Quantum Nonlinear Switching Model

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(Dated: January 10, 2002)

We present a method, the dynamical cumulant expansion, that allows to calculate quantum corrections for time-dependent quantities of interacting spin systems or single spins with anisotropy. This method is applied to the quantum-spin model $\hat{H} = -H_z(t)S_z + V(S)$ with $H_z(\pm \infty) = \pm \infty$ and $\Psi(\pm \infty) = |\Psi\rangle$ we study the quantity $P(t) = (1 - \langle S_z \rangle(\tau)/\langle S_z \rangle(0))/2$. The case $V(S) = -H_z S_z$ corresponds to the standard Landau-Zener-Stueckelberg model of tunneling at avoided-level crossing for $N = 2S$ independent particles mapped onto a single-spin-$S$ problem, $P(t)$ being the staying probability. Here the solution does not depend on $S$ and follows, e.g., from the classical Landau-Lifshitz equation. A term $-DS^2$ accounts for particles’ interaction and it makes the model nonlinear and essentially quantum mechanical. The $1/S$ corrections obtained with our method are in a good accord with a full quantum-mechanical solution if the classical motion is regular, as for $D > 0$.

PACS numbers: 03.65.-w, 75.10.Jm, 05.45.Mt

The classical limit of quantum spin systems has been discussed for decades in terms of spin coherent states. Yet little is known, in particular, about quantum corrections to the classical dynamics of systems with large spin $S$. One possible reason is that initially narrow quantum wave packets are spreading with time $t$, i.e., quantum states depend on the combined parameter $\sqrt{t}/S (\alpha > 1)$. Therefore the limits $t \to \infty$ and $S \to \infty$ do not commute and a perturbation theory in $1/S$ is usually impossible. One is left here with the question of how well defined asymptotic ($t \to \infty$) quantities behave in the limit $S \to \infty$. Are they continuous functions of $1/S$ for $1/S \to 0$?

One of the problems of this kind is a recent generalization of the Landau-Zener-Stueckelberg (LZS) problem of transitions at avoided level crossings that is a well known quantum effect ubiquitous in physics of atomic and molecular collisions. Still the standard LZS effect formally allows classical description since Schrödinger equation for a pseudospin $S = 1/2$ is equivalent to the classical Landau-Lifshitz (LL) equation

$$\dot{\mathbf{m}} = \gamma [\mathbf{m} \times \mathbf{H}_{\text{eff}}], \quad \mathbf{H}_{\text{eff}} = -\partial \mathcal{H}/\partial \mathbf{m},$$

(1)

where $\mathbf{m} \equiv (\mathbf{S})/S$, $\gamma = g\mu_B/h$, and, in particular, $\mathbf{H}_{\text{eff}} = \mathbf{H} = H_x \mathbf{e}_x + H_z(t) \mathbf{e}_z$ with $H_z = \Delta$ (level splitting) and $H_z(t) = E_1(t) - E_2(t)$ (level bias) satisfying $H_z(\pm \infty) = \pm \infty$. Typical time dependence of $H_z$ is linear, $H_z = vt$, or nonlinear. The probability to stay at the initially populated level $-1$ is

$$P(t) = [1 - m_z(t)]/2, \quad P(-\infty) = 1$$

(2)

and for the linear sweep

$$P(\infty) \equiv P = e^{-\varepsilon}, \quad \varepsilon \equiv \frac{\pi \Delta^2}{2 \hbar v}.$$  

(3)

This “degeneracy” between quantum and classical descriptions of the LZS effect disappears if tunneling particles interact with each other, as is the case for molecular magnets (see Ref. [13] for a recent review) or for the Bose-Einstein condensate. Finding the time-dependent wave function for the whole system of $N \gg 1$ particles that is specified by $2^N$ coefficients is of course a tremendous problem. As a plausible first step one can consider a simplified model in which each particle interacts with all $N - 1$ other particles with the same strength $J$ that maps onto a large-spin model (we use $g\mu_B = \hbar = 1$ below)

$$\hat{H} = -H_z(t)S_z + V(S), \quad S = N/2$$

(4)

$$V(S) = -H_x S_x - DS^2, \quad D = 2J,$$  

(5)

whereas $P(t)$ is still defined by Eq. (2). On one hand, our original model of interacting particles in the limit $N \to \infty$ is described exactly by the mean-field approximation (MFA) that simplifies the problem to a nonlinear Schrödinger equation for a single tunneling particle with $S = 1/2$. On the other hand, the mean-field limit $N \to \infty$ corresponds to the classical limit $S \to \infty$ of Eq. (4) that results into the LL equation

$$\mathcal{H} = -H_z(t)m_z - v(m), \quad m = -H_x m_x - dm_x/dt$$

and $d \equiv SD$. However for $N \neq \infty$ that better suits to real systems with finite range interactions one is confronted with an essentially quantum effect.

One can also look upon the large-spin problem of Eq. (1) from another perspective and consider it as a kind of scattering problem for a spin: Sweeping the field across the region of strong interaction, $H_z(\pm \infty) = \pm \infty$, with the task to find $\langle S_z \rangle_{t \to \infty}$ if in the initial state the spin was down $\langle S_z \rangle_{t \to -\infty} = -S$. We call it the “Quantum Nonlinear Switching (QNS) model”. This model is more general than the LZS model and it can be applied to real large spins rather than to large pseudospins composed of many pseudospins $1/2$. It could be relevant for molecular magnets, and one can even include in $V(S)$ terms that are absent in the LZS tunneling model, e.g., the biaxial anisotropy $E(S_2^2 - S_1^2)$ characteristic to Fe$_6$. The purpose of this Letter is to investigate the semi-classical limit $S \gg 1$ and to work out an analytical method to calculate $1/S$ corrections to the mean-field...
result for the staying probability $P$ for the QNS model. This provides a crucial test for the MFA that was applied to the LZS effect in systems of interacting particles in the absence of a more accurate and still tractable method\[1, 2, 3, 4\]. In comparison to statics, application of the MFA to dynamics is questionable since the stability of the solution of the nonlinear LL equation Eq. \[1\] with respect to quantum fluctuations is not guaranteed. It becomes clear as soon as one realizes that dynamical MFA describes classical dynamics that might exhibit chaos, that is, classically computed quantities $P(t)$ might be poorly defined.

The method we propose here is a dynamic cumulant expansion (DCE) based on the normalized and symmetrized spin cumulants that are defined by

$$m_\alpha \equiv \langle S_\alpha \rangle / S,$$

$$m_{\alpha\beta} \equiv \frac{1}{S^2} \left( \langle S_\alpha S_\beta \rangle + \langle S_\beta S_\alpha \rangle - \langle S_\alpha \rangle \langle S_\beta \rangle \right)$$  \hspace{1cm} (6)

eq 0, \beta = x, y, z = 1, 2, 3. We use the Heisenberg representation for spin operators $S_\alpha$ and compute matrix elements with respect to the initial state that is the particular case $\theta = \pi$ of the spin coherent state $|\Omega(\theta, \phi)\rangle = \sum_{m=-S}^S C_m|m\rangle$ with

$$C_m = \left( \frac{2S}{S+m} \right)^{1/2} \left( \cos \frac{\theta}{2} \right)^{S+m} \left( \sin \frac{\theta}{2} \right)^{S-m} e^{i(S-m)\phi}$$  \hspace{1cm} (7)

\[1\] \[2\] \[3\] \[4\]. Matrix elements of products of operators factorize in the classical limit, and, correspondingly, normalized cumulants of $n$ spin operators are as small as $S^{1-n}$\[17, 18\]. Thus a theory formulated in terms of cumulants yields a quasiclassical expansion. It is sufficient to retain pair cumulants and drop higher-order cumulants to obtain $1/S$ corrections. If the state of the system would be close to a coherent state during all the time, the cumulant method would yield the result without any difficulties. We will see, however, that this assumption fails at large times because the nonequidistant spectrum of a nonlinear spin system leads to dephasing of different $|m\rangle$-components of the coherent state. One can think of a strongly localized cloud of classical spins that, however, are precessing with slightly different frequencies. Whatever narrow is this cloud, it will spread along the classical trajectory within a dephasing time $T_{\text{dep}}$. That is, cumulants cease to be small at large times and the DCE diverges. Mathematically it means, as we will see below, that $P(t)$ depends on both $S^{-1}$ and on the combined parameter $S^{-1}t^2$, where the pure $S^{-1}$ describes the quantum correction to the QNS and $S^{-1}t^2$ describes the dephasing effect. The DCE picks both terms and thus it diverges at $t \to \infty$. Fortunately this divergence can be eliminated to yield a pure $1/S$ correction to $P \equiv P(\infty)$, as shown in Fig. \[1\] for the model described by Eq. \[1\].

To construct DCE, we use the Heisenberg equation of motion for spin operators with an interaction $V(S)$ that is taken to be a fully symmetrized function of $S_\alpha$

$$\dot{S}_\alpha = \epsilon_{\alpha\beta\gamma} \tilde{H}_\gamma(t, S) S_\beta$$  \hspace{1cm} (8)

$$\tilde{H}_\gamma(t, S) = H_\gamma(t) - \partial V(S) / \partial S_\gamma.$$  \hspace{1cm} (9)

Eq. \[8\] is in fact fully symmetrized but we don’t write it explicitly to save space. Our quantity of interest is $m_\alpha(t) = \langle -S | S_\alpha(t) | -S \rangle / S$. Taking this matrix element makes Eq. \[8\] nonclosed because $\partial V(S) / \partial S_\gamma$ is an operator. After expanding the matrix element of Eq. \[8\] up to the pair cumulants and dropping higher-order ones being of order $O(1/S^2)$ it becomes an equation for $m_\alpha$ coupled to the pair cumulants $m_{\alpha\beta}$. Equation for the latter also follows from Eq. \[8\] and it is closed at order $1/S$. Initial conditions to these equations at $t = -\infty$ are $m_\alpha = -1$ and $m_{xx} = m_{yy} = 1/(2S)$ while other components are zero. As equations for $m_\alpha$ and $m_{\alpha\beta}$ are coupled, its solution generates all powers of $1/S$ that should be suppressed to obtain a pure $1/S$ expansion. To this end, we expand

$$m_\alpha = \mu^{(0)}_\alpha + \frac{1}{2S} \mu^{(1)}_\alpha + \ldots,$$  \hspace{1cm} (10)

$$m_{\alpha\beta} = \frac{1}{2S} \mu^{(1)}_{\alpha\beta} + \ldots$$  \hspace{1cm} (9)
It is convenient to express the results in terms of
\[ v_{\alpha_1 \cdots \alpha_n}(m) = S^{n-1}V_{\alpha_1 \cdots \alpha_n}(S^m) \]
\[ V_{\alpha_1 \cdots \alpha_n}(S) = \frac{\partial^n V(S)}{\partial S_{\alpha_1} \cdots \partial S_{\alpha_n}}. \]  
As a result one obtains the three equations
\[ \dot{\mu}_0^{(0)} = T_{\alpha\beta}(t, \mu^{(0)}(0))\dot{\mu}_0^{(0)} \]
\[ \dot{\mu}_{\alpha\beta}^{(0)} = \tilde{T}_{\alpha\gamma}(t, \mu^{(0)}(0))\dot{\mu}_{\gamma\beta}^{(0)} + \tilde{T}_{\alpha\gamma}(t, \mu^{(0)}(1))\dot{\mu}_{\gamma\beta}^{(0)} \]
\[ \dot{\mu}_1^{(1)} = \tilde{T}_{\alpha\beta}(t, \mu^{(0)}(0))\dot{\mu}_1^{(1)} + k_{\alpha\beta\gamma}(\mu^{(0)})\dot{\mu}_{\beta\gamma}^{(1)}, \]
where
\[ T_{\alpha\beta}(t, m) = \epsilon_{\alpha\beta\gamma}H^{\alpha\beta}(t, m) \]
\[ \tilde{T}_{\alpha\beta}(t, m) = T_{\alpha\beta}(t, m) - \epsilon_{\alpha\gamma\delta}m_{\gamma}\dot{v}_{\beta\delta}(m) \]
and
\[ k_{\alpha\beta\gamma}(m) = -\epsilon_{\alpha\beta\delta}\dot{v}_{\beta\gamma}(m) - \epsilon_{\alpha \gamma \delta} \frac{1}{2!} \dot{v}_{\beta\gamma\eta}(m) m_{\delta}. \]

Equations (13)–(15) should be solved numerically in order of their appearance. The closed Eq. (13) is a nonlinear Landau-Lifshitz equation, whereas Eqs. (14) and (15) are linear. Although Eqs. (13), (15) and the method of their derivation resemble decoupling schemes that are usually not based on small parameters, the DCE is a rigorous (although formal) expansion in powers of $1/S$.

Let us now check Eqs. (13)–(15) for the exactly solvable toy model of Eqs. (1) with $H_x = 0$, starting at $t = 0$ with an arbitrarily directed spin coherent state, Eq. (4). Their solution is nothing else than a $1/S$ expansion of the exact solution taken for
\[ t \ll T_{\text{rec}} = \pi/D = \pi S/d, \]
where $T_{\text{rec}}$ is the recurrence period, i.e., $1/S$ expansion of
\[ m_z(t) = \frac{Im\psi(t)}{1 + \frac{\partial^2 t^2}{S} - S S} \exp \left[-\frac{\partial^2 t^2}{S} \sin^2 \theta \right] \sin \theta. \]

In fact this expansion is only valid for $t \ll T_{\text{deph}}$, where
\[ T_{\text{deph}} = \sqrt{S/d} \ll T_{\text{rec}}, \]
is the dephasing time mentioned above Eq. (5).

Consideration of the toy model shows that spin switching and dephasing are different effects that are well separated in time for $S \gg 1$: QNS occurs in the vicinity of the resonance, $T_{\text{QNS}} \sim S^0$, while dephasing occurs much later, $T_{\text{deph}} \sim S^{1/2}$. Thus one concludes that the divergence of DCE because of dephasing is harmless and it can be cured. In the QNS setup, dephasing affects dynamics of $m_z(t)$ via $V(S)$. For $S \to \infty$, time dependence of $m_z$ far past the resonance can be found perturbatively in $v(m)/H_z(t) \ll 1$. Here precession of $m$ around the $z$ axis is much faster than the temporal change of $H_z$, i.e.,
\[ \dot{H}_z(t)/H_z^2(t) \ll 1. \]
Thus the classical energy $\mathcal{H}$ is nearly conserved over the period of precession, and it yields
\[ m_z(t) = m_z^{(As)} + \delta m_z(t) \]
\[ \delta m_z(t) = v(m^{(As)}(t))/H_z(t), \]
where $m^{(As)}(t)$ corresponds to the asymptotic form $H^{(As)} = -H_z(t)m_z$. Precession of $m^{(As)}(t)$ causes oscillatory behavior of $\delta m_z(t)$ and thus of $P(t)$ that however vanishes in the limit $H_z(t) \to \infty$ (see Fig. 1). For finite $S \gg 1$, dephasing of $\delta m_z(t)$ follows from the first-order quantum-mechanical perturbation theory
\[ \delta m_z(t) \approx \frac{1}{S H_z(t)} \text{Re} \sum_{m,m' = -S} c_m^{*}c_{m'} V_{mm'} e^{i\delta \Phi_{mm'}}, \]
where $V_{mm'} = \langle m \mid V(S) \mid m' \rangle$, $\delta \Phi_{mm'} = \Phi_{m'}(t) - \Phi_{m}(t)$, and $\Phi_{m}(t) = -m \int^t dt' H_z(t') + V_{mm} t$. Dephasing stems from the nonequidistant spectrum, i.e., from the nonlinear $m$-dependence of $V_{mm}$. As coefficients of the unperturbed state $c_m$ after crossing the resonance are close to those of Eq. (7) and are localized around a classical value $m_0 = \cos \theta$, calculation in Eq. (24) yields results similar to those for the toy model. Expanding $\delta m_z(t)$ in $1/S$ similarly to that of Eq. (20) yields a linear time divergence
\[ \delta m_z(t) \propto t S^{-1} \cos [\Phi_{H_z}(t) + \psi(t)], \]
for the linear sweep $H_z(t) = v t$, where $\Phi_{H_z}(t) = \int^t dt' H_z(t')$ and $\psi(t)$ is a slowly varying phase generated by the interaction: $\psi(t) \sim v(m(t))$. This is exactly the raw result of DCE shown in Fig. 1.

We have found the source for the divergence of the DCE and seen that the divergent contribution $\delta m_z(t)$ in fact tends to zero, if described properly by Eq. (24) while the QNS effect with $1/S$ corrections is entirely contained in the asymptotic value $m_z(\infty) = m_z^{(As)}$ in Eq. (22). Now it is clear that one should simply project out parts of $m_z(t)$ that are oscillating at large times. The latter can be done, e.g., by the "antiphasing"
\[ \bar{m}_z(t) = \frac{1}{2} \left[ m_z \left( t + \frac{T_{H_z}}{4} \right) + m_z \left( t - \frac{T_{H_z}}{4} \right) \right], \]
where $T_{H_z} = 2\pi/H_z(t)$ is the period of oscillations. This transformation kills the diverging factor $t$ in Eq. (20). To improve convergence, one can repeat antiphasing several times, including that with multiple precession frequencies arising due to nonlinearity. The result shown in Fig. 1 at $t \to \infty$ is in a good accord with the exact numerical solution of the Schrödinger equation for $P \equiv P(\infty)$ that is shown by a horizontal line.

Fig. 2 shows $P$ vs the quantum parameter $1/(2S)$ for different sweep-rate parameters $\epsilon$ of Eq. (3). The agreement between the exact numerical solution and the result from DCE for $d \equiv SD = 3$ (that corresponds to strong
ferromagnetic interaction) and \( H_x = 1 \) is very good. Particularly surprising is the linearity of \( P \) vs \( 1/(2S) \) down to \( S = 5/2! \) Quantum corrections to \( P \) become more pronounced for slow sweep, \( \varepsilon \gg 1 \), and the slope of the curves tends to infinity in the limit \( \varepsilon \to \infty \). This is in accord with the finding of Ref. [7]. For \( h_x \equiv H_x/(2d) < 1 \) in the classical case \( \lim_{\varepsilon \to \infty} P = (1 - h_x^{2/3})^{3/2} \), whereas in the quantum case \( \lim_{\varepsilon \to \infty} P = 0 \) because of high-order tunneling analogous to tunneling in molecular magnets.

We have seen above that divergence of DCE because of dephasing is in fact a formal problem that can be solved to yield an excellent description of quantum effects in the QNS model. However there is another more fundamental problem related to the smoothness of the classical solution. For the antiferromagnetic sign of the interaction, \( d < 0 \), the classical solution for \( P \) is a nonmonotonic function of \( \varepsilon \) dropping fast to \( P = 0 \) close to critical values \( \varepsilon_{\nu}, \nu = 1, 2, \ldots \) (see Figs. 6 and 8 of Ref. [7]). \( \varepsilon_{\nu} \) are bifurcation points for classical trajectories at which for finite \( S \) the quantum distribution \( |c_m(t)|^2 \) that was a narrow packet at the beginning splits into two packets when crossing the resonance. One can say that two parts of the quantum packet follow different classical trajectories. In this case DCE cannot be expected to work well since the assumption of a narrow packet close to a coherent state is not fulfilled. Accordingly quantum corrections are very large, and one needs an extremely large spin to approach the classical limit near these points (see Fig. 8 of Ref. [7]). The curves \( P \) vs \( 1/(2S) \) are not straight in the scale of Fig. 2 and they are very different for different \( \varepsilon \).

Classical models with more complicated interactions such as the biaxial model with the transverse field along the hard axis show dynamical chaos for time dependent \( H_z \) that makes the classical nonlinear spin switching effect poorly defined. In the chaotic regime the Schrödinger equation provides a more adequate description of the problem than classical equations of motion as it “smoothes” the chaos (see, e.g., Ref. [19]). As a result, quantum effects are much stronger than \( 1/S \) corrections in case of regular motion. On the other hand, for the field along the medium axis classical motion is regular and smoothly depends on parameters, thus DCE provides an excellent description of quantum corrections at order \( 1/S \) that is comparable with that of Fig. 2.

In this Letter, we have tested the dynamical cumulant expansion for the particular model of interacting tunneling species with a special interaction that is equivalent to a large spin. We have shown that in the case of a smooth classical motion it works well. This method can be generalized for systems with realistic interactions that cannot be mapped onto a large spin and thus are problematic if one uses the Schrödinger equation for the whole system. The system of cumulant equations will include that for the magnetization \( m_{\alpha,i} \) that in the simplest case is independent of the site \( i \) and those for the pair cumulants \( m_{\alpha\beta,ij} \) that depend on the distance between \( i \) and \( j \). One has to check however before applying DCE that the classical (i.e., the mean-field) solution of the problem is not chaotic.

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