.

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