Quantum dynamics of spins in time-dependent field

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We explore the quantum dynamics of spin systems in time-dependent fields. In particular, the quantum hysteresis phenomena in microscopic molecular magnets in a sweeping field and fundamental process of so-called quantum computing are investigated. We also discuss the manipulation of spins in quantum spin systems.

Introduction

Explicit real-time quantum dynamics has become possible to be observed due to the recent developments of microscopic technology and the synthesis of functional materials with microscopic structures. Regarding the dynamical properties, we can point out the following approaches. Using the Kubo formula we can obtain the dynamical susceptibility of strongly correlated quantum systems. For this purpose, we may diagonalize the full Hamiltonian[1)] or make use of the real-time evolution of state.[2)] The real-time quantum dynamics is a key ingredient of 'quantum computing' for which various attempts have been proposed.[3)] In the relaxation process at low temperature, the role of 'quantum tunneling' is crucial.[4)] These approaches would be interesting to develop new concepts of physics or to introduce new technology concerning nanoscale phenomena. In this article we briefly review on the quantum dynamics of spin systems.

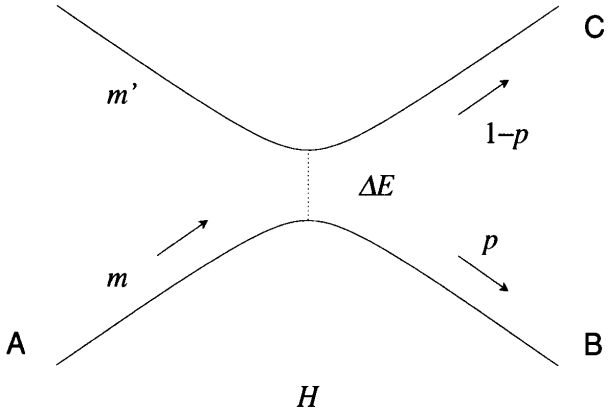

Fig. 1. Avoided level crossing and non adiabatic transition.

Nonadiabatic transition

In microscopic molecular magnets such as Mn_{12},[5)] Fe_8,[6)] and V_{15},[7)] the uniaxial magnetization shows a peculiar hysteresis in a sweeping field at low temperature, which reflects the discrete energy level of the small system. In particular, changes in the magnetization occur only at discrete values of the external field $\{H_i\}$ where the energy levels cross. Precisely speaking, the crossing levels are hybridized by a small perturbation and form so-called avoided level crossing structure. (See Fig. 1). This change of the magnetization at crossing points is due to a transition from a state of a magnetization m to another state with m' which is a kind of tunneling phenomenon and is a quantum-mechanical process. It is thus called resonant tunneling. Classically, magnetization needs to jump up the energy barrier due to the uniaxial anisotropy of the system.

In order to analyze what processes occur at the crossing point, we consider the time dependence of the state when the field is swept,

$$H(t) = H_0 + ct, \tag{1}$$

where H_0 is the initial value of the field and c is the sweeping rate of the field. The magnets of Mn_{12} and Fe_8 are regarded as an ensemble of independent $S = 10$ spins which are regularly located in crystals. Thus, here we adopt here a Hamiltonian,

$$\mathcal{H} = -DS_z^2 - H(t)S_z + Q, \tag{2}$$

where Q represent additional terms due to the details of the material. Here, we simply take

$$Q = -\Gamma S_x, \tag{3}$$

which causes quantum fluctuation, *i.e.*, $[S_z, \mathcal{H}] \neq 0$. For detailed analysis we need further terms, such as $E(S_x^2 - S_y^2)$ for Fe_8 because of lack of symmetry in the hard plane. For Mn_{12} we may consider $(S^+)^4 + (S^-)^4$, *etc.* When the field is swept as Eq. (1), the system undergoes the so-called nonadiabatic transition. That is, if the sweeping rate is small the state changes adiabatically, namely it stays in the ground state at the temporal value of the field. In this process the magnetization of the state changes with the field and thus it represents the tunneling process. On the other hand, if the field is swept rapidly, the state can not follow the change of the field and stays in almost the same state as the initial one which is called nonadiabatic transition.[8)] This process corresponds to channel B in Fig. 1 while the adiabatic process corresponds to channel C. In the intermediate rate of the sweeping, the state after the crossing point is given by a linear combination of the states in the two channels

$$|\Psi(t)\rangle = b|\mathrm{B}\rangle + c|\mathrm{C}\rangle, \tag{4}$$

where $|\mathrm{B}\rangle$ and $|\mathrm{C}\rangle$ represent the states in channel B and C, respectively. The probability to observe the system in channel B is

$$p_{\rm LZS} = |b|^2 = 1 - \exp\left(-\frac{\pi(\Delta E)^2}{\hbar g \mu_{\rm B} \Delta M} c\right), \quad (5)$$

as described by Landau, Zener and Stückelberg[9] in 1932. Here ΔE is the energy gap at the crossing point, ΔM is the change of the magnetization $|m - m'|$ and g is the g-factor of the magnet. We have studied the quantum-mechanical aspects of the transition in nanoscale magnets in molecular magnets from the view point of nonadiabatic transition.[10] The energy gap is related to the resonant frequency,

$$\omega = \Delta E/\hbar, \quad (6)$$

which could be observed by a resonant method. Thus, in principle, the energy gap is obtained by two methods, *i.e.*, observation of the change in the magnetization after the sweeping field and observation of the resonance with an oscillating field, which are complementary to each other.

Here, let us consider the time scale of the processes. If the energy gap ΔE is 10^{-m} K, the resonant frequency is given by

$$\omega = 10^{-m}\,{\rm K}/\hbar = 1.31 \times 10^{11-m}[{\rm s}^{-1}] \quad (7)$$

and the probability of the adiabatic motion $p_{\rm LZS}$ is given by

$$p_{\rm LZS} = 1 - \exp\left(-\frac{0.15 \times 10^{12-2m}}{\Delta M \cdot c}\right), \quad (8)$$

where we take $g = 2$ and c is given in units of [T/s] and ΔM is given by magnetic moment. If we take $\Delta E = 10^{-5}$, the correspond values are $\omega = 1.31 \times 10^6$ and $p_{\rm LZS} = 0.53$ for $\Delta M = 20$ and $c = 1\,[{\rm t/s}]$. To observe the nonadiabatic transition clearly p is preferable to be different from 1.0 (*i.e.*, adiabatic limit). Thus for larger energy gap, we need a faster sweeping rate of the field. In particular, for the measurements in microscopic molecular magnets, the cases with $\Delta E \sim 10^{-7}$ are studied.

If the energy gap is very small, the pure quantum process would be easily affected by the noise, although the fundamental processes are given by a combination of nonadiabatic transitions.[11] Thus, we consider the effects of noise on the processes and try to extract information about the quantum mechanical processes.

Quantum hysteresis in nanoscale magnets

In particular, experimental data show a strong temperature dependence at $T = 1$–$2\,{\rm K}$, although the magnetization curve still shows stepwise increases, which is a characteristic of resonant tunneling. We have studied the effects of the environment using a quantum master equation which is derived by tracing out the degree of freedom of the thermal bath from the density matrix of the whole system.[12,13]

For such a temperature dependence, the idea of 'thermally assisted resonant tunneling' was introduced, where tunneling between the excitation levels play an important role. Furthermore it has been pointed out that the amount of relaxation at the resonant points change alternately in the resonant tunneling process, which has been called the 'parity effect'. We have pointed out that the parity effect can be naturally interpreted from the view point of the nonadiabatic transition of the excited states.[14]

When the temperature is lowered the magnetization curve saturates at a certain temperature. There we may consider that the process represent a pure quantum process. However, the curve has been found to be affected by the dissipative environment inevitably. At such a low temperatures the activated process is suppressed and is almost prohibited. However relaxation processes which do not require activation energy take place regardless of the temperature. This additional relaxation process must be taken into account to analyze the quantum-mechanical transition probability, $p_{\rm LZS}$. We demonstrated an example of such a process and also showed that $p_{\rm LZS}$ can be obtained if we analyze date appropriately. We call this temperature-independent magnetization process 'deceptive nonadiabatic process'.[15]

The above observations were made in Mn_{12} and Fe_8 which are regarded as $S = 10$ spin. Recently V_{15} has been studied. Although this molecule has 15 V atoms, the total magnetization of the ground state is 1/2. Thus it is very simple model and we expect that the features of quantum dynamics can be clearly seen. However, it has been found that an effect of the environment plays an important role. Actually, in this molecule the energy gap is rather large ($\Delta E \sim 0.1\,{\rm K}$) and the probability $p_{\rm LZS} \simeq 1$, as we find from Eq. (5). In the experiment a deviation from the adiabatic behavior is still found. This deviation is interpreted in term of the lack of a number of phonons in the crystal ('phonon-bottleneck' effect). We have demonstrated this phenomena and also studied the magnetization curve as a function of the sweeping velocity including fast sweeping, where the LZS transition becomes dominant.[16,17]

Spin manipulation

In so-called spin-gap systems, such as the Haldane and dimer systems, localized magnetizations are induced near the inhomogeneity of the lattice. Because those magnetizations behave almost independently, it would be an interesting problem to manipulate such a moment by a time-dependent external field.[18,19] This problem maybe contribute to the realization of quantum computing, although it seems difficult practically. We have studied a spin dynamics related to the quantum computing and have shown that the real-time dynamics is rather unstable against small changes which are ignored in usual study of quantum computing.[20]

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