

# Numerical simulation of quantum dynamics in dissipative environment : Study of resonant tunneling of nanoscale molecule magnets

Seiji Miyashita and Keiji Saito  
Department of Applied Physics, University of Tokyo

Quantum dynamics of nanoscale molecular magnets, such as  $Mn_{12}$  and  $Fe_8$ , in a dissipative environment is studied by numerical methods. Due to the discreteness of the energy levels in small systems the nonadiabatic transition plays an important role. However effects of environments disturb the quantum process. 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). 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 processes of such dynamics by the method of QME.

## Numerical methods on quantum dynamics of nanoscale magnets

Recently, evidences of quantum mechanical dynamics have been found in processes of the magnetization of uniaxial micro-size molecules under sweeping the magnetic field. Such dynamics of nanoscale magnets is studied from the view point of the nonadiabatic transition (NAT). In small magnets such as  $Mn_{12}$  or  $Fe_8$  which have  $S = 10$ , the discreteness of the energy levels cause interesting phenomena. At low temperatures, it is almost 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 in original Hamiltonian, *i.e.* by simply solving the Schrödinger equation to study pure quantum mechanical motion. Here higher order decomposition of exponential operator introduced by Suzuki<sup>1)</sup> is very useful.

Nanoscale molecular magnets, such as  $Mn_{12}$ ,<sup>2-8)</sup>  $Fe_8$ ,<sup>9-11)</sup> 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

$$\mathcal{H} = -DS_z^2 - HS_z + Q, \quad (1)$$

where  $S_z = -10, -9, \dots, 10$  and  $Q$  denotes terms which cause 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 as shown in Fig. 1(a). At each crossing point, a small energy gap is formed due to  $Q$ . This local structure is called avoided level crossing, when the field is swept through such avoided level crossing point (Fig. 1(b)), so-called nonadiabatic transition occurs.

The nonadiabatic transition plays important roles in microscopic quantum dynamics such as the level dynamics of semi-

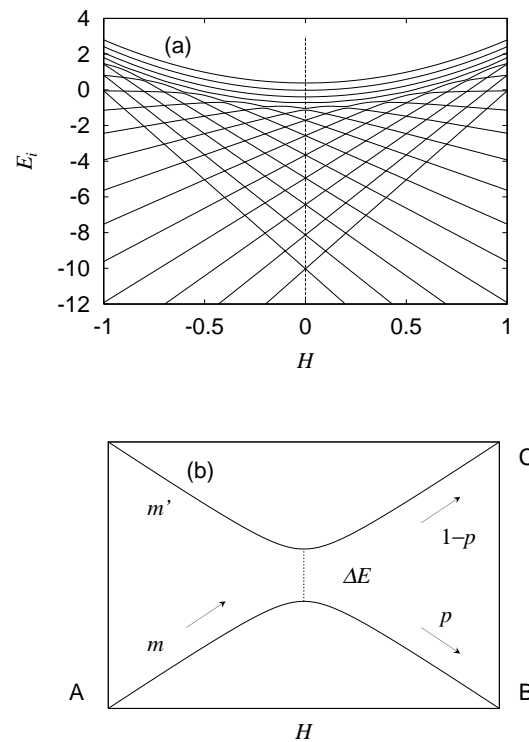


Fig. 1. Energy structure of  $S = 10$  Uniaxial magnet as functions of the external field  $H$ . (a) global structure and (b) avoided level crossing.

conductor, chemical reaction and optics. The transition probability in various cases of nonadiabatic transitions has been reviewed by Nakamura.<sup>12)</sup> The present type of nonadiabatic transition is called Landau-Zener-Stückelberg transition.<sup>13-15)</sup> We have studied the dynamics of nanoscale magnets from the view point of the nonadiabatic transition.<sup>16-18)</sup>

In real experiments, however, we cannot neglect the effect of thermal environment, because the energy scale of the magnetic material is very low. 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.<sup>19)</sup> Another method is to use the quantum master equation<sup>20)</sup> which describes the equation of the motion of the reduced density matrix of the system  $\rho(t)$ . The equation is derived by tracing out the degree of the freedom of the environment from the density matrix of the total system which consists of the system  $\mathcal{H}_S$ , the thermal bath  $\mathcal{H}_B$  and interaction between them:

$$\mathcal{H} = \mathcal{H}_S + \mathcal{H}_I + \mathcal{H}_B \quad (2)$$

and

$$\rho(t) = \text{Tr}_B e^{-\beta \mathcal{H}}. \quad (3)$$

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

$$\frac{d}{dt}\rho(t) = \frac{1}{i\hbar}[\mathcal{H}, \rho(t)] + \Gamma\rho(t), \quad (4)$$

where  $\Gamma$  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.<sup>21)</sup>

$$\begin{aligned} \frac{\partial \rho(t)}{\partial t} = & -i[\mathcal{H}, \rho(t)] \\ & - \lambda \left( [X, R\rho(t)] + [X, R\rho(t)]^\dagger \right), \end{aligned} \quad (5)$$

where

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

Here  $\beta$  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 is attached to bosons of the reservoir linearly representing the interaction of the system with the thermal bath. 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.

At very low temperatures, magnetization processes do not depend on the temperature any more. We have found that the dissipative effect cause additional relaxation which changes the amount of  $\Delta M_i$  even at very low temperatures. We have demonstrated such processes by QME and compared with the pure quantum process which shows successive Landau-

Zener-Stückelberg transition. We called this apparent steps ‘deceptive nonadiabatic transition’. We also showed that taking this effect into account, we can estimate the quantum mechanical transition probability  $\{p_i\}$ .<sup>22)</sup>

At higher temperatures, the excitation levels also contribute to the magnetization processes. As a characteristic of such process, a parity effect has been pointed out.<sup>8)</sup> Namely, the amount of relaxation at resonant points change alternately. We pointed out that this effect is a general features of quantum relaxation of uniaxial magnets, where NAT of excitation levels plays an important role.<sup>23)</sup>

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