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MACROSCOPIC QUANTUM TUNNELING IN SMALL MAGNETIC PARTICLES

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Macroscopic quantum tunneling of magnetization in small antiferromagnetic clusters is studied for the spin-1/2 anisotropic ($J_z \equiv J$, $J_x = J_y \equiv J_{xy}$) Heisenberg model employing both perturbation theory and exact numerical diagonalization technique. Splitting between the two lowest levels due to the lifting of degeneracy between them (occurring for $J_{xy} = 0$) by the in-plane exchange ($J_{xy} \neq 0$) is found to occur in the $N/2$-th order of the perturbation theory. The tunneling is absent in systems with odd number of sites where all levels are doubly degenerate in zero magnetic field.

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In the last few years the phenomenon of macroscopic quantum tunneling (MQT) has been attracting much interest from the viewpoint of both fundamental physics and technology [1]. Small magnetic particles are the suitable candidates for the observation of macroscopic quantum tunneling of the magnetization [2–4]. From the viewpoint of applications, the interest to MQT is explained by the general trend of miniaturization of the magnetic recording devices. The fundamental question in this respect is about the ultimate possible reduction of the information storage device size where macroscopic quantum fluctuations still do not destroy written information [1].

Most frequently quantum tunneling in small magnetic particles is considered within the following framework [5–9]. A particle is assumed to possess a single-domain magnetic structure (ferro- or antiferromagnetic) characterized by the easy-axis anisotropy. When minimizing the system energy, the spin is considered to be large (to behave classically) [5–9] and the energy minima are found for the classical magnetic moment. In doing so it is in fact assumed that the interaction aligns the spins of all electrons along the same direction so that the equilibrium magnetization can be described by a single classical vector which has several (usually two) orientations corresponding to the minimum energy; as a next step the magnetization vector dynamics in the arising double-well potential is described by quantum mechanical laws [5–9]. The rate of quantum tunneling is assumed to be determined by the probability of tunneling through the barrier between the two wells. Such a coherent magnetization tunneling results in splitting of the doubly degenerate ground states corresponding to the minimum of classical energy. The splitting is usually calculated in the saddle point approximation (instanton approximation) [5–9].

For a number of models it was found that the quantum tunneling is only possible for a system with integer total spin [9] and is absent in the case of a half-integer spin.

Below we shall use the exact numerical diagonalization technique and the perturbation theory applied the spin-1/2 Heisenberg model to show that MQT in some sense does exist in antiferromagnetic systems and is actually nothing else than the usual quantum beatings. In this approach description of MQT does not require the complicated instanton language [5–9].

We shall first consider the system with even number N of sites (total number of states being 2^N) described by a spin-1/2 Heisenberg Hamiltonian possessing an easy-axis anisotropy. The system Hamiltonian has the form

$$\hat{H} = J \sum_{<ij>} \hat{\sigma}_{zi}\hat{\sigma}_{zj} + \frac{J_{xy}}{2} \sum_{<ij>} (\hat{\sigma}_{xi}\hat{\sigma}_{xj} + \hat{\sigma}_{yi}\hat{\sigma}_{yj}) = \tag{1}$$

$$= J \sum_{<ij>} \hat{\sigma}_{zi}\hat{\sigma}_{zj} + J_{xy} \sum_{<ij>} (\hat{\sigma}_{+i}\hat{\sigma}_{-j} + \hat{\sigma}_{-i}\hat{\sigma}_{+j})$$

with the antiferromagnetic exchange $(J, J_{xy} > 0)$. Summation in (1) is performed over the nearest neighbours. The full basis consists of the vectors

$$\{|\sigma_1\sigma_2 \ldots \sigma_N >\} = |\uparrow\uparrow\uparrow \ldots \uparrow>, |\downarrow\uparrow\uparrow \ldots \uparrow>, \ldots |\uparrow\downarrow\uparrow\downarrow \ldots>, |\downarrow\uparrow\downarrow\uparrow \ldots>, \ldots |\downarrow\downarrow\downarrow \ldots \downarrow> . \tag{2}$$

The system eigenenergies and eigenfunctions are determined by diagonalizing the Hamiltonian (1). For definiteness we shall consider an antiferromagnetic ring (closed chain).

In the basis (2) the Hamiltonian (1) has a block-diagonal form since it commutes with the total spin component along the z-axis S_z, the size of each block being determined by the number of possible permutations of up and down spins. For example, for a ring we have

$$\hat{H} = \begin{pmatrix} \boxed{NJ} & & & & \\ & \begin{array}{|c|c|}\hline \cdots & J_{xy} \\ \hline J_{xy} & \cdots \\ \hline \cdots & \cdots \\ \hline \end{array} & & & \\ & & \begin{array}{|c|c|c|}\hline -NJ & J_{xy} & \cdots \\ \hline J_{xy} & \cdots & \cdots \\ \hline \cdots & \cdots & J_{xy} \\ \hline \cdots & J_{xy} & -NJ \\ \hline \end{array} & & \\ & & & \begin{array}{|c|c|}\hline \cdots & J_{xy} \\ \hline J_{xy} & \cdots \\ \hline \cdots & \cdots \\ \hline \end{array} & \\ & & & & \boxed{NJ} \end{pmatrix} \tag{3}$$

The first and last blocks each contain a single state which is not coupled to any other states. Of special interest is the block corresponding to $S_z = 0$, i.e. a half of electrons have spin up and a half spin down. This block contains the two states $|\uparrow\downarrow\uparrow\downarrow \ldots >$ and $|\downarrow\uparrow\downarrow\uparrow \ldots >$ corresponding to the two standard antiferromagnetic ground states. If the in-plane exchange interaction is neglected $(J_{xy} = 0)$, these two states are degenerate and possess the minimum energy. All the other states are at least J higher in energy. In the case of small J_{xy} one can use the perturbation theory for degenerate levels [10]. For $N > 2$ the states $|\uparrow\downarrow\uparrow\downarrow \ldots >$ and $|\downarrow\uparrow\downarrow\uparrow \ldots >$ are not directly coupled to each other.

The secular equation in the subspace spanned by these two states

$$|\Psi > = c_1|\Psi_1^{(0)} > + c_2|\Psi_2^{(0)} > = c_1|\uparrow\downarrow\uparrow\downarrow \ldots > + c_2|\downarrow\uparrow\downarrow\uparrow \ldots > \tag{4}$$

has the form

$$\begin{pmatrix} E - E_1^{(0)} + V_1 & \Delta \\ \Delta & E - E_2^{(0)} + V_2 \end{pmatrix} \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = 0, \tag{5}$$

where $E_{1,2}^{(0)} = -NJ$ is the unperturbed doubly degenerate level.

It is easily seen that because of spin reversal symmetry $V_1 = V_2 \equiv V$ in all orders of the perturbation theory. The leading terms in V are quadratic in J_{xy}. Indeed, e.g. for the state $|\uparrow\downarrow\uparrow\downarrow\ldots>$ we have

$$V = \sum_m \frac{V_{1m} V_{m1}}{E_1^{(0)} - E_m^{(0)}} = \frac{J_{xy}^2}{J} f(N), \tag{6}$$

where V_{1m} is the perturbation matrix element

$$V_{1m} = J_{xy} \sum_{<ij>} < \Psi_1^{(0)}| \sum_{<ij>} (\hat{\sigma}_{+i}\hat{\sigma}_{-j} + \hat{\sigma}_{-i}\hat{\sigma}_{+j})|\sigma_1\sigma_2\ldots>,$$

where $|m> \equiv |\sigma_1\sigma_2\ldots>$ is one of the states with energy $E_m^{(0)}$ in the same block which differs from $|\uparrow\downarrow\uparrow\downarrow\ldots>$ by the reversal of a pair of interacting (nearest-neighbour) spins, and $f(N)$ is a function of N only. Thus, the shift of the levels is mainly quadratic in J_{xy}.

Off-diagonal matrix elements can be written as

$$\Delta = \sum_{m_1,m_2,m_3\ldots m_{\frac{N}{2}}} \frac{V_{1m_1} V_{m_1m_2} V_{m_2m_3} \ldots V_{m_{\frac{N}{2}}2}}{(E_1^{(0)} - E_{m_1}^{(0)})(E_{m_1}^{(0)} - E_{m_2}^{(0)})(E_{m_2}^{(0)} - E_{m_3}^{(0)})\ldots(E_{m_{\frac{N}{2}}}^{(0)} - E_2^{(0)})} . \tag{7}$$

Starting from the state $|\uparrow\downarrow\uparrow\downarrow\ldots>$, one can reach the state $|\downarrow\uparrow\downarrow\uparrow\ldots>$ through the intermediate states each of which differs from the preceding one by the reversal of a pair of interacting spins. Such a transition cannot involve less than $N/2$ intermediate states. Therefore, on the order of magnitude

$$\Delta \sim J\,(J_{xy}/J)^{N/2}\, f_\Delta(N), \tag{8}$$

where $f_\Delta(N)$ is a function of N only (we cannot estimate $f_\Delta(N)$ for arbitrary N).

The two states have the energies

$$E_\pm = -NJ - V \pm |\Delta|, \tag{9}$$

the splitting between them

$$|\Delta| = J f_\Delta(N) \left|\frac{J_{xy}}{J}\right|^{N/2} \sim J \exp\left(-\frac{N}{2}\ln\left|\frac{J}{J_{xy}}\right|\right), \tag{10}$$

depending exponentially on the system size which is in agreement with the instanton approach [5–9] (although the dependence on the anisotropy in the exponent is logarithmic rather than power).

We have also numerically diagonalized the Hamiltonians for the antiferromagnetic rings with $N = 4, 6, 8,$ and 10 sites. The shift of two lowest level as a function

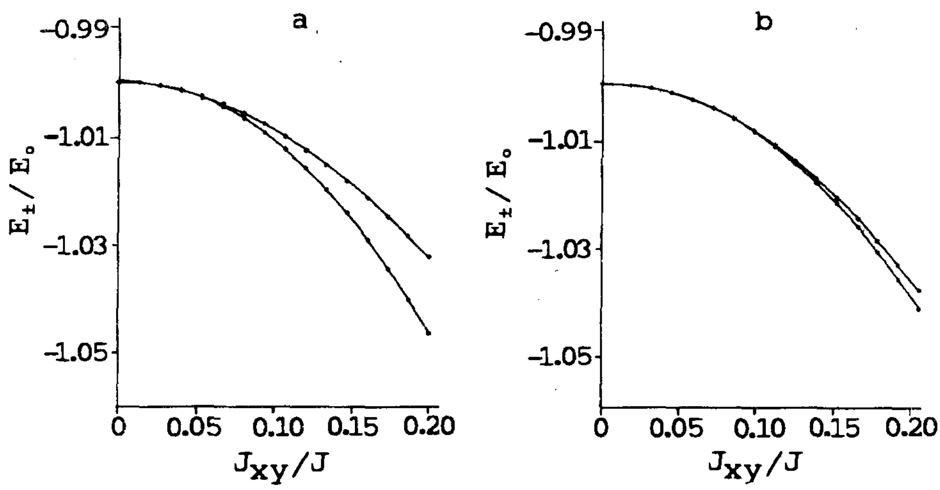

Fig.1. Energy of the two lowest levels $E_\pm(J_{xy})$ normalized to $E_0 = |E_-(J_{xy} = 0)| = |E_+(J_{xy} = 0)|$ for $N = 6$ (a) and 8 (b). Dots are numerical results and the lines are quadratic splines

of J_{xy} is shown in Fig.1. It is seen that the shift is rather well described by a parabolic curve. The splitting scales as $(J_{xy}/J)^{N/2}$ (Fig.2).

The wave functions of the two lowest levels found from Eqs. (5) and (9) are:

$$|\Psi_\pm> = \frac{1}{\sqrt{2}}(|\Psi_1^{(0)}> \pm |\Psi_2^{(0)}>) = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\uparrow\downarrow\dots> \pm |\downarrow\uparrow\downarrow\uparrow\dots>), \tag{11}$$

so that the quantum mechanical average spin on each site is zero in both states.

Going back to the ideology of Refs. [5–9], one can prepare the system in the state $|\uparrow\downarrow\uparrow\downarrow\dots>$ corresponding to one of the two minimums of the system energy [5–9] which is not an eigenstate. The staggered magnetization in this state $\hat{S}_\Sigma = -\sum_i (-1)^i \hat{S}_{zi}$ (here $\hat{S}_{zi} = \sigma_z/2$) is

$$\overline{S}(0) = <\Psi(0)|\hat{S}_\Sigma|\Psi(0)> = N/2. \tag{12}$$

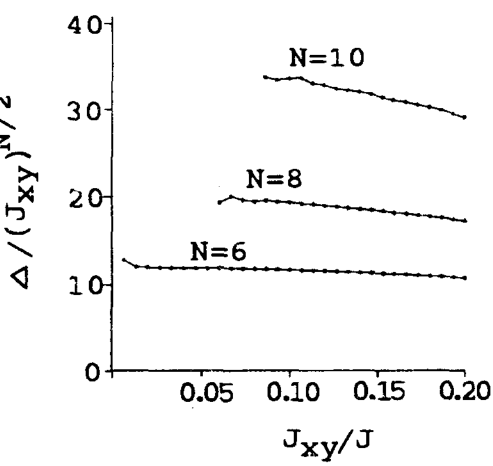

Fig.2. Splitting of the two lowest levels as a function of the in-plain exchange parameter

The system evolution is described by equations

$$|\Psi(t)>=\exp(-i\hat{H}t)|\Psi(0)>, \quad \overline{S}(t)=<\Psi(t)|\hat{S}_\Sigma|\Psi(t)>. \tag{13}$$

Expanding the state $|\Psi(0)>$ in the Hamiltonian eigenvectors and taking into account Eq.(11) one obtains

$$\overline{S}(t) \propto \cos(\Delta t) + \text{the terms with frequencies } \omega \sim J \gg \Delta, \tag{14}$$

i.e. ordinary quantum-mechanical interference in a two-level system occurring if the initial state is not an eigenstate. The low-frequency term with $\omega = \Delta/J$ corresponds to the interference between the two lowest states. The frequency $\omega = \Delta/J$ can be interpreted as the energy of tunnel splitting between the two doubly degenerate states $|\Psi_{1,2}^{(0)}>$ with energy $E_{1,2}^{(0)} = -NJ$. The tunneling itself arises only due to the in-plane exchange. The low-frequency oscillation with $\omega = \Delta/J$ is modulated with the high-frequency oscillations with frequencies $\omega \sim J$, corresponding to the admixture of the excited states.

However, actually it is hardly possible to prepare an interacting system in an arbitrary state. The low-frequency tunneling mode should manifest itself in the fluctuation spectrum of the system, i.e. in the frequency dependence of the corresponding susceptibility. According to the FDT-theorem [11], the imaginary part of the generalized susceptibility for the operator $\hat{S}_\Sigma$ can be written as

$$\overline{\hat{S}^2_{\Sigma\omega}} = \hbar\chi''_{zz}(\omega)\coth(\omega/2T), \tag{15}$$

$$\chi''_{zz}(\omega) = \frac{\pi}{\hbar}\sum_{\beta=+,\text{other excited states}} |<\Psi_\beta|\hat{S}_\Sigma|\Psi_->|^2[\delta(\omega+\omega_\beta)-\delta(\omega-\omega_\beta)]. \tag{16}$$

The sum in Eq. (10) is taken over all the excited states including E_+. The contribution we are interested in at the frequency $\omega = \Delta/\hbar$ comes from the term $|<\Psi_+|\hat{S}_\Sigma|\Psi_->|^2$. Taking into account that

$$\hat{S}_\Sigma|\Psi_->=\left(-\sum_{i=1}^{N}(-1)^i\hat{S}_{zi}\right)(|\uparrow\downarrow\uparrow\downarrow\ldots>-|\downarrow\uparrow\downarrow\uparrow\ldots>)/\sqrt{2}=\frac{N}{2}|\Psi_+>, \tag{17}$$

one obtains the coherent contribution

$$\chi''_{zz}(\omega) \propto \delta(\omega - \Delta/\hbar). \tag{18}$$

The δ-function singularity in the fluctuation spectrum corresponds to a collective mode with the frequency $\omega \sim \Delta/\hbar$, which can be interpreted as the quantum tunneling of the magnetization. For large N all the rest non-coherent oscillations yield an almost continuous frequency spectrum.

Unfortunately, it is not clear whether the fluctuation spectrum of $\hat{S}^z_\Sigma$ can be directly measured experimentally. The quantum nature of the singularity in $\chi''_{zz}(\omega)$ will manifest itself in its presence at low (compared with J) temperatures $T \sim \Delta \ll J$. As the in-plane exchange is increased, the low-frequency mode is expected to become less and less prominent until the coherent spin evolution of the system is completely destroyed.

Suppose now that the number of states is odd. In that case all the energy levels are at least doubly degenerate (Kramers degeneracy), which is easily seen directly from the Hamiltonian matrix (3) containing now pairs of identical blocks (all corresponding to non-zero S_z values) mapped to each other by the spin reversal of all electrons. Therefore, there is no low-frequency mode in the system which means the absence of MQT. It is also interesting to note that the tunneling can be absent even in an antiferromagnetic cluster with even number of sites if its geometry is such that the ground state (in the limit $J_{xy} = 0$) has non-zero total spin S_z, and consequently, is doubly degenerate (it is easily seen that such clusters can be constructed for any $N \geq 4$). Here again because of the spin-reversal symmetry the degeneracy between these two states cannot be lifted by $J_{xy} \neq 0$.

In the ferromagnetic system the ground state is formed by two degenerate functions $|\uparrow\uparrow\uparrow\ldots\uparrow>$ and $|\downarrow\downarrow\downarrow\ldots\downarrow>$ with the energy $-N|J|$ $(J < 0)$ independent of the parity of the total number of sites. These states cannot be split by the in-plane exchange so that the MQT is also prohibited in this case.

Note that the spin dynamics of a ferromagnetic cluster has recently been studied numerically [12] for the spin-1/2 Heisenberg model in a uniform constant magnetic field. However, the coherent oscillations of the system magnetization found in Ref.[12] seem to be related to the ordinary spin resonance and have nothing to do with the macroscopic quantum tunneling discussed in Refs.[5–9].

It should also be noted that there is currently much interest in the possibility of realization of the so-called single-electron spin logical gates where the bit values 0 and 1 are associated with the electron spin direction at a particular site [13–15]. Computations are preformed by acting on the input sites of the gate by local magnetic fields. One could expect that the quantum fluctuations are able to destroy the spin configuration representing the results of calculations. However, it is seen from the above results that for a sufficiently large system ($N \gg 1$) the spin configurations created by the external fields can be rather stable, the probability of a spontaneous transition to another state being exponentially small.

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