Quantum internal dynamics of solitons in one-dimensional antiferromagnets

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We show that the dynamics of the internal degrees of freedom of solitons in one-dimensional antiferromagnets is essentially of quantum origin even in the “classical” case of large spin ($S=\frac{5}{2}$) and differs drastically in the cases of integral and half-odd $S$. We predict the possibility of observing a resonance on the internal soliton modes in the easy-axis quasi-one-dimensional antiferromagnet CsMnI,3, © 1996 American Institute of Physics.

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1. INTRODUCTION

At present the important role of solitons in the physics of one-dimensional magnetic materials is well known and is supported by extensive experimental data, which suggest that the contribution of solitons to the response functions and the thermodynamic characteristics of magnetic systems is considerable (see the reviews in Refs. 1 and 2).

The most direct way of detecting solitons is to observe their contribution to the inelastic neutron scattering cross section, which is known as the soliton central peak in the dynamic structure factor. This contribution is determined by the presence of a finite number density of solitons that are thermally excited and can move freely in the crystal. The presence in the spectrum of such a zero (translational) free-motion mode leads to the appearance of a resonance peak at zero frequency in the system’s response function (the dynamic structure factor $S(q,\omega)$). Because of thermal motion of the scattering centers this peak proves to be Doppler-broadened and has a halfwidth of order $\delta q$, where $\delta q$ is the root-mean-square thermal velocity of the soliton.

In addition to translational modes solitons can have internal degrees of freedom, say, magnon modes localized on the soliton or the projection $S_z$ of the intrinsic spin. Such solitons with additional quantum numbers are sometimes called dyons by analogy with the physics of monopoles.8 The presence of internal modes makes it possible to observe resonances at the corresponding frequencies. The so-called soliton magnetic resonance on the dyon levels with $S_z=\pm \frac{1}{2}$ that are split in an external magnetic field was observed in the quasi-one-dimensional antiferromagnet CsCoCl,3 which has the Ising type in inelastic neutron scattering experiments and electron paramagnetic resonance (EPR) experiments.9 Elementary excitations in Haldane systems are also of a soliton nature10 and have an additional quantum number, $S_z$, which makes it possible to observe the transitions between states with different values of $S_z$ in EPR experiments11,12 (it must be noted that in these experiments the nature of these solitons differ significantly from that of ordinary solitons in the antiferromagnetic order and is based on the hidden-order concept13).

The present study is an analysis of the internal dynamics of solitons in one-dimensional Heisenberg (weakly anisotropic) antiferromagnets in the “semiclassical” limit $S \geq 1$ ($S=\frac{5}{2}$ for all practical purposes). Recently we found14,15 that dyons contribute considerably to the dynamics and thermodynamics of Heisenberg antiferromagnets, but due to an error resulting from ignoring the topological term in semiclassical quantization the dyon spectrum was calculated incorrectly. Using the model of an easy-axis antiferromagnet with weak rhombic anisotropy placed in an external magnetic field, we show that the dynamics of the internal degrees of freedom of solitons in one-dimensional antiferromagnets is essentially of quantum nature even when $S$ is large and varies considerably for cases of integral and half-odd $S$. The well-known method of soliton phenomenology14,15 can easily be generalized to the dyon case. We use it to study the thermodynamics of the dyon gas in easy-axis and weakly rhombic antiferromagnets and predict the possibility of observing resonances on the internal soliton modes in EPR experiments. We also make numerical estimates of the resonance frequencies for CsMnI,3, which at temperatures above the temperature of three-dimensional ordering is a good realization of a quasi-one-dimensional easy-axis Heisenberg antiferromagnet with spin $S=\frac{5}{2}$.

2. THE MODEL: AN EASY-AXIS ANTIFERROMAGNET IN A MAGNETIC FIELD

We start with the model of a one-dimensional easy-axis Heisenberg antiferromagnet with rhombic anisotropy located in an external magnetic field. The model Hamiltonian is

$$H=\sum_1 \{J S_i S_{i+1}+D_1 (S^z_i)^2+D_2 (S^z_i)^2-g\mu_B H S_z\}
\tag{1}$$

where $J>0$ is the exchange integral, $D_1$ and $D_2$ are the anisotropy constants, the $S_z$ are the spin operators describing magnetic ions with spin $S$ placed at the sites of a one-dimensional lattice with a lattice constant $a$, $H$ is the magnetic field, $\mu_B$ is the Bohr magneton, and $g$ is the Landé factor of the magnetic ion. Let us assume that $D_1>D_2>0$. Then $z$ is the easy axis, and $zy$ is the easy plane. It is also convenient to introduce the “rhombicity” parameter $\rho=D_1/D_2<1$; in a purely easy-axis antiferromagnet we have $\rho=0$, while the “nearly easy-plane” case corresponds to $\rho>1$. Despite its simplicity, the model gives a good description of a number of real one-dimensional magnetic materials. For instance, CMC
(CnMnCl₂·2H₂O) corresponds to \( p = 3 \) (Ref. 16) and TMMC (CH₃₃NdMNCl₆) to \( p = 1 \) (Ref. 2), while CnMnI₄, in which hexagonal anisotropy is extremely low and is of the same order of magnitude as interchain exchange, can be assumed for all practical purposes to be an easy-axis magnetic material described by the model (1) with \( p = 0 \) (Refs. 17 and 18).

We also note that the model can be applied to magnetic materials with a symmetry higher than rhombic, for instance, hexagonal symmetry, where in the ideal case the invariant \((S^2)^2\) is symmetry-forbidden. The point is that often in real samples of hexagonal magnetic materials there are rhombic distortions related to crystal growth processes. For instance, in TMMC the rhombic anisotropy of this nature is larger than the natural crystallographic anisotropy and may reach 15% of the uniaxial anisotropy. Moreover, noticeable rhombic anisotropy can be induced by weak external uniaxial pressure, which is often used in analyzing the dynamics of solitons of the domain-wall type in weak ferromagnets (see Refs. 20 and 21).

It is known that the long-wave dynamics of an antiferromagnetic in the \( S^>1 \) limit can be described by the nonlinear sigma-model (see the reviews in Refs. 2, 13, and 22). The effective Lagrangian corresponding to (1) can be obtained by means of a path integral over coherent states and has the form

\[
\mathcal{L} = \frac{1}{2} JS^2 a \left[ \frac{1}{c^2} \left( \frac{\partial^2}{\partial t^2} - \left( \nabla \right)^2 \right) \right] - \Delta a + \frac{g \mu_a S}{2c} (H \cdot \nabla) \left[ \frac{g \mu_a S}{2Sa} (H \cdot \nabla)^2 + \frac{S}{2} \frac{\partial}{\partial t} \right].
\]  

(2)

Here the field \( I \) is the continuum limit of the antiferromagnetic vector defined as a linear combination of spins in the unit magnetic cell, i.e., \( I = (S_a - S_b) / 2S \), with magnetization in a similar manner, \( m_\alpha = (S_a + S_b + 1) / 2S \). In the absence of an external field the Lagrangian (2) is Lorentz-invariant, \( c = 2JSa \), and acts as a limit of the spin-wave velocity, and we write \( \Delta a = \Delta a_f \) with \( f = 1, 2 \) for the characteristic spatial scales determined by the anisotropy. The magnetization \( m \) can be expressed in terms of \( I \) and its derivatives:

\[
m = \frac{H}{4S} \left[ \frac{1}{c^2} \frac{\partial^2}{\partial t^2} + (H \cdot \nabla)^2 \right] \frac{1}{2} \left( \nabla I \right).
\]  

(3)

where \( \gamma = g \mu_a / k \) is the gyromagnetic ratio, and the last term stems from the way in which \( m \) and \( I \) are defined, which clearly violates translational invariance. The vectors \( m \) and \( I \) satisfy the constraints \( m \cdot 1 = 0 \) and \( m^2 + I^2 = 1 \), and since according to (3) \( m \parallel I \) holds in the long-wave approximation, we can assume that in (2) \( I \) is a unit vector. The last term on the right-hand side of Eq. (2) determines what is known as the topological term, responsible for the difference in the physical properties of one-dimensional antiferromagnets with integral and half-odd \( S \) (see the review in Ref. 22).

This term is a total divergence and therefore has no effect on the equations of motion in the classical case. However, under quantization the allowance for this term changes the definition of the canonical momentum and proves important, as we will shortly see. Note that in deriving (2) and (3) we assumed that the number of spins in the chain is even.

Let the \( z \) axis specify the direction of the field. Then in the purely easy-axis case \( p = 0 \) \( (D_1 = D_2 = 0 \) and \( \Delta a = \Delta a_0 = \Delta a_f \) the classical dyon solution describing a two-parameter precession kink can be written explicitly because the \( z \)-component of the total spin is a constant of motion. At this point it is convenient to introduce the angular variables \( I_f = e^{it} \cos \theta \) and \( I_z = \text{const} \). Then the dyon solution has the form

\[
\cos \theta = \alpha \tanh \left( \frac{\xi}{\Delta a_0} \right), \quad \varphi = \varphi_0 + \alpha t + \gamma H t,
\]  

(4)

where \( \varphi_0 = \text{const} \), \( \xi \) and \( \tau \) are the ordinary relativistic combinations

\[
\xi = \frac{x - at}{\sqrt{1 - \gamma^2 c^2}}, \quad \tau = \frac{t - xu/c^2}{\sqrt{1 - \gamma^2 c^2}},
\]  

(5)

\( v \) is the soliton velocity, \( \omega \) is the precession frequency in the soliton reference frame, \( \omega_0 = c\gamma \Delta a_f \) is the magnon activation frequency in the absence of a field, and \( \alpha = \pm 1 \) is the kink’s topological charge. The value of the \( z \)-projection of system’s total spin \( S_z = S(\alpha) \int dS \) corresponding to the solution (4) is given by the following formula:

\[
S_z = \alpha S + \frac{S_o \cos \theta}{\sqrt{1 - \gamma^2 c^2}},
\]  

(6)

where \( S \) is the statistical contribution related to the last term on the right-hand side of Eq. (3). In the classical case \( \omega \) can assume arbitrary values ranging from \( -\omega_0 \) to \( \omega_0 \), and the energy minimum is reached for a static solution with \( \omega = -\gamma H \). With semiclassical quantization we can require that the value of \( S_z \) be integral irrespective of whether \( S \) is integral or half-odd (since the system contains an even number of spins). Then the kink’s ‘intrinsic quantum number’ \( m = S_z - \alpha S \) must be an integer when \( S \) is an integer and a half-odd number when \( S \) is half-odd. The dyon energy is determined by the dyon momentum and the intrinsic variable \( m \) and can be written as follows:

\[
E(P, m) = -g \mu_a H m + E_d(m) + c^2 P^2,
\]  

(7)

where

\[
E_d(m) = h \omega_0 \sqrt{S^2 + m^2}, \quad P = E_d(m) \frac{u/c}{\sqrt{1 - \gamma^2 c^2}}.
\]  

(8)

On the whole, the above reasoning for a purely uniaxial antiferromagnet follows Haldane’s derivation,\(^2\) with slight modifications in argumentation and the way the Lagrangian of the \( \sigma \)-model is derived. However, even with weak anisotropy in the base plane (\( p = 0 \)) there is no way to construct an exact dyon solution, with the result that only approximate methods can be employed.

For \( p \neq 0 \) the simplest stable static soliton solution corresponds to a sine-Gordon kink.
Linear analysis of the excitation spectrum against the kink background suggests that there is a localized magnon mode corresponding to uniform ($\xi$-independent) oscillations of the angle $\varphi$ about the equilibrium value $\varphi_0 = \pi n/2$, with $n$ an integer. In the limit $\rho \to 0$, the frequency of this mode, $\omega_{\varphi} = \omega_{\varphi}(\rho)$, tends to zero and the oscillations become a rotation, as in (4). Quantum fluctuations, however, markedly narrow the range of applicability of the linear approximation. Indeed, estimates of the amplitude of the zero-point oscillation of the angle $\varphi$ yield $< (\varphi - \varphi_0)^2 > = 1/2 \langle \delta \rho \rangle$, so that for real values of $S$ the linear regime is reached only in the easy-plane case, $\rho_\perp > 1$. To analyze the dynamics of the internal mode in the "nonlinear quantum" regime we note that in both limits, $\rho_\perp \to 1$ and $\rho_\perp = 0$, the angle $\varphi$ is independent of the spatial variable $\xi$. We select the following ansatz for the dyon solution:

$$\varphi = \varphi(t), \quad \Delta_\varphi = \Delta_\varphi(\varphi(t)), \quad \cos \theta = \sigma \tanh \frac{x}{\Delta_\varphi(\varphi(t))},$$

where $\varphi$ and $x$ are the slow variables in the sense that $\dot{x}, \ddot{x} \ll \dot{\varphi}, \ddot{\varphi}$, with the dot standing for time derivatives. In the case of weak rhombicity ($\rho_\perp < 1$), to which we limit our discussion, the effective Lagrangian of the problem obtained after (10) is plugged into (2) has the following form in the leading approximation in the small parameters $\rho, \dot{x}, \ddot{x}$, and $\varphi(t)$:

$$L = \frac{E_0}{2 \alpha t^3} \dot{x}^2 + \frac{E_0}{2 \alpha} \dot{\varphi}^2 + \hbar \gamma S \left[ \sigma \varphi \right] \dot{\varphi} + \frac{\hbar \gamma \varphi}{\alpha \omega_0} \dot{\varphi},$$

where $E_0 = \hbar \omega_0 S$. Thus, the initial field problem with an infinite number of degrees of freedom has been reduced to a finite-dimensional problem. Note that in the given geometry with low kink velocities $\dot{x}, \ddot{x} \ll \dot{\varphi}$, the internal and translational degrees of freedom are approximately separated; generally, however, this is not the case and the free motion of a kink with an excited internal mode is not necessarily motion with constant velocity. The presence of the term with the first time derivative is due to the topological term (the last term) in (2); since this term is a total derivative, it has no effect on the classical dynamics but becomes very important in quantization (see, e.g., Ref. 23).

Canonical quantization of the Lagrangian (11) leads to a Schrödinger equation $i \hbar \dot{\Psi} = - \frac{\hbar \gamma}{\alpha} \dot{\varphi} \Psi$, for the wave function $\Psi(\varphi, t)$ describing the internal mode, with a Hamiltonian of the form

$$\hat{H} = \frac{\hbar \omega_0}{2 \alpha t^3} \dot{\varphi} + \frac{\dot{\varphi}^2}{\hbar \gamma S} + \frac{\hbar \gamma \varphi}{\alpha \omega_0} \dot{\varphi}^2 + \frac{\hbar \gamma}{\alpha \omega_0} \sin^2 \varphi + \frac{\hbar \gamma}{\alpha \omega_0} \cos \theta \tanh \frac{x}{\Delta_\varphi(\varphi(t))}.$$

The factor $\Phi$ in (15) appears because the Lagrangian (11) contains terms proportional to the first time derivative; note that $\dot{x} \times \Phi$ holds for any value of $\rho_\perp$. In the absence of a field this factor forbids tunneling at half-odd $S$, similar to the effect of tunneling in small particles with uncompensated spin. An external field lifts the degeneracy and leads to an oscillatory dependence of the spectrum structure on $H$ with a period $\Delta H = \omega_0 / \gamma$. Note that Eq. (15) is valid only for $\rho_\perp = 0$.
grees of freedom are "frozen," the correction to the kink’s Gordon model:

\[ \kappa \sim h \omega_c N/\pi T \]

with the expression for the kink number density in the and 13, so that here we give only a brief description of the features main results. In the classical case the total number density and antisolons can be written as

\[ n_\pm = \pm \frac{L}{2\pi} \sum_{\pm} \int \frac{dk}{2\pi} \frac{\delta(k)}{k} \left[ 1 - e^{-h\omega_0/kT} \right], \]

where \( L \) is the length of spin chain, \( P \) is the soliton momentum, \( n \) numbers the internal modes, \( E_s(p) = \kappa + c^2 P^2/2E_0 \) is the energy of a dyon with momentum \( P \) in the nth excited state, and \( \Sigma_2 \) is the change in the free energy of the magnon gas brought on by production of a single kink:

\[ \Sigma_2 = \frac{L}{2\pi} \sum_{\pm} \int \frac{dk}{2\pi} \frac{\delta(k)}{k} \left[ 1 - e^{-h\omega_0/kT} \right]. \]

Here \( \delta(k) \) is the asymptotic phase shift experienced by a magnon of the \( j \)th branch of the continuous spectrum after interaction with a kink, \( \omega_{1/2} \) are the frequencies of continuous-spectrum magnons (for small \( p \) and weak fields, \( \omega_1(k) = \omega_0(k) = \omega_0(1 + k^2/\Sigma_2^2) \). Generally speaking, \( \delta(k) \) can depend on \( P \) and \( n \), but at low temperatures \( T < E_0 \), dyons with \( \pm P \) and \( n \leq S \) provide the main contribution to the thermodynamics, with the result that this weak dependence can be ignored. The \( \delta \) vs \( k \) dependence can be found approximately from the exact expressions for the wave functions of magnons superposed on a sine-Gordon kink (9),

\[ \delta_{1/2}(k) = -2 \arctan \left( \frac{\xi}{3} \right) \]

Here \( \xi \) is the root-mean-square soliton velocity, and \( \kappa \) is the characteristic soliton energy to a response function we must know how to calculate the corresponding averages. At low temperatures \( T < E_0 \), the statistical mechanics of a dyon gas can be built by the well-known method of soliton phenomenology.\(^{12}\) The dyon distribution function normalized to the total number of solitons and antisolons can be written as

\[ \rho(P,n) = \frac{L}{2\pi} \sum_{\pm} \int \frac{dk}{2\pi} \frac{\delta(k)}{k} \left[ 1 - e^{-h\omega_0/kT} \right]. \]

Generally speaking, the \( \delta \) functions contribute to the dynamic structure factor \( S^{\pm}(Q,\Omega) \), which is the Fourier component of the spin correlation function \( \langle S^z(x,t)S^z(x',t') \rangle \). In the case of an antiferromagnet the main contribution to the dynamic structure factor is provided by the correlations of the antiferromagnetism vector \( \vec{I} \) and is concentrated near the magnetic Bragg wave vector \( Q = Q_{2B} = \pi/a \) (the contribution of \( m \) is concentrated near \( Q = 0 \) and is \( D/J \) times weaker in intensity). The longitudinal component \( S^{\perp}(Q,\Omega) \) (in relation to the easy-axis) of the dynamic structure factor is practically insensitive to internal soliton dynamics, with the result that we can limit ourselves to the transverse components. It is general knowledge that solitons give rise to what is known as the central peak, i.e., a contribution to the dynamic structure factor in the range of \( \Omega \) values close to zero caused by the Doppler-broadened response on the Goldstone translational mode. Classical calculations yield\(^{16}\)

\[ S^{\perp}(Q,\Omega) = f_\perp(Q,\Omega), \]

where

\[ f_\perp(Q,\Omega) = \frac{2\pi}{2\pi} \sum_{\pm} n_\pm \left| f_\perp(q,\Omega) \right|^2 \exp \left( -\frac{\Omega^2}{2q^2\sigma_v^2} \right) \]

Here the halfwidth of such a central peak, \( \Gamma_\perp \), is approximately equal to \( qv_T \), where \( v_T = \sqrt{T/E_0} \) is the root-mean-square soliton velocity, and \( F_\perp(q,\Omega) = \exp(-\Omega^2/2q^2\sigma_v^2) \) is the traverse form factor, which describes the geometric structure of the soliton. The second transverse component, \( S^{\parallel}(Q,\Omega) \), in the linear approximation contains a delta-function peak at the localized magnon mode frequency \( \Omega = \omega_0/\sqrt{p} \).
The quantum case \( pS^<e^4 \) the system is almost axisymmetric and satisfies \( S^>{}^=S^=S^1 \), where \( S^\rangle \) can be written as

\[
S^\rangle (Q^\sphericalangle, \Omega) = \sum_n e^{-i\Omega_n/\omega_0} f(Q, \Omega, \Omega_n) + e^{-i\Omega_n/\omega_0} f(Q, \Omega, -\Omega_n),
\]

where \( \Omega_n/\omega_0 \) are the dyon levels and \( \Omega_n/\omega_0 \) is the corresponding partition function; the quantum number \( m \) assumes the values \( 0, \pm 1, \pm 2, \ldots \) for integral \( S \) or \( 1/2, \pm 3/2, \ldots \) for half-odd \( S \). This expression describes a set of Gaussian peaks at the resonance frequencies \( \Omega_n = \pm \Omega_n \), \( \Omega_n = \Omega_n + \omega_0 \), and a dispersion \( \Omega_n = \omega_0 \), where \( \omega_0/\omega_0 \) is the meaning of the root-mean-square thermal frequency of precession.

For \( |m|<S \) the resonant frequencies \( \Omega^\rangle = \omega_0(|m| + 1/2)/\omega_0 \), which provides the condition for resolving separate peaks,

\[
\omega_0 = S^{-1} \sqrt{\omega_0^2 - \omega_0^2},
\]

and the condition for observing the splitting of resonance lines caused by an external field,

\[
yT \ll \frac{\omega_0^2}{\omega_0^2}.
\]

According to Ref. 18, in the one-dimensional antiferromagnet \( \text{CsMn}_13 \), \( S^=5/2, f^=198 \text{ GHz} \), and \( \Omega^\rangle = 1.07 \text{ GHz} \), so that estimates yield \( \Omega^\rangle = 28 \text{ GHz} \). Such frequencies are more likely to be accessible in EPR experiments than in neutron scattering experiments (note, however, that transitions with \( q=0 \) can be observed only because of the presence of the correlator \( \langle m^m \rangle \)), with the result that the intensity of such transitions is low; the EPR linewidth is determined not by the spread in the dynamical parameters, as it is in the dynamic structure factor, but by relaxation mechanisms, not considered in this paper). Neutron scattering experiments have a low resolution and can be used, at the best, to detect the envelope central peak with a halfwidth \( 1/2 = \omega_0 \), independent of the wave vector (in contrast to the ordinary central peak with \( 1/2 = \omega_0 \)).

4. CONCLUDING REMARKS

Thus, using the example of the simplest model of a nearly easy-axis antiferromagnet with a rhombic anisotropy, we have studied, in the limit of an asymptotic large spin \( S \), the dynamics of the internal degrees of freedom of topological solitons (kinks) in one-dimensional antiferromagnets. Note that the rhombic nature of the anisotropy is not important and was chosen solely to simplify analysis. Indeed, in the case of higher-order symmetries (tetragonal, hexagonal, etc.) we would have a potential with a large number of minima in \( (12) \) instead of a two-well potential, tunneling calculations that are more complicated, and different selection rules in \( S \).

The specific features of the antiferromagnet equations of motion lead to a situation in which the kinks are the light particles, their effective mass being only \( S \) times larger than the effective magnon mass (in contrast to ferromagnets, where this ratio is proportional to \( \sqrt{H_s/H_\text{ex}}, \omega_0 \) and \( H_\text{ex} \) the exchange and anisotropy fields). We found that because of this the internal kink dynamics is of an essentially quantum nature even in the classical case of fairly large spin \( S^>{}^=5/2 \). Quantum coherence effects lead to a structure of the internal modes spectrum that strongly depends on whether \( S \) is integral or half-odd. The presence of an external magnetic field may give rise to complicated effects. In the simplest case of a purely easy-axis antiferromagnet such a field only splits the spectrum, thus lifting the degeneracy in the z-projection of the total spin. Generally, however, the field mixes the internal dynamics and the translational mode of soliton motion.

In addition to drastically changing the thermodynamics of the soliton gas, the presence of internal degrees of freedom in solitons leads to the possibility of observing a resonance involving transition between internal levels, say by EPR or by neutron spectroscopy techniques. We have made numerical estimates of the resonant frequencies for the antiferromagnet \( \text{CsMn}_13 \), which proved to be a good realization of a quasi-one-dimensional easy-axis spin-5/2 Heisenberg antiferromagnet.

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A semi-quantum analysis done in the spirit of this approach shows that in the classical case \( pS^>{}^=4 \) there is still an important quantum effect: the tunneling between two energy-equivalent kink states. Accordingly, the dynamic structure factor \( S^\rangle \) contains a resonance peak at the frequency \( \Omega_n = 2\omega_0/\hbar \), where \( \omega_0 \) is given by Eq. (15) for \( p>1 \) and by

\[
\Lambda = \omega_0(\Phi(0.42, p^2))^{-1/2},
\]

for \( p>1 \) (see Ref. 26). However, it is extremely difficult to decide whether such a resonance peak can be observed, since the problem requires a detailed microscopic analysis of the various mechanisms of coherence destruction, including the mechanism of the interaction with nuclear spin.


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