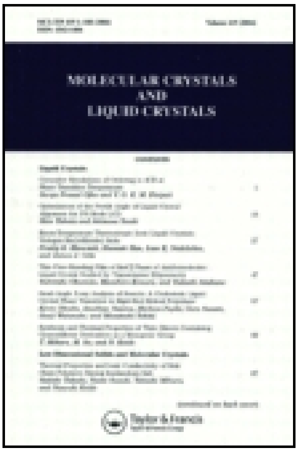

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Quantum Dynamics and Response in Nanoscale Spin Systems

Seiji Miyashita [a], Masamichi Nishino [a] & Keiji Saito [a]
[a] Department of Applied Physics, University of Tokyo, 7-3-1 Bunkyo-ku, Tokyo, 113-8656, Japan

Version of record first published: 18 Oct 2010

To cite this article: Seiji Miyashita, Masamichi Nishino & Keiji Saito (2002): Quantum Dynamics and Response in Nanoscale Spin Systems, Molecular Crystals and Liquid Crystals, 376:1, 327-334

To link to this article: http://dx.doi.org/10.1080/10587250210725

Mol. Cryst. Liq. Cryst., Vol. 376, pp. 327-334

1058-725X/02 \$12.00 + .00

Quantum Dynamics and Response in Nanoscale Spin Systems

SEIJI MIYASHITA, MASAMICHI NISHINO and KEIJI SAITO

Department of Applied Physics, University of Tokyo 7-3-1 Bunkyo-ku, Tokyo, 113-8656 Japan

We study the quantum dynamics with the effect of dissipative environments in nanoscale magnetic systems, e.g. the molecular magnets such as Mn_{12}, Fe_8 and V_{15} and also the locally induced magnetizations in the so-called gaped spin systems. We analyze the dynamics from a view point of nonadiabatic transition at the avoided level crossing points which is called the resonant tunneling point. By the quantum master equation, we study effects of the heat contact on an adiabatic process (magnetic Föhn effect) which is relevant for systems with a large gap such as V_{15}. By the quantum Langevin equation we study how successive random crossings cause a nonexponential decay of the magnetization in the systems with very small energy gap such as Mn_{12} and Fe_8.

Keywords nanoscale magnets, quantum dynamics, nonadiabatic transition quantum master equation, quantum Langevin equation

INTRODUCTION

Since the technology to treat nanoscale objects has been developed, it has become possible to observe the real time behavior which reflects the quantum mechanical dynamics of the system. For example, we see the quantum hysteresis in nanoscale molecular magnets such as Mn_{12}[1, 2, 3] and Fe_8[4, 5, 6] which is called the resonant tunneling. This phenomenon is directly attributed to the nonadiabatic transitions at the avoided level crossing points [7]. The transition probability was obtained by Landau, Zener and Stückelberg years ago[8]. In realistic materials, however, there exist various effects which cause the decoherent process, and the pure quantum mechanical process is easily smeared out[9]. In this paper we will study the following two typical examples of dissipation effects.

When the energy gap is large, the system shows an almost adiabatic transition. An interesting structure has been found in the magnetization process of V_{15} where the probability of the nonadiabatic transition is almost zero[10]. That is, the magnetization curve shows a plateau just after the resonant point. It is found that this plateau is due to an inflow of the heat from the thermal reservoir. We will see the sweeping velocity dependence of the magnetic process.

In the so-called gaped spin systems, such as the bond-alternate antiferromagnetic Heisenberg chain, almost free local magnetizations are induced when the lattice has some inhomogeneity [11, 12]. There magnetizations responds to the sweeping field in a similar way to the case of V_{15} [12].

For the molecules Mn_{12} and Fe_8, the energy gap at the resonant point is very small and the random noise on the external field causes successive nonadiabatic transitions. We simulate such processes by solving Schrödinger equation with a noise (quantum Langevin equation), and analyze the data by a simplified master equation. There we study how the random successive transition causes nonexponential decay of the magnetization, which would be relevant for the square-root time dependences observed in experiments.

MAGNETIZATION PROCESS IN A SWEEPING FIELD

In nanoscale magnets, the energy structure consists of discrete levels because of small number of atoms. When the external field is swept and cross the resonant point, the system undergoes a nonadiabatic transition between the two levels. This situation can be modelized by a two-level system, whose Hamiltonian is

$$\mathcal{H} = -\Gamma\sigma_x - h(t)\sigma_z. \tag{1}$$

Let us set the initial value of the field at far from the resonant point, say $-H_0$. The ground state $|\mathrm{G}\rangle$ at this point is almost the down state $|-\rangle$. Here we use the eigenstate of σ_z as the basis set

$$\sigma_z|\pm\rangle = \pm|\pm\rangle. \tag{2}$$

When we sweep the field with a velocity c, the nonadiabatic transition occurs near the crossing point and the population of the ground state after the crossing is given by LZS as

$$p_{\mathrm{AD}} = 1 - \exp\left(\frac{\pi(\Delta E)^2}{2c}\right), \tag{3}$$

and the population of the excited state $|\mathrm{E}\rangle$ is $p = 1 - p_{\mathrm{AD}}$. Here the scattering occurs only in very narrow region of the field (resonant point) and the scattering rate depends on the velocity ($\propto c^{-1}$). These properties are the characteristics of the nonadiabatic transition, and have been observed in experiments, and the relation (3) has been used to estimate the energy gap ΔE[5].

In realistic materials, this pure quantum mechanical process is affected by the effects of dissipative environments. In order to study such effects, we exploit the quantum master equation where the effect of the thermal bath is taken into account in a perturbational way[13]. When the noise causes strong effects we study the dynamical process with noise directly by the quantum Langevin equation. In this paper we will study two typical examples of dissipation effects, i.e. the case where the energy gap is large and the system shows almost adiabatic process, and the case where the gap is very small and the noise causes the field to cross the resonant point successively.

MAGNETIC FOEHN EFFECT

Chiorescu, et al. have found almost perfect adiabatic transition in the system of V_{15}[10]. There they found a plateau in the magnetization process just after the crossing, which shows the deviation from the adiabatic behavior. The nonadiabatic transition process maybe a possible candidate of the origin for this deviation. However, when the sweeping velocity decreases, the magnetization at the plateau decreases, which means that the deviation increases. This is the opposite direction to that we expect in the nonadiabatic transition process. This process has been found as a phenomenon due to the insufficient heat inflow during the process. Chiorescu, et al. attributed the shortness of the heat to the small heat capacity of the lattice at low temperatures, and called it 'phonon-bottleneck effect'. We simulated the system (1) with a contact to the thermal reservoir by quantum master equation

$$\frac{\partial \rho(t)}{\partial t} = \frac{1}{i\hbar}\left[\mathcal{H}, \rho(t)\right] - \lambda\left(\left[X, R\rho(t)\right] + \left[X, R\rho(t)\right]^{\dagger}\right), \qquad (4)$$

where X is a system operator through which the system and the bath couple. The first term of the right-hand side describes the pure quantum dynamics of the system while the second term represents effects of environments at a finite temperature. There R is an operator which reflects the effects of the thermal reservoir[13]. We studied overall sweeping velocity dependence of the magnetization curve. There we find two regions: When the velocity is very large the system undergoes the nonadiabatic transition and the plateau appears with the magnetization (Fig.1(a))

$$m_{\rm LZS} = M_0(1 - 2p_{\rm AD}). \qquad (5)$$

Here the the deviation is brought coherently and we see the precession between the states. When the velocity decreases, $m_{\rm LZS}$ increases (Fig.1(b)).

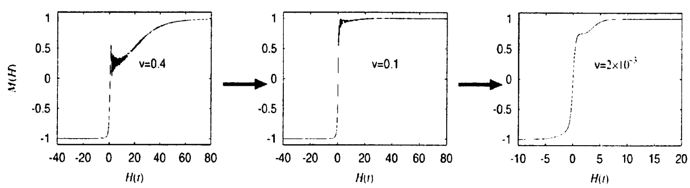

FIGURE 1 Sweeping velocity dependence of the magnetization process. (a) nonadiabatic process, (b) almost adiabatic process, and (c) magnetic Föhn phenomenon

On the other hand, we find another plateau when the velocity decreases further (Fig.1(c)). The plateau in this case decreases as the velocity decreases and finally the magnetization tends to the isothermal one, i.e.

$$m_{\rm therm} = M_0 \frac{H}{\sqrt{H^2+\Gamma^2}} \tanh(\frac{\sqrt{H^2+\Gamma^2}}{k_BT}). \qquad (6)$$

We find that this dependence does not depends on the details of the system, and is general for a wide range of systems[14]. The plateau at the slow sweeping region can be understood as follows. The ratio of the populations p_E/p_G at the initial field is very small because the energy gap there is much larger than the temperature. However the gap at the crossing point is very small and the ratio corresponds to a very low temperature, i.e.

$$\frac{p_E}{p_G} = \exp\left(-\frac{2H_0}{k_BT}\right) = \exp\left(-\frac{\Delta E}{k_BT_{\rm eff}}\right), \quad T \gg T_{\rm eff}. \qquad (7)$$

Thus, there is a big temperature difference between the system and the environments, and the heat tends to flow in the system. This inflow of heat causes the deviation from the adiabatic behavior, and the population at the excited state increases. Here the deviation is brought incoherently and the precession does not occur in this case. This situation is similar to the Föhn effect in the meteorology. That is, when the wet air climbs up a mountain and undergoes the adiabatic expansion and the temperature decreases. There the vapor becomes the rain and releases the heat. When the air goes down the mountain, the temperature becomes higher than that before. Noting this analogy, we call the present phenomenon 'magnetic Föhn phenomenon'[15].

NONEXPONENTIAL DECAY AT RESONANT POINTS

When the amplitude of noise is larger than the resonance region, the noise causes successive crossings which brings successive nonadiabatic transitions. In this section we study how the magnetization decays in the noise[18]. Because the relevant transition occurs only when the field crosses the resonant point, the relaxation of the magnetization is mainly governed by the frequency of the crossings. Here we assume that the external field $H(t)$ behaves as a random walk with a weak restoring force, which is expressed by the Ornstein-Uhlenbeck process

$$\frac{dH}{dt} = -\gamma H(t) + \eta(t), \tag{8}$$

where $\eta(s)$ is a white gaussian noise $\langle \eta(t) \rangle = 0$, and $\langle \eta(t)\eta(s) \rangle = 2D\delta(t-s)$. The pure random walk, i.e. the Wiener process, corresponds to the case without the damping factor $\gamma = 0$. The distribution of $H(t)$ for this process is given by

$$P(H,t) = \frac{1}{\sqrt{2\pi\sigma^2(t)}} \exp\left(-\frac{(H - \langle H \rangle)^2}{2\sigma^2(t)}\right), \tag{9}$$

where

$$\sigma^2(t) = \frac{D}{\gamma}\left[1 - \exp(-2\gamma t)\right]. \tag{10}$$

We solve the time dependent Schrödinger equation with various sample of $H(t)$, and study the dynamics of the magnetization. In Fig. 2, we show the time dependence of the magnetization $\langle M(t) \rangle$ averaged over 10000 samples. At the early stage, where the process is regarded as the Wiener process, we find a stretched exponential function

$$M = M_0 \exp\left(-\alpha \frac{(\Delta E)^2}{\sqrt{D}} \sqrt{t}\right), \tag{11}$$

while at the late stage where the noise distribution reaches the stationary one, we find a normal exponential decay

$$M = M_1 \exp\left(-\beta (\Delta E)^2 \sqrt{\frac{\gamma}{D}} t\right), \tag{12}$$

where α and β are positive constants. Making use of a master equation for the populations in the ground and the excited states, we derived a formula for $\langle M(t)\rangle$ under assumption that each crossing gives the same mixing of the populations between the states

$$\langle M(t)\rangle = \exp\left(-\frac{p_0\nu_0(\Delta E)^2}{\sqrt{2\pi\gamma D}} \log\left|\frac{\sqrt{1-e^{-2\gamma t}}-1}{\sqrt{1-e^{-2\gamma t}}+1}\right|\right). \tag{13}$$

This gives the relations (11) and (12) in limiting cases, putting

$$\alpha = \tilde{p} \qquad \text{and} \qquad \beta = \frac{\tilde{p}}{\sqrt{2}} \quad \text{where} \quad \tilde{p} \equiv \frac{2p_0\nu_0}{\sqrt{\pi}}. \tag{14}$$

Furthermore, (13) fits the overall behavior shown in Fig.2 very well.

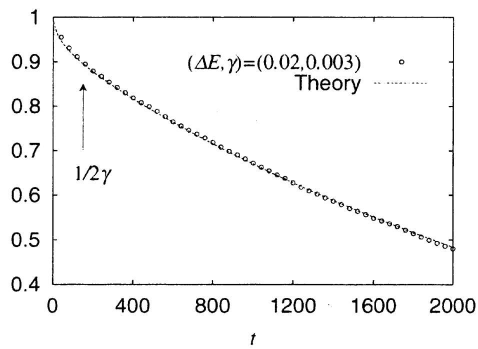

FIGURE 2 $\langle M(t)\rangle$ obtained by simulation and the formula (13)

In the nanoscale molecular magnets, various types of $\sqrt{t}$ dependence have been found [16, 17]. For example, in the experiment of Fe_8[5, 6], the magnetization increases from zero proportionally to $\sqrt{t}$

when they apply a weak field. For the hole digging phenomena in this experiment, the present study predicts that both of the depth and the width of the hole increase with $\sqrt{t}$, which is apparently consistent with the experiment although further detail analysis is necessary[6]. Relaxation of the saturated magnetization also shows the $\sqrt{t}$ dependence which is very similar to that in Fig.2 [4, 3].

Acknowledgments

We thank Professors B. Barbara, H. De Raedt, Y. Ajiro and E. Chudonovsky and also Drs. W. Wensdorfer and I. Chiorescu for their valuable discussions. The present work is partially supported by the Grant-in-Aid for Scientific Research.

References

[1] L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli and B. Barbara, Nature **383**, (1996) 145. B. Barbara, et al. J. Phys. Soc. Jpn. Suppl. **69**, 383 (2000).

[2] J. M. Hernandez, X. X. Zhang, F. Luis, T. Tejada, J. R. Friedman, M. P. Sarachik, and R. Ziolo, Phys. Rev. B **55**, 5858 (1997).

[3] L. Thomas, et al. Phys. Rev. Lett. **83** 2398 (1999).

[4] C. Sangregorio, T. Ohm, C. Paulsen, R. Sessoli, and D. Gatteschi, Phys. Rev. Lett. **78**, 4645 (1997). T. Ohm, C. Sangregorio, C. Paulsen, Euro. Phys. J. B **6**, 195 (1998); T. Ohm, C. Sangregorio, C. Paulsen, J. Low Temp. Phys. **113**, 1141 (1998).

[5] W. Wernsdorfer and R. Sessoli, Science **284**, 133 (1999). W. Wernsdorfer, et al, Phys. Rev. Lett. **82**, 3903 (1999). W. Wernsdorfer, et al, J. Phys. Soc. Jpn. Suppl. **69**, 375 (2000).

[6] W. Wernsdorfer, A. Caneschi, R. Sessoli, D. Gatteschi, A. Cornia, V. Villan, C. Paulsen, Phys. Rev. Lett. **84** 2965 (2000).

[7] S. Miyashita, J. Phys. Soc. Jpn. **64**, 3207 (1995) ; **65**, 2734 (1996). H. De Raedt, S. Miyashita, K. Saito, D. García-Pablos, and N. García, Phys. Rev. B, **56** 11761 (1997). S. Miyashita, K. Saito, and H. De Raedt, Phys. Rev. Lett. **80**, 1525 (1998).

K. Saito, S. Miyashita, and H. De Raedt, Phys. Rev. B **60** 14553 (1999). S. Miyashita, K. Saito, H. Kobayashi, and H. de Raedt, J. Phys. Soc. Jpn. Suppl. **69**, 395 (2000).

[8] L. Landau, Phys. Z. Sowjetunion **2**, 46 (1932). C. Zener, Proc. R. Soc. London, Ser. A **137**, 696 (1932). E. C. G. Stückelberg, Helv. Phys. Acta **5**, 369 (1932).

[9] Y. Kayanuma and H. Nakayama, Phys. Rev. B **57**, 13099 (1998). D. A. Garanin and E. M. Chudnovsky, Phys. Rev. B**56**, 11102 (1997).

[10] I. Chiorescu, W. Wernsdorfer, A. Müller, H. Bögge, and B. Barbara, Phys. Rev. Lett. **84** 3454 (2000).

[11] P. Roos and S. Miyashita, Phys. Rev. B**59**, 13782 (1999). M. Nishino, et al., Phys. Rev. B**61**, 4033 (2000).

[12] M. Nishino, et al., Phys. Rev. B**62**, 9463 (2000).

[13] K. Saito, S. Takesue, and S. Miyashita, Phys. Rev. E**61** 2397 (2000).

[14] Y. Shapira, et al., Phys. Rev. B**59**, 1946 (1999). K. L. Taft,et al., J. Am. Chem. Soc. **116**, 823 (1994).

[15] K. Saito and S. Miyashita: condmat 0004027.

[16] N. V. Prokof'ev and P. C. E. Stamp, Phys. Rev. Lett. **80**, 5794 (1998), J. Low Temp. Phys. **113**, 1147 (1998).

[17] E. M. Chudnovsky, Phys. Rev. Lett. **84**, 5676 (2000), N. V. Prokof'ev, P. C. E. Stamp, ibid, 5677, and W. Wernsdorfer, C. Paulsen, R. Sessoli, ibid, 5687.

[18] S. Miyashita and K. Saito: condmat 0005013.