Quantum Mechanical Transitions in a Dissipative Environment

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We have constructed an algorithm to treat quantum mechanical dynamics, such as the nonadiabatic transitions in a dissipative environment, by making use of the formula of reduced density matrix, i.e., Quantum Master Equation (QME). We applied the method to investigate dynamics of magnetization of nanoscale magnets, such as Mn_{12} and Fe_8, in a time dependent field. Due to the discreteness of the energy levels in small systems the resonating tunneling phenomena play an important role. At relatively high temperatures the excited levels contribute to the transition and the transition rate depends significantly on the temperature. On the other hand, at very low temperatures the magnetization process does not depend on the temperature. Even at such a low temperature, the effect of environments is still relevant. We demonstrate such a process with the method of QME. We also analyze temperature dependent phenomena of hysteresis of the nanoscale magnets.

§1. Introduction

Relaxation phenomena of metastable state have been one of the most interesting topics of statistical physics. Mechanisms of the hysteresis phenomena of ferromagnets have been understood in the picture of free energy function in the mean-field analysis, which is very intuitively understandable. For real processes of relaxation in bulk system, however, microscopic analyses are necessary, i.e. from the view point of the critical nucleation and motion of the domain wall. [1)-3)]

Recently, phenomena of real time dynamics of quantum mechanics are attracting much interest, because of the developments of microscopic design technique and methods to synthesize micro-size molecular magnets. Indeed, in many experiments, saturation of the relaxation time at low temperatures has been reported, which suggests that some temperature independent mechanism of relaxation exists. The quantum fluctuation is one of the most attractive candidate for such a mechanism, although there are other mechanisms which could give temperature independent relaxation. [4)]

Evidences of quantum mechanical dynamics have been found in processes of the magnetization of uniaxial micro-size molecules under sweeping the magnetic field. In small magnets such as Mn_{12} [5)-10)] or Fe_8 [11)-13)] which have $S = 10$, the discreteness of the energy levels causes interesting phenomena. At low temperatures, it is almost

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impossible to jump up the energy barrier. But the system can relax to state at every avoided crossing point of the energy levels where a kind of quantum tunneling occurs.

We have investigated the dynamics of the quantum state numerically by simply solving the Schrödinger equation to study pure quantum mechanical motion. Here the higher order decomposition of exponential operator introduced by Suzuki [14] is very useful. In order to change the state quantum mechanically, we change the field with a finite speed. The resonant tunneling is quite naturally explained from the viewpoint of the nonadiabatic transition (NAT). [15-17]

In real experiments, however, we cannot neglect the effect of thermal environment, because the energy scale of the magnetic material is very low. The temperature dependence of such quantum mechanical process became an important topics. [18-20,23] We have constructed an algorithm to treat quantum mechanical dynamics, such as the nonadiabatic transitions in a dissipative environment. First we study effects of random field on the quantum dynamics by simulating quantum dynamics with time dependent random filed. Next we introduce an equation of motion of reduced density matrix making use of the projection method. [21,22] Here we assume that the thermal bath consists of infinite number of independent bosons which contacts with the system very weakly.

At relatively high temperatures the excited levels contribute to the transition and the transition rate depends significantly on the temperature. On the other hand, at very low temperatures the magnetization process does not depend on the temperature. [10] Even at such a low temperature, the effect of environments is still relevant. [23]

We have found that the dissipative effect cause additional relaxation which changes the amount of jumps of the magnetization at the resonant points ΔM_i. [23] But taking this effect into account, we can estimate the quantum mechanical transition probability.

We also point out some general features of the thermally assisted resonant tunneling, such as the parity effect of the amount of relaxations. [5]

§2. Nonadiabatic Transition

Nanoscale molecular magnets, such as Mn_{12} [6-9], Fe_8 [11,12], consist of small number of atoms forming an effective $S = 10$ spin and interactions among molecules are very small. Thus each atom can be regarded as an $S = 10$ single spin. The Hamiltonian of such model is generally given by

$$\mathcal{H} = -DS_z^2 - HS_z + Q, \tag{2·1}$$

where $S_z = -10, -9, \cdots 10$ and Q denotes a term which causes the quantum fluctuation, such as $S_x, S_x^2 - S_y^2$, or $(S^+)^4 + (S^-)^4$. In these systems the energy levels as a function of the field have a discrete structure (Fig.1(a)) such that very explicit quantum mechanical dynamics can be expected.

At each crossing point, a small energy gap is formed due to Q (Fig.1(b)). This local structure is called avoided level crossing. When the field is swept through such

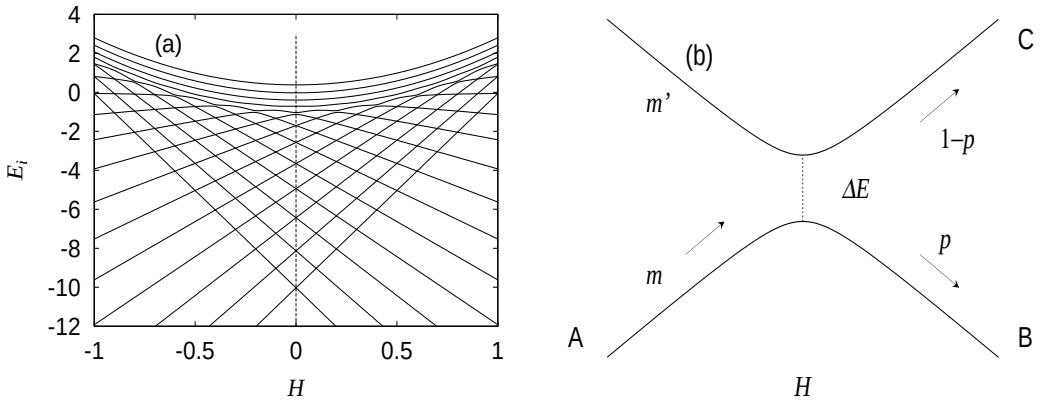

Fig. 1. Energy structure of $S = 10$ Uniaxial magnet as functions of the external field H. (a) global structure and (b) avoided level crossing.

avoided level crossing points, so-called nonadiabatic transition occurs. The nonadiabatic transition plays important roles in microscopic quantum dynamics such as the level dynamics of semiconductor, chemical reaction and optics. The transition probability in various cases of nonadiabatic transitions has been reviewed by Nakamura. [24)]

In the present paper, nonadiabatic transition between two crossing levels as shown in Fig.2(b) will be considered. That is, the population coming in from the channel A is scattered to B and C with a probabilities p and $1 - p$, respectively. Here p is a probability to stay the same level, i.e. to behave adiabatically. This process corresponds to tunnel. On the other hand, $1 - p$ is a probability to jump up to the channel C. The states of channel A and C are the same state when the perturbation $Q = 0$ and are similar in character. This unperturbed state is called diabatic state. In this sense, $1 - p$ is a probability to stay in the diabatic state. This process corresponds to un-tunnel. The probability p plays an important role in quantum mechanical relaxation of the system. This probability was studied by Landau [25)], Zener [26)] and Stückelberg [27)] in 1932 and is given by

$$p = 1 - \exp\left(-\frac{\pi(\Delta E)^2}{2c\hbar g\mu_{\rm B}\Delta m}\right), \qquad (2\cdot2)$$

where ΔE is a gap at the avoided level crossing and Δm is the difference of magnetization of the levels. c is the speed of the sweeping field, $c = dH/dt$. Thus $g\mu_{\rm B}\Delta mc$ is the changing rate of the Zeeman energy. [15)-17)]

This LZS type nonadiabatic transition has two characteristics. One is the localization of transition. That is, the transition occurs only in the vicinity of the crossing point. The other is the sweeping rate dependence of the transition probability. It has been also pointed out that the application of an alternate field at the resonant points causes a nontrivial oscillation of magnetization due to phase interference. [28)]

For quantitative analysis of experimental data, we have to take into account the effect of environment. It is found that even at very low temperatures where the relaxation process does not depend on the temperature, the process is not a pure quantum process. We call such a process "deceptive nonadiabatic transition". At higher temperatures, excitation levels begin to contribute to the relaxation phenomena. Alternative enhancements of relaxation at resonant points are observed, which is called 'parity effect'. [5] This effect is also interpreted as NAT of the population of excited levels.

§3. Numerical Method

In order to investigate quantum dynamics in dissipative environments, we have used two kinds of numerical method. We have studied quantum dynamics with temporally fluctuating external field by solving the Schrödinger equation. Statistical properties are obtained by averaging over the distribution of the randomly fluctuating field. [29]

Another method is to use the quantum master equation which describes the equation of motion of the reduced density matrix of the system $\rho(t)$. The equation is derived by tracing out the degree of freedom of the environment from the density matrix of the total system which consists of the system $\mathcal{H}_{\rm S}$, the thermal bath $\mathcal{H}_{\rm B}$ and interaction between them:

$$\mathcal{H} = \mathcal{H}_{\rm S} + \mathcal{H}_{\rm I} + \mathcal{H}_{\rm B} \tag{3·1}$$

and

$$\rho(t) = {\rm Tr}_{\rm B} e^{-\beta\mathcal{H}}. \tag{3·2}$$

In the limit of weak coupling, i.e. the second order of $\mathcal{H}_I$, and assuming that the correlation time of the bath variable is very short (Markovian approximation), we have an equation in the following form

$$\frac{d}{dt}\rho(t) = \frac{1}{i\hbar}[\mathcal{H}, \rho(t)] + \Gamma\rho(t), \tag{3·3}$$

where Γ is a linear operator acting on $\rho(t)$. In the cases where the bath consists of infinite number of bosons, a general expression can be derived. [22]

$$\frac{\partial\rho(t)}{\partial t} = -{\rm i}\,[\mathcal{H}, \rho(t)] - \lambda\left([X, R\rho(t)] + [X, R\rho(t)]^\dagger\right), \tag{3·4}$$

where

$$\langle\bar{k}|R|\bar{m}\rangle = \zeta(\frac{E_{\bar{k}} - E_{\bar{m}}}{\hbar})n_\beta(E_{\bar{k}} - E_{\bar{m}})\langle\bar{k}|X|\bar{m}\rangle,$$
$$\zeta(\omega) = I(\omega) - I(-\omega), \quad \text{and} \quad n_\beta(\omega) = \frac{1}{e^{\beta\omega} - 1}.$$

Here β is an inverse temperature of the reservoir $1/T$, and we set $\hbar$ to be unity. $|\bar{k}\rangle$ and $|\bar{m}\rangle$ are the eigenstates of $\mathcal{H}$ with the eigenenergies $E_{\bar{k}}$ and $E_{\bar{m}}$, respectively. $I(\omega)$

is the spectral density of the boson bath. X is an operator of system which interacts with the bosons linearly. Here we take $X = \frac{1}{2}(S_x + S_z)$. The relaxation process can be affected by the form of X. Generally $X = S_x$ is more efficient than $X = S_z$ for the relaxation. A detailed comparison with other choices will be presented elsewhere. The concrete form of $\Gamma\rho$ depends on the way of coupling between the system and the thermal bath, and also on the nature of the thermal bath, e.g., the spectrum density, etc. However, here we discuss only natures which does not depend on the detail of the model.

§4. Quantum Dynamics in Dissipative Environment

4.1. *Deceptive nonadiabatic transition*

In an avoided level crossing point (m, m') the change of magnetization ΔM is given by

$$\Delta M = pm' - (1-p)m - m = p(m' - m). \tag{4·1}$$

At the lowest point $(-S, S)$, this relation should be applicable.[16] However at higher crossing points (m, m') with $m' < S$, the population scattered from m to m' is unlikely to stay on the level of m' and easily decays to the ground state, i.e., $m' \to S$, even if the dissipative effect is so small that the population at the metastable level of m hardly decays. This difference can be easily understood from the intuitive picture of Fig. 1(a). That is, the relaxation in the same valley, i.e., $m' \to S$, is easy while the relaxation over the barrier $m \to S$ is hard. In this situation, we can not apply the relaxation (4·1) directly to estimate the LZS probability p. However we can still estimate p from ΔM because the relaxation from the level of m occurs only by the LZS probability and the following relaxation to the ground state occurs in a rather short time. Taking these processes into account, we modify the relation (4·1) by replacing the final magnetization m' by S:

$$\Delta M = pS - (1-p)m - m = p(S - m) \tag{4·2}$$

We have confirmed that this relation works by performing simulations using the QME.[23]

4.2. *Parity effect*

Experimentally it has been observed that the amount of relaxation at the resonant points change alternatively.[5] The transition probabilities at the resonant points increase monotonically as the difference of magnetizations $|m - m'|$ of levels decreases. Thus we expect that the transition probabilities at resonant points with the same value of $|m - m'|$ are nearly the same. At finite temperatures, there are some populations at excited states. When the field is swept, they also move with field until a resonant point of appreciable transition rate. Those points are located at the same horizontal level in Fig. 1(a). Because those points locate every second resonant points and thus main relaxations occurs at these points which causes the alternate change of the amount of relaxation.[30]

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