Quantum coherence oscillations in antiferromagnetic chains

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Macroscopic quantum coherence oscillations of the Néel vector are proved to appear in the antiferromagnetic nuclear spin chain of eight ^{129}Xe atoms placed on a magnetic surface. The oscillation period at zero temperature is calculated numerically as a function of the chain constant. The environmental decoherence effects at finite temperature are accounted assuming a dipole coupling between the spin chain and the fluctuating magnetic field of the surface. With this coupling the Néel vector oscillations are damped by a rate $(N-1)/\tau$, where N is the number of spins and τ is the relaxation time of a single spin. [S0163-1829(99)08909-2]

The observation of macroscopic quantum coherence (MQC) phenomena in complex many-particle systems represents a subject of wide interest, ranging from the conceptual foundations of quantum mechanics,[1] to the physics of the microelectronic devices. During the last years particular attention was given to the macroscopic quantum tunneling and quantum coherence oscillations.[2] In these phenomena the quantum dynamics of a prepared nonstationary wave packet is directly reflected by the non-classical behavior of a macroscopic observable. However, it is difficult to identify physical situations where such coexistence between the classical and quantum aspects could be observed.

Quantum coherence oscillations may occur in the localization probability of relatively complex systems as individual atoms trapped in the surface-tip junction of the scanning tunneling microscope.[3–5] Particularly suitable candidates to observe MQC phenomena are also the magnetic systems.[6] In the anisotropic antiferromagnets the Néel vector may change the orientation by quantum tunneling[7] or quantum coherence oscillations.[8] The observation of these oscillations still represents a challenging problem,[9] but important results have been obtained from the measurements of the AC magnetic susceptibility in antiferromagnetic (AF) ferritin.[10,11]

The study of the non-elementary excitations in AF systems is essential also for understanding the high-T_c superconductivity.[12,13] Therefore, a detailed study of the MQC phenomena in antiferromagnets appears highly interesting.

The purpose of this paper is to study the MQC oscillations of the Néel vector for an anisotropic AF chain of spins 1/2. The Hamiltonian corresponds to the system of nuclear spins for eight atoms of the ^{129}Xe isotope placed on a magnetic surface. Such chains can be constructed, for instance, using the scanning tunneling microscope in the ''atomic switch'' operation mode, proved during the last years to be an efficient instrument to displace in a controlled way the Xe atoms on a Ni surface.[14] The interaction between spins is due to the magnetic dipole forces, and the effect of the surface is taken into account using the simple method of images. The spins $\vec{\mathbf{I}}_i$, $i=1,8$ are supposed equally spaced by the chain constant d, and at zero temperature the model Hamiltonian is

$$H_0 = J\sum_{i=1}^{7} [c_x \mathbf{I}_{i,x}\mathbf{I}_{i+1,x} + c_y \mathbf{I}_{i,y}\mathbf{I}_{i+1,y} + c_z \mathbf{I}_{i,z}\mathbf{I}_{i+1,z}]. \quad (1)$$

Here, $J>0$ is the coupling strength and c_x, c_y, c_z are the anisotropy coefficients. The coordinate system has the z axis normal to the surface and the y axis along the chain. Each magnetic moment $\vec{m}=(m_x,m_y,m_z)$ lying above the surface at $z=r_0$ has an image $\vec{m}'=(-m_x,-m_y,m_z)$ at $z=-r_0$, where $r_0=2.17$ Å is the radius of the Xe atom. The nearest neighbors of $\vec{m}$ interact also with $\vec{m}'$, and the parameters of H_0 are $J=\mu_0\hbar^2\gamma^2/(4\pi d^3)$ and $c_x=1-2\sin^3\alpha$, $c_y=-2-2\sin^3\alpha(1-3\sin^2\alpha)$, $c_z=1+2\sin^3\alpha(1-3\cos^2\alpha)$, where $\tan\alpha=d/2r_0$, $\mu_0=4\pi 10^{-7}N/A^2$ is the vacuum permeability and $\gamma=-1.54\mu_N/\hbar$ the gyromagnetic factor of the ^{129}Xe isotope.

Classically, the AF ordering of a spin chain $\{\mathbf{I}_1,\mathbf{I}_2,\ldots,\mathbf{I}_N\}$, is described in terms of two subchains, containing the odd and the even spins, respectively. The magnetization vectors of these subchains are $\vec{M}_o=\gamma\hbar\Sigma_{i=odd}\vec{\mathbf{I}}_i$ and $\vec{M}_e=\gamma\hbar\Sigma_{i=even}\vec{\mathbf{I}}_i$. The chain is antiferromagnetic when $\vec{M}_o$ and $\vec{M}_e$ have the same magnitude M_0, but a relative antiparallel orientation. The energy in this case is a function of the Néel vector $\vec{n}=(\vec{M}_o-\vec{M}_e)/2M_0$, and is expressed by

$$E^A(\vec{n}) = -J(N-1)(c_x n_x^2 + c_y n_y^2 + c_z n_z^2)/4. \quad (2)$$

An anisotropic system with $c_{x,y}<c_z$ has two degenerate minima, $E^A_{min}=-J(N-1)c_z/4$ attained when $\vec{n}$ has the two possible orientations along the z axis, $\vec{n}=\pm\vec{e}_z$. These minima are separated by a two-dimensional potential barrier with the maximum $E^A_{max}(\phi)=-J(N-1)(c_x\cos^2\phi + c_y\sin^2\phi)/4$, in the x-y plane, where ϕ denotes the angle between the Néel vector and the x axis.

The spectrum of H_0 was calculated for 6.8 Å$<d<$9.4 Å by solving numerically the eigenvalue equation

$$H_0|\psi_n\rangle = E_n|\psi_n\rangle. \quad (3)$$

The basis was defined by the common eigenstates for the z components of all the spin operators $\mathbf{I}_{i,z}$, $i=1,8$. These basis states are denoted by $|k\rangle \equiv |m_1^k,m_2^k,m_3^k,m_4^k,m_5^k,m_6^k,m_7^k,m_8^k\rangle$, $k=1,2^8$, with $m_i^k=\pm 1/2$. The eigenstates of H_0 have the general form $|\psi_n\rangle=\Sigma_k y_k^n|k\rangle$. It is found that the ground $(n=0)$, and the first excited state $(n=1)$, have the largest overlap with only two

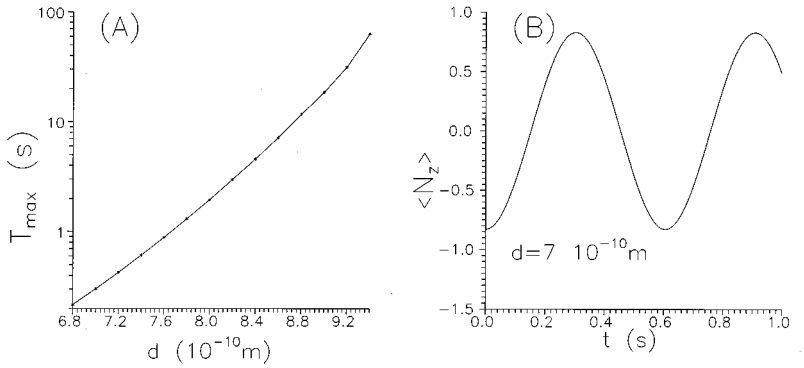

FIG. 1. T_{max} as a function of the chain constant d (A) and the expected value $\langle N_z \rangle$ as a function of time when $d=7$ Å (B).

basis states, which are antiferromagnetic in the classical sense. These basis states are denoted $|\uparrow\rangle$ and $|\downarrow\rangle$, and are eigenstates of z component of the Néel operator $\vec{N}=\Sigma_{i=1,4}(\vec{\mathbf{I}}_{2i-1}-\vec{\mathbf{I}}_{2i})/4$ with eigenvalues $+1$ and -1, respectively.

Detailed results are presented for the particular value $d=7$ Å, when $J=0.17\hbar$/ms and the anisotropy coefficients are $c_x=-0.22$, $c_y=-0.58$, and $c_z=1.2$. The overlap amplitudes between the eigenstates $|\psi_0\rangle$, $|\psi_1\rangle$ and $|\uparrow\rangle$, $|\downarrow\rangle$ are $\langle\uparrow|\psi_0\rangle=\langle\downarrow|\psi_0\rangle=0.57$, $-\langle\uparrow|\psi_1\rangle=\langle\downarrow|\psi_1\rangle=0.63$. Thus, the two classical AF states contribute by more than 65% to the norm of the eigenstates. The eigenvalues $E_0=-0.406\hbar$/ms and $E_1=-0.395\hbar$/ms are separated by $\Delta=E_1-E_0=0.011\hbar$/ms, which is sensibly smaller than $E_2-E_1=0.053\hbar$/ms. The small value of Δ shows that the system has a quasidegenerate ground state. This appears as a ''tunneling doublet'' determined by the two-dimensional potential barrier $E^A_{max}(\phi)=(0.066\cos^2\phi+0.17\sin^2\phi)\hbar$/ms separating the AF energy minima of $E^A_{min}=-0.353\hbar$/ms.

The tunneling by MQC oscillations appears for the nonstationary wave packets prepared at the ground-state energy with a well defined AF configuration. States having these properties are represented by the linear combinations

$$|\psi_\downarrow\rangle=\frac{1}{\sqrt{2}}(|\psi_0\rangle+|\psi_1\rangle),\quad |\psi_\uparrow\rangle=\frac{1}{\sqrt{2}}(|\psi_0\rangle-|\psi_1\rangle) \quad (4)$$

of $|\psi_0\rangle$ and $|\psi_1\rangle$. If the system is prepared at $t=0$ in the state $|\psi_\downarrow\rangle$, then at the moment t it will be found in the state $|\psi_\uparrow\rangle$ with the probability $\mathcal{P}_\uparrow(t)=\sin^2(\pi t/2T_{max})$, where $T_{max}=\hbar\pi/\Delta$. The half period of oscillation T_{max} is represented as a function of the chain constant d in Fig. 1(A). For $d=7$ Å the expectation values

$$\langle N_\mu\rangle(t)=\langle\psi_\downarrow|e^{iH_0t/\hbar}N_\mu e^{-iH_0t/\hbar}|\psi_\downarrow\rangle \quad (5)$$

are $\langle N_x\rangle(t)=0$, $\langle N_y\rangle(t)=0$, and $\langle N_z\rangle(t)=\langle\psi_0|N_z|\psi_1\rangle\cos(\pi t/T_{max})$, with $\langle\psi_0|N_z|\psi_1\rangle=-0.83$ and $T_{max}=0.3$ s [Fig. 1(B)].

It is important to emphasize the extreme sensitivity of the MQC resonance oscillations with respect to the preparation of the initial state. The expected value of the energy in the classical antiferromagnetic states, $\langle\downarrow|H_0|\downarrow\rangle$ and $\langle\uparrow|H_0|\uparrow\rangle$, is $E^A_{min}=-0.353\hbar$/ms, higher than $E_1=-0.39\hbar$/ms by $\approx 4\Delta$. This energy is far outside the interval $[E_0,E_1]$, and therefore between the classical antiferromagnetic states $|\downarrow\rangle$ and $|\uparrow\rangle$ there are no MQC oscillations.

At finite temperature the MQC oscillations are damped by the coupling between the spin chain and the fluctuating magnetic field of the surface, created by the phonon modulation of the crystalline electric field and the lattice spin waves.[15] This coupling will be described by the residual interaction term

$$H_r(t)=-\gamma\hbar\sum_{i=1,8}\vec{\mathbf{I}}_i\cdot\vec{B}^e_i(t), \quad (6)$$

where $\vec{B}^e_i(t)$ denotes the fluctuating external field at the site i of the chain. At low temperatures the typical wave length of the surface phonons and magnons[16] is ~ 400 Å, much larger than the length of the chain $L=49$ Å. Therefore, $\vec{B}^e_i$ will be considered to be the same for all spins, $\vec{B}^e_i(t)\equiv\vec{B}^e(t)$. With these approximations the residual interaction term becomes $H_r(t)=-\gamma\hbar\vec{B}^e(t)\cdot\vec{\mathbf{I}}$, where $\vec{\mathbf{I}}=\Sigma_{i=1,8}\vec{\mathbf{I}}_i$.

The ''atomic switch'' experiments[14] have been performed at the environmental temperature $T=4$ K, when the thermal energy $k_BT=0.34$ meV is very high compared both to the tunnel splitting $\Delta=6.83\times10^{-12}$ meV and to the maximum barrier height $V_B=E^A_{max}(\pi/2)=0.11\times10^{-9}$ meV. Therefore, the thermal environment can be considered as classical, and the field components $B^e_\mu(t)$, $\mu=x,y,z$, will be treated as white noise with zero mean. The normalization of this fluctuating field is ensured by the fluctuation-dissipation theorem[17] (FDT) $\langle\langle B^e_\mu(t)B^e_{\mu'}(t')\rangle\rangle=\delta_{\mu,\mu'}\delta(t-t')/(\gamma^2\tau)$. Here, $\langle\langle\cdots\rangle\rangle$ denotes the average over the statistical ensemble describing the environment, and the parameter τ is supposed to depend on temperature as $1/T$.

For a single spin of the chain, $H_r(t)$ induces transitions between the states $|1/2\rangle$ and $|-1/2\rangle$ with a rate[18] $\lambda=(|\langle-1/2|\mathbf{I}_x|1/2\rangle|^2+|\langle-1/2|\mathbf{I}_y|1/2\rangle|^2)/\tau=1/2\tau$. Therefore, the relaxation rate of the population difference $n_{1/2}-n_{-1/2}$ is $2\lambda=1/\tau$, showing that τ has the meaning of spin-surface relaxation time.

The damping of the MQC oscillations cannot be treated by using a similar two-level approximation, because the matrix elements of H_r within the subspace generated by $|\psi_0\rangle$ and $|\psi_1\rangle$ are 0. The operators $\mathbf{I}_x$ and $\mathbf{I}_y$ contained in H_r act on the initial state $|\psi_\downarrow\rangle$ by flipping the individual spins, and in time the quantum state aquires components over the whole spectrum. This process is described in principle by a transport equation for the density matrix[1,17] but due to the relatively large number of states ($=2^8$), such numerical calculations are not feasible. Moreover, at high temperatures,

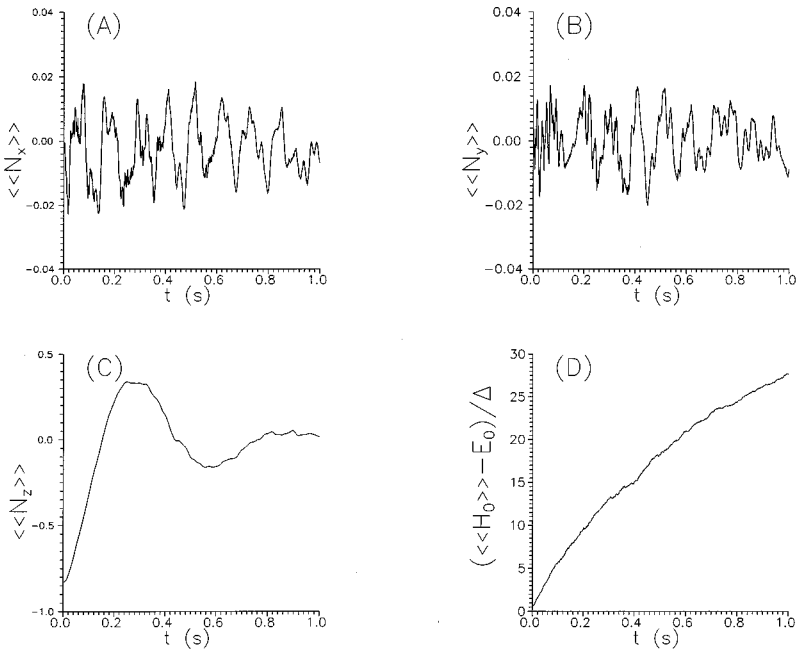

FIG. 2. Ensemble averages of the Néel vector components $\langle\langle N_x\rangle\rangle$ (A), $\langle\langle N_y\rangle\rangle$ (B), $\langle\langle N_z\rangle\rangle$ (C), and $(\langle\langle H_0\rangle\rangle - E_0)/\Delta$ (D) as functions of time when $d=7$ Å and $\tau=2.5$ s.

if the initial state of the system is a pure state and there are only few observable of interest, as in the present case, the computational effort required to find the evolution of the whole density matrix is not justified. Instead, an equivalent description,[18] which can be applied efficiently is provided by the statistical ensemble of N_t Brownian trajectories $|\psi^r(t)\rangle$, $r=1,N_t$ determined by the Schrödinger equation

$$i\hbar\partial_t|\psi^r(t)\rangle=[H_0-\gamma\hbar\vec{B}^e(t)\cdot\vec{\mathbf{I}}]|\psi^r(t)\rangle. \tag{7}$$

This was integrated using the procedure presented in Ref. 3, as a classical system of Hamilton equations for the real and imaginary parts of the amplitudes $y_k(t)=\langle k|\psi(t)\rangle$, $k=1,2^8$. In the real variables $u_k(t)\equiv\mathrm{Re}[y_k(t)]$ and $v_k(t)\equiv\mathrm{Im}[y_k(t)]$, Eq. (7) takes the form

$$2\hbar\dot{u}_k=\frac{\partial\mathcal{H}(t)}{\partial v_k},\quad 2\hbar\dot{v}_k=-\frac{\partial\mathcal{H}(t)}{\partial u_k}, \tag{8}$$

where

$$\mathcal{H}(t)=\sum_{k,k'=1}^{2^8}(u_ku_{k'}+v_kv_{k'})\mathrm{Re}[\langle k|H_0+H_r(t)|k'\rangle] -(u_kv_{k'}-v_ku_{k'})\mathrm{Im}[\langle k|H_0+H_r(t)|k'\rangle]. \tag{9}$$

The dominant AF states are annihilated by $\mathbf{I}_z$, and therefore the contribution of the term $B_z^e(t)\mathbf{I}_z$ from H_r is very small, and it was neglected. The remaining x and y components of the fluctuating field at the moment $t_n=ndt$, normalized according to the FDT, have the form $B^e(t_n)=R_n\sqrt{1/(\gamma^2\tau dt)}$, where $\{R_n,\quad n=1,2,3,\ldots\}$ is a sequence of random numbers with 0 mean and variance 1.

The time-evolution of the ensemble average for an observable $\mathcal{O}$ is defined by

$$\langle\langle\mathcal{O}\rangle\rangle(t)=\frac{1}{N_t}\sum_{r=1}^{N_t}\langle\psi^r(t)|\mathcal{O}|\psi^r(t)\rangle. \tag{10}$$

The results obtained for the Néel vector and the energy when $d=7$ Å, $\tau=2.5$ s and $N_t=20$ are presented in Fig. 2. The components $\langle\langle N_x\rangle\rangle$ and $\langle\langle N_y\rangle\rangle$ of Figs. 2(A) and 2(B) have thermal fluctuations around 0, while $\langle\langle N_z\rangle\rangle$ shown in Fig. 2(C) has damped oscillations. These oscillations can be well approximated by the analytical expression

$$\langle\langle N_z\rangle\rangle(t)=e^{-\Lambda t}\langle N_z\rangle(t), \tag{11}$$

where the damping constant obtained by fit is $\Lambda=3\ \mathrm{s}^{-1}$. Similar calculations with τ in the range of seconds, as expected at low temperatures, indicate that $\Lambda=7.5/\tau$.

The ratio $[\langle\langle H_0\rangle\rangle(t)-E_0]/\Delta$ between the average excitation energy and the doublet splitting Δ is a suitable measure of the heating effect produced by the environment, and is represented in Fig. 2(D). The ensemble average of the energy appearing here is accurately reproduced by the formula $\langle\langle H_0\rangle\rangle(t)=E_0+\Delta(w_1t-w_2t^2)$, with $w_1=45.9\,\mathrm{s}^{-1}$ and $w_2=19.3\ \mathrm{s}^{-2}$.

The calculations presented in this work indicate that the chain of nuclear spins for eight ^{129}Xe atoms placed on a magnetic surface is antiferromagnetic when the chain constant is ~ 7 Å, or larger. This antiferromagnetic system has a low-lying resonance, observable as a macroscopic oscillation of the Néel vector between the two opposite orientations along the normal to the surface. The orientation reversal takes place during a time T_{max}, which increases exponentially with d [Fig. 1(A)]. At finite temperature the resonance oscillations are damped by the residual dipole interactions between the nuclear spins and the fluctuating magnetic field of the surface. This residual coupling was described by one parameter, the relaxation time τ of a single spin. The results indicate that when τ is in the range of seconds, the damping rate is close to $(N-1)/\tau$, where N is the number of spins.

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