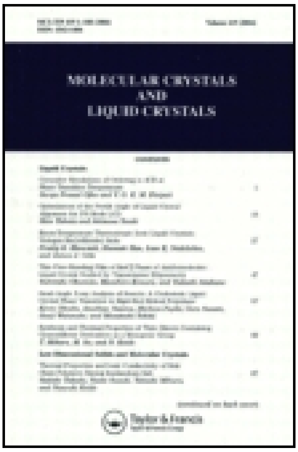

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Quantum Mechanical Dynamics in Microscopic Structures

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Dynamics of the magnetization of microscopic molecules in a sweeping field is studied, where the adiabatic motion in quantum dynamics and thermal incoherent effects are combined and various interesting phenomena appear. We have also studied possible mechanisms of the tunneling in half-integer spin systems, and discuss methods to manipulate the spin state by making use of characteristics of quantum dynamics.

Keywords Microscopic magnetic molecules; Adiabatic change; Landau-Zener-Stückelberg transition; Magnetic Foehn effect

INTRODUCTION

By recent developments of synthesis of microscopic high spin molecular magnets, such as Mn_{12}[1] and Fe_8[2, 3], etc., it has been possible to study direct evidences of quantum mechanical dynamics in discrete energy levels. In particular, so called resonant tunneling phenomena were discovered, where the relaxation of magnetization occurs only at discrete values of magnetic fields at which two energy levels of different magnetization cross. This causes a stepwise magnetization process. In order to understand mechanism of this process, the idea of the quantum tunneling has been introduced in the potential picture[4], which explained qualitative features of the phenomena. In order to study the quantitative properties, we need to study the explicit hamiltonian of the system. For this purpose we introduced the idea that the tunneling phenomenon is a kind of adiabatic process when the field is changed. The fundamental process is described by the Landau-Zener-Stückelberg(LZS) mechanism[5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15], where the probability to find the system in the adiabatic state depends on the energy gap ΔE of the avoided level crossing structure and the sweeping velocity of the external field $c = dH/dt$

$$p_{\rm LZS} = 1 - \exp(-\frac{\pi(\Delta E)^2}{2c}). \tag{1}$$

Beside the high spin molecules, quantum spin dynamics is observed in low spin molecules such as V_{15}[16, 17] where the adiabatic change of magnetization is described more clearly. The local magnetizations are provided not only by micro molecules but also by spatial inhomogeneity of the lattice in so-called 'spin gap systems' which are non magnetic states such as the dimer state in bond alternate antiferromagnetic Heisenberg systems[18] or the Haldane state in $S = 1$ antiferromagnetic Heisenberg systems[19, 20, 21]. In systems of such local magnetizations, it is expected that the quantum dynamics would be observed.

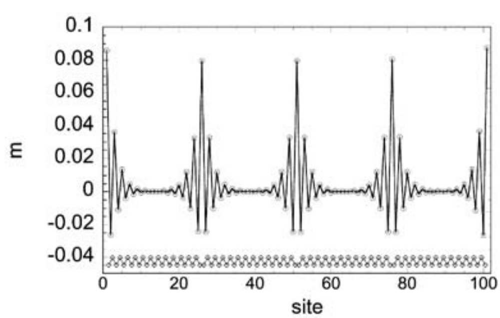

FIGURE 1. Local magnetic structure induced around defects of alternation of the bonds[18]

QUANTUM DYNAMICS OF LOCAL MAGNETIZATION

The energy scale of the quantum dynamical process is so small that effects of noise from environments play an essential role for the observed phenomena. To study such effects of noise we have developed two types of methods. One is the quantum Langevin equation where we solve the Schrödinger equation with a time dependent field which represents random noise. For each process of noise, the system shows various peculiar phenomena due to the quantum interference and the process seems stochastic. However if we average over many processes, we obtain a process representing a dissipative process. By this method, we studied the origin of so called square-root-time behavior of Mn_{12} and Fe_8[22] in static resonant field.

We also study the modification of the transition probability of LZS, when we sweep the magnetic field in a random field $\boldsymbol{h}(t)$

$$\mathcal{H} = 2\Gamma S^x - H(t)S^z - \boldsymbol{h}(t)\cdot\boldsymbol{S}, \tag{2}$$

where $H(t) = -H_0 + ct$ is the sweeping field and

$$\langle h_\alpha(t)\rangle = 0, \quad \text{and} \quad \langle h_\alpha(0)h_\alpha(t)\rangle = \frac{A_\alpha^2}{2\gamma}e^{-\gamma t}, \quad \alpha = x, y, \quad \text{or}, \quad z, \tag{3}$$

where A and γ denote the amplitude and relaxation rate of the noise, respectively. We have studied how the state starting from the ground state at $H = -H_0$ changes while the field is swept. The probability staying in the ground state is given by the LZS formula when $A = 0$. When we sweep the field in the noise field, the probability staying in the ground state is modified as a function of A and γ.[23] From this modified probability p_{eff} we may define the effective energy gap in the analogy of (1) $\Delta\bar{E}_{\text{eff}}$ as

$$\Delta\bar{E}_{\text{eff}} = \sqrt{-\frac{2c}{\pi}\ln(1-p_{\text{eff}})}. \tag{4}$$

The ratio $\Delta \equiv \Delta\bar{E}_{\text{eff}}/\Delta E_0$ describes the degree of modification. ΔE_0 is the gap ($\Delta E_0 = 2\Gamma$) for the pure case. In the case of $S = 1/2$ we find a relevant modification as shown in Figure 2, where we show the dependence keeping $B^2 \equiv (A_x^2 + A_y^2 + A_z^2)/2\gamma$ to be constant ($B = \Gamma$), because B represents the physical amplitude of the noise. When the sweeping velocity is fast the ratio Δ saturates. In this limit Δ is analytically given as

$$\Delta = \sqrt{1 + \frac{B^2}{6\Gamma}}. \tag{5}$$

On the other hand, for $S = 10$, we found that the ratio shows little dependence on the amplitude as far as the sweeping velocity of the external field is fast enough. In experiment[3], the transition probability depends on the arrangement of isotope atoms by which the property of the noise is changed. Thus, we have to study further for the source of the dependence.[24]

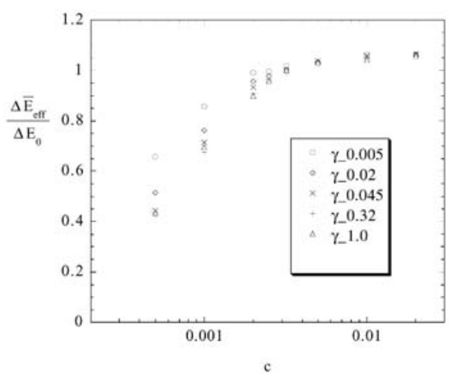

FIGURE 2. Dependence of the transition probability to non-adiabatic state on the amplitude of the noise field.[23]

The other method is the quantum master equation which describes equation of motion of the density matrix where both quantum dynamics and dissipative dynamics due to the contact with the thermal bath are taken into account.[25] By this method we have shown that the effect of thermal relaxation causes an essential difference of amount of the change of the magnetization in stepwise magnetization process even at very low temperatures, which we called deceptive resonant tunneling effect.[12] We also study the mechanism of the plateau in the magnetization process of V_{15} which has been studied as 'Phonon-bottleneck phenomena' by Chiorescu et al. [16, 17], and proposed that this is one of very universal phenomena which appear when the adiabatic process is exposed to heat inflow, and we, in general, call such phenomena 'Magnetic Foehn (MF) phenomena'. Using this idea we analyzed the magnetization process of Fe-rings in the pulsed magnetic field.[26, 27] In Figure 3, we show temperature dependence of dM/dH in a pulsed field. There we see regular big peaks which correspond to the change of magnetization by 1. At the high temperature, we find extra peaks besides the main peaks. The positions of these satellite peaks depend on whether the field is increasing or decreasing. At low temperature (Figure 3(b)), the satellite peaks disappear. The data in Figures 3 explain the experimental data of Fe_{12} very well.[28, 29] So far the strength of the coupling to the thermal bath is quantified by the relaxation times such as T_1 and T_2. The satellite peaks due to the MF effect can be a new quantity to characterize the strength.

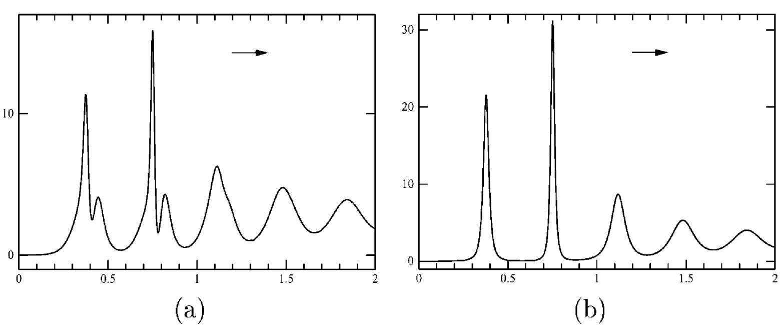

FIGURE 3. dM/dH vs. the increasing magnetic field for a system modeling Fe_{12} at (a)1.3K and (b)0.09K.

Although in V_{15} we find the adiabatic change of magnetization clearly, generally the tunneling is thought to be prohibited because of the time reversal symmetry which causes the Kramers doublet at $H = 0$ and the level does not form the avoided level crossing structure. Signs of adiabatic transition have been observed in other half-integer spin systems such as Mn_4 ($S = 9/2$)[30]. We studied the mechanism of adiabatic change of magnetization in half-integer spin systems.[31] In particular for antiferromagnetic triangle system, the ground state is four-fold degenerate and when the rotational and inversion symmetries are broken, for example, as

$$\mathcal{H} = \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1 + \alpha(S_1^x S_2^z - S_1^z S_2^x) \tag{6}$$

two sets of avoided level crossing structures appear. Both of them cross at $H = 0$ and thus the Kramers theory is satisfied. But the magnetization processes in both avoided level crossing structures follow the LZS mechanism individually.

DISCUSSION

Finally let us discuss possible utilization of the adiabatic change of magnetization. The main difference between the thermal change (Arrhenius process) and quantum change (tunneling) is the dissipation of the energy. In the memory devices, the hysteresis phenomenon gives key ingredient to keep the 'memory'. In order to change the state a string magnetic field which is larger than the spinodal point is applied. Thus at every switching, the heat is generated in the system by the so-called hysteresis-loss, which makes the temperature of the system rise. Here let us consider a new way of switching by making use of adiabatic transition. Microscopically, the hysteresis is a phenomenon due to the non-adiabatic process where the system can not follow the change of the external field. As we know in the experiment of Fe_8 in the transverse field, the energy gap can be controlled by the external field. If the gap at the avoided level crossing point is small, the adiabatic change requires a very slow change of the parameter. This means that it takes a long time to switch the state and the hysteresis appears by which the memory is kept in the same state. On the other hand, if the gap is large then the state easily follows the change of the external field, where the switch is done without energy loss. In order to control the gap we need to apply some external field, e.g., the transverse field. However, the energy to change the external field does not cause heating the system because the work is done by the external field and no dissipative energy is released in the system. Making use of the above mentioned mechanism, say quantum switching mechanism, we may make a memory system with very small energy loss. This kind of new developments of technology would bring a fruitful future of systems with macroscopic assembly of quatum mechanically interacting microscopic local magnets, or more generally of local energy structures.

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