Magnetic Dynamics of Nanoscale Magnets: From Classical to Quantum

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The various uncommon properties of magnetic nanoparticles (Stoner nanoparticles) and molecular magnets have attracted much attention in industry as well as in the academic community. In this review we begin with going through important experiments in this field and possible applications of these materials. Then we introduce the theoretic models currently used for Stoner nanoparticles and molecular magnets respectively. They are the Stoner-Wohlfarth model and the Landau-Lifshitz-Gilbert(LLG) theory in the former case and the quantum macrospin model for the latter one. In the end we give a brief discussion on the shortcomings of these models and propose a possible approach to build a mesocopic magnetic dynamical theory.

Keywords: magnetic nanoparticles, molecular magnets, magnetic data storage, resonant tunneling

I. INTRODUCTION

In recent years, nanoparticles and nano-thin films with large magnetic moments and magnetic anisotropy have attracted much interest. In these materials, all or most atoms' magnetic moments are strongly coupled and aligned in the same direction, creating a single magnetic domain. And because of its mesoscopic scale, these materials usually exhibit non-classical behaviors. From the application point of view, these kinds of materials are optimal candidates for high density data storage and, possibly, quantum computation. Also from the theory point of view, study of the properties of magnetic nanoclusters requires deeper understanding of the transition from the quantum to the classical world.

Generally speaking, there are two kinds of magnetic nanoparticles under discussion. One of them is a crystal or semi-crystal composed of magnetic ions such as Fe or Mn. Its total spin is large enough that it can be treated classically. We call these kinds of particles Stoner nanoparticles because they can be properly described by the classical model proposed by Stoner and Wohlfarth [1]. The first experiment in this area was done by S. Sun, et al.[2], who discovered the spontaneous self-organization of magnetic FePt nanoparticles on a surface (Fig. 1). It is generally believed that if magnetic media based on such nanoparticles could be made, the data storage density would be as much as 100 times larger than that in current hard drives. This prediction is based on the small size and large magnetic anisotropy of these nanoparticles, which can help to go beyond the magnetic storage limit set by the phenomenon known as superparamagnetism[3].

Besides storage density, high speed of reading and writing is also required by future information storage applications. Data reading and writing involve switching the magnetization of magnetic storage cells (magnetization reversal). As far as Stoner nanoparticles are concerned, a workhorse to study the dynamics of magnetization is classical Landau-Lifshitz-Gilbert theory [4][5].

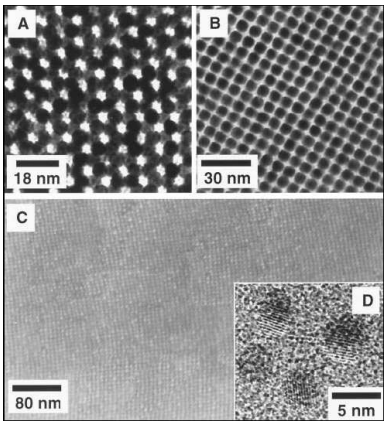

FIG. 1: (**A**) TEM micrograph of a 3D assembly of 6-nm as-synthesized $Fe_{50}Pt_{50}$ particles deposited from a hexane/ octane (v/v 1/1) dispersion onto a SiO-coated copper grid. (**B**) TEM micrograph of a 3D assembly of 6-nm $Fe_{50}Pt_{50}$ sample after replacing oleic acid/oleyl amine with hexanoic acid/hexylamine. (**C**) HRSEM image of a $\sim$ 180-nm-thick, 4-nm $Fe_{52}Pt_{48}$ nanocrystal assembly annealed at 560°C for 30 min under 1 atm of N_2 gas. (**D**) High-resolution TEM image of 4-nm $Fe_{52}Pt_{48}$ nanocrystals annealed at 560°C for 30 min on a SiO-coated copper grid.(Figure from [2])

Another kind of magnetic particle is the so-called molecular magnet, which is a single large molecule with a nonzero magnetic moment. One of these is the nowadays famous $Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4$, or Mn_{12} for short. Mn_{12} was first synthesized by Lis [6] in 1980. He found that the compound contains 4 Mn^{4+} (S=3/2) ions in a central tetrahedron surrounded by eight Mn^{3+} (S=2)

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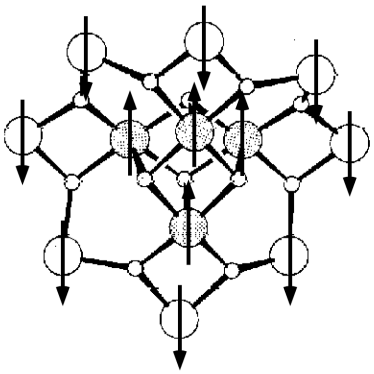

FIG. 2: Structure of magnetic core of the Mn_{12} molecule. Only the Mn^{4+} (large shaded circles), Mn^{3+} (large open circles) and oxygen (small circles) ions are shown. The arrows indicate the configuration of the spins that results in a total spin of 10 for the molecule. The diameter of Mn_{12} is ~ 17Å

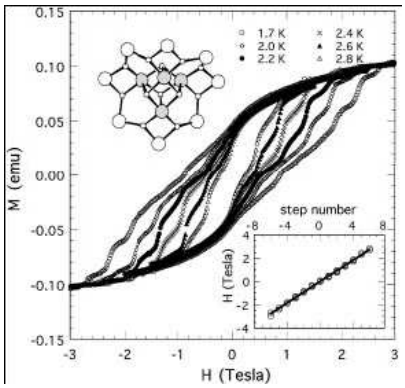

FIG. 3: Magnetization versus applied field. Inset shows values of field where jumps in the magnetization are observed. (Figure from [7])

ions in a non-coplanar ring, as shown in Fig. 2.

The Mn ions are strongly superexchange-coupled through oxygen bridges. One distinctive property of Mn_{12} is that the coupling between Mn ions within this molecule is so strong that at low temperatures it can be treated as a single macrospin with $S = 10$.

In 1995 J. Friedman and collaborators measured the hysteresis loops of Mn_{12} at temperatures from 1.7 to 3K (Fig. 3) [7]. All the hysteresis curves exhibited clear steps, which had never been seen in any other magnetic systems. Actually these steps are evidence for a quantum phenomenon much sought after by theorists and experimenters in recent years: the tunneling of a spin through a potential barrier from one orientation to another. The importance of this experiment lies in that it is one of few direct evidences for the so-called macroscopic quantum tunneling (another worth mentioning is tunneling in a Josephson Junction provided by Clark, et al. [8]). For the explanation of these steps we will go into detail in Sec. III.

After Mn_{12}, a lot of other molecular magnets were discovered, such as Fe_8, V_{15}, Ni_{12} etc. It is worthwhile to mention that, because of the apparent quantum nature of these molecular magnets and their mesoscopic size, they are now considered promising candidates for materials of real life quantum computation [9].

The remaining of this paper are organized as follows: In Sec. II we will briefly introduce the classical Stoner-Wohlfarth model and Landau-Lifshitz-Gilbert theory, as well as their application to the Stoner nanoparticles. In Sec. III we will introduce the quantum macrospin model of molecular magnets, taking Mn_{12} and Fe_8 as examples. Finally we discuss the necessity and general requirements of building a mesoscopic magnetic dynamical theory in a bottom-up way.

II. CLASSICAL MODEL AND LANDAU-LIFSHITZ-GILBERT THEORY

A. Static Model: Stoner-Wohlfarth Theory

Because of their large magnetic moment and small size, the nanoparticles of the first category mentioned in the introduction, such as FePt, can be treated without much loss of accuracy as the classical Stoner-Wohlfarth particle. In this subsection we will introduce the Stoner-Wohlfarth theory and, based on that, an explanation of the magnetic hysteresis curve, as a preparation for the LLG theory in Sec. II B.

Stoner and Wohlfarth proposed a simple model to explain the magnetization hysteresis curve of heterogeneous alloys in 1948 [1]. We now know the generally accepted explanation of hysteresis is the irreversible motion of magnetic domain walls. When an external magnetic field is applied, energy minimization requires domains oriented along the field to grow, with smooth motion of the domain walls. Though ideally this motion is reversible, in real materials it is not since the motion of domain walls is always hindered by imperfections, and if the applied field is strong enough to force the domain walls to pass them, the resulting abrupt relaxation of stress will make the process irreversible.

However, Stoner and Wohlfarth argued that in those materials in which particle-like single-domain magnetic units are separately embedded in a non-magnetic matrix, the above process cannot proceed since there are no domain walls, and in this case another mechanism is dominant, that is, the direct rotation of each particle's magnetization. And in this mechanism the irreversibility is from the large magnetic anisotropy energy of those particles. Now we can see that this is just the situation of magnetic nanoparticles like FePt.

Basically the model Stoner and Wohlfarth studied is a uniformly magnetized single domain particle. With this assumption, classical macroscopic magnetostatics can be

used without any problems. Additionally, they require the particle to have spheroidal symmetry and uniaxial anisotropy along the rotational-symmetry axis. Generally, the Gibbs free energy of a classical magnetic system in an external magnetic field can be expressed as [10]:

$$\begin{aligned} G(\mathbf{M}, \mathbf{H}_a) &= F_{ex} + F_{an} + F_m + G_a \\ &= F_{ex} + F_{an} + \int_\Omega [-\frac{1}{2}\mu_0 \mathbf{M}\cdot\mathbf{H}_m - \mu_0 \mathbf{M}\cdot\mathbf{H}_a] dV \end{aligned} \quad (1)$$

where the four terms respectively stand for the exchange energy (between spins composing the system), anisotropy energy, magnetostatic energy without external field and Zeeman energy. Now the exchange energy can be neglected since the particle is always uniformly magnetized. Then, assuming the easy axis of the particle is the z-axis, in the simplest quadratic form, the anisotropy energy can be expressed as:

$$F_{an}(\mathbf{m}) = K_1(1 - m_z^2)V_0 \quad (2)$$

where $m_z = \frac{M_z}{M}$, V_0 is the volume of the particle and K_1 is a constant. As the magnetic easy axis is along the geometrical principal axis of the spheroid, the magnetostatic energy term is simplified to be

$$\begin{aligned} F_m(\mathbf{m}) &= \frac{1}{2}K_2(N_x m_x^2 + N_y m_y^2 + N_z m_z^2) \\ &= \frac{1}{2}K_2 N_\perp (1 - m_z^2) + \frac{1}{2}K_2 N_z m_z^2, \end{aligned} \quad (3)$$

where N_x, N_y and N_z are the diagonal elements of the diagonalized demagnetization tensor, and K_2 is another constant.

Then, we can neglect constant terms in G, since to get the equilibrium property we only require its derivative to be zero. Thus in a compact dimensionless form, the Gibbs free energy is written as

$$g(\mathbf{m}, \mathbf{h}_a) = -\frac{1}{2}k_{eff} m_z^2 - \mathbf{m}\cdot\mathbf{h}_a. \quad (4)$$

The behavior of g is depicted in Fig. 4, in which θ, θ_h are respectively the spherical angles between $\mathbf{m}$, $\mathbf{h}_a$ and the z-axis. Fig. 5 shows the hysteresis curve Stoner and Wohlfarth got from their model, which is not hard to imagine with the help of Fig. 4. Correspondingly, Fig. 6 shows hysteresis curves of FePt nanoparticles from experiments.

Fig. 4 also shows that the free energy of the system may have either one or two global minima, depending on the value of $\mathbf{h}_a$. The separating curve, which represents the values of critical field $\mathbf{h}_{SW}$, between the region where two minima exist and the region where there is only one is the so-called Stoner-Wohlfarth astroid (Fig. 7). In the particular case of $\theta_h = 0$, it is easy to verify that $h_{SW} = k_{eff}$.

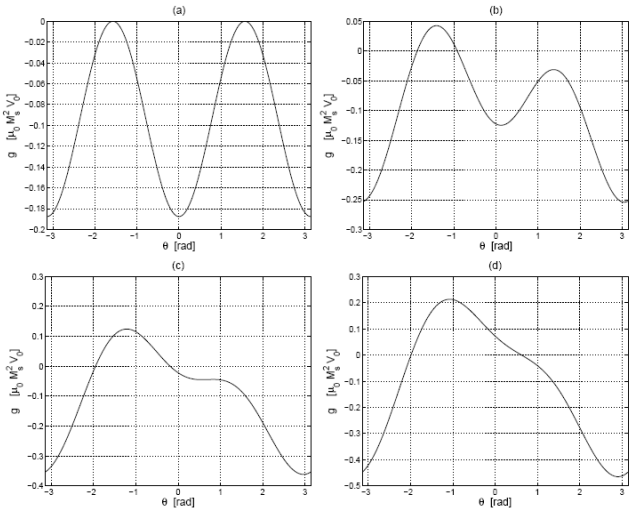

FIG. 4: Free energy as a function of angle θ, $k_{eff} > 0$. (a) for $\mathbf{h}_a = 0$ two minima $\theta = 0$, π and two maxima $\theta = \pm\pi/2$ exist. (b) for small $\mathbf{h}_a$ with given $\theta_h \neq 0$ there still exist two minima and two maxima. (c) a critical value $h_{SW}(\theta_h)$ of h_a exists such that a saddle point appears in place of one minimum and one maximum. (d) for $h_a > h_{SW}(\theta_h)$ only one minimum and one maximum remain.

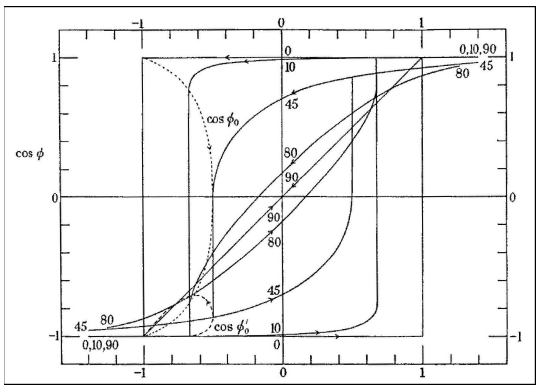

FIG. 5: Hysteresis loops in Stoner's original paper. The external field in abscissa is measured in units of k_{eff}. Magnetization in ordinate is measured in units of M_s.

B. Landau-Lifshitz-Gilbert Theory

As mentioned in the introduction, one major possible application of magnetic nanoparticles is in magnetic storage of information, where one must study the dynamics of a single magnetic cell. Most of the theoretical analysis is based on the classical model proposed by Landau and Lifshitz in 1935 [4], and later modified by Gilbert [5] in 1955. The basic idea of this theory is very simple. Let's begin with the relation between the magnetic moment $\mathbf{M}$ and the angular momentum $\mathbf{L}$ in classical electromagnetic theory:

$$\mathbf{M} = -\gamma \mathbf{L}, \quad (5)$$

where $\gamma = \frac{g|q|}{2mc}$ is the gyromagnetic ratio and g is the Landé factor. Then classical angular momentum theorem

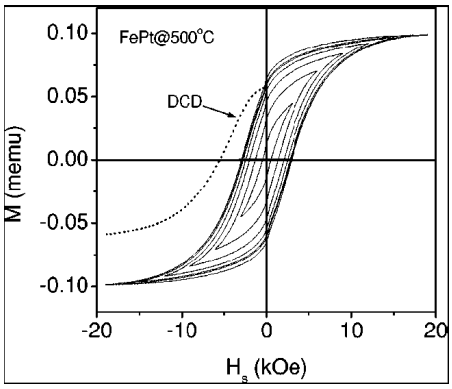

FIG. 6: Hysteresis loops of self assembled FePt nanoparticles annealed at 500°C for 30 min.(Figure from [11])

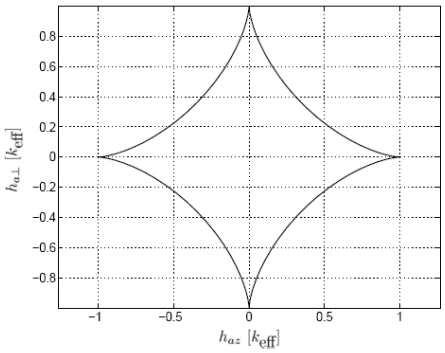

FIG. 7: The Stoner-Wohlfarth astroid in the h_{az}, $h_{a\perp}$ plane, which can be found by searching the saddle points of free energy on the plane.

gives:

$$\frac{d\mathbf{L}}{dt} = \mathbf{M} \times \mathbf{H} \tag{6}$$

where $\mathbf{H}$ is the external magnetic field. Substituting Eq. (5) into this equation we get the dynamical equation of the magnetization $\mathbf{M}$:

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}. \tag{7}$$

Assuming the field $\mathbf{H}$ to be time independent, multiplying Eq. (7) by $\mathbf{M}$ and $\mathbf{H}$ respectively gives

$$\frac{d}{dt}[\mathbf{M}(t)]^2 = 0, \frac{d}{dt}[\mathbf{M}(t) \cdot \mathbf{H}] = 0. \tag{8}$$

These two equations tell us that the magnitude of $\mathbf{M}$ and the angle between $\mathbf{M}$ and $\mathbf{H}$ are unchanged during the motion, which means Eq. (7) represents a precessional motion.

Until now our system has been conserved. The idea of Landau and Lifshitz is to introduce damping, hence the interaction with the environment in this system. In analogy with damping motion in Newton dynamics, one can introduce a damping term proportional to the time derivative of $\mathbf{M}$ in Eq. (7):

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times (\mathbf{H} - \frac{\lambda}{\gamma M_s}\frac{d\mathbf{M}}{dt}), \tag{9}$$

where λ is the damping factor and $M_s = |\mathbf{M}|$ is the saturation magnetization. Inserting Eq. (7) into Eq. (9) gives the Landau-Lifshitz-Gilbert equation [12]:

$$\begin{aligned}(1+\lambda^2)\frac{d\mathbf{M}(t)}{dt} = & -\gamma[\mathbf{M}(t) \times \mathbf{H}(t)] \\ & - \frac{\lambda\gamma}{M_s}\mathbf{M}(t) \times [\mathbf{M}(t) \times \mathbf{H}(t)].\end{aligned} \tag{10}$$

When written in the dimensionless variables $\tau = \gamma M_s t$, $\mathbf{m} = \mathbf{M}/M_s$, $\mathbf{h} = \mathbf{H}/M_s$, the above LLG equation becomes

$$\begin{aligned}(1+\lambda^2)\frac{d\mathbf{m}(t)}{d\tau} = & -[\mathbf{m}(t) \times \mathbf{h}(t)] \\ & - \lambda\mathbf{m}(t) \times [\mathbf{m}(t) \times \mathbf{h}(t)].\end{aligned} \tag{11}$$

Now we have all the necessary tools to study the dynamics of magnetic nanoparticles. To incorporate the Stoner-Wohlfarth model into LLG theory, one only needs to replace $\mathbf{h}$ in Eq. (11) by

$$\mathbf{h}_{eff} = -\frac{\partial g}{\partial \mathbf{m}}, \tag{12}$$

where g is given by Eq. (4). The general dynamical behavior of Stoner particles is that, from an initial non-equilibrium configuration, it will evolve through precession and damping to a final configuration with globally minimal free energy. Generally people start from Eq. (11) and Eq. (4), try different external fields and anisotropy terms and solve the LLG equation numerically or, in very limited cases, analytically to look for optimal speed of magnetization reversal [14][15]. However, it is not always safe to use this approach because one cannot know when the quantum effects will be too strong for classical equations to hold. We will discuss this in more detail in Sec. IV.

III. QUANTUM MACROSPIN MODEL OF MOLECULAR MAGNETS

One motivation of a microscopic theory for molecular magnets is to explain the uncommon steps in their magnetization hysteresis curves (Fig. 3). Now people generally believe that the origin of such steps is quantum resonant tunneling. Before going into details, one can get the simple idea of this mechanism with the help of Fig. 8, which is just a quantum version of Fig. 4.

In classical case the system must first climb up the potential barrier in order to go to the global minimum

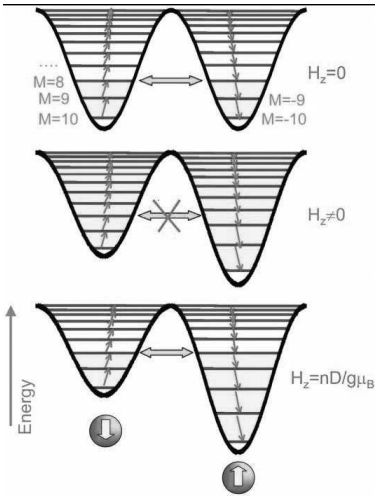

FIG. 8: Energy levels for an $S = 10$ spin under different applied fields. (Figure form [16])

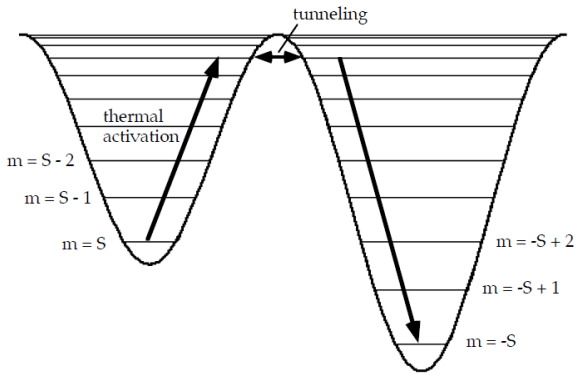

FIG. 9: Double-well potential of a uniaxial spin. The arrows schematically illustrate the thermally assisted resonant tunneling process. (Figure form [19])

in another well, which result in the common hysteresis curves as in Fig. 5. However in quantum case, it is well-known that when any two energy levels separated by the potential barrier "match", the transition probability will be much higher than that when they are different. This phenomenon is the so-called quantum resonant tunneling. With the help of this mechanism, at certain values of the external field the system will directly tunnel through the barrier, resulting in abrupt change of magnetization, hence the steps in the hysteresis curves.

At this stage one may wonder why there had been no evidences for this simple model until very recently. Actually until the discovery of Mn_{12}, there had been no good materials for magnetization tunneling experiments. Unlike most ensembles of magnetic clusters, a macroscopic sample of a molecular magnet has unique, chemically determined properties. Another important feature of these systems is that although each Mn_{12} cluster's spin ($S = 10$) is large for a single molecule, it is small relative to most superparamagnetic systems. This small spin value together with the its large magnetocrystalline anisotropy yields an appreciable energy separation between spin levels, which makes the observation of tunneling much easier.

Now it is easy to write down a desired Hamiltonian for this system:

$$H = -DS_z^2 - g\mu_B S_z H_z + H', \tag{13}$$

where, in close analogy with Eq. (4), the first two terms are separately anisotropy energy and Zeeman energy, and H' contains all terms not commuting with S_z (otherwise there would be no tunneling).

We call the model of molecular magnets using a Hamiltonian like Eq. (13) a semi-microscopic model, because the inner structure of the molecule has been neglected. On one hand, the exchange coupling between magnetic ions inside are usually strong enough to rationalize our simplification. On the other hand, it is not practical to start from the atomic level– remember that even the simplest hydrogen molecule cannot be solved analytically. It is also worthwhile to mention that the quantum computing scheme proposed in [9] is also based on this macrospin model.

From Eq. (13) one can easily deduce the values of the external field when the energy levels on either side of the energy barrier coincide. The result reads:

$$H = -Dn/g\mu_B. \tag{14}$$

One can verify this result by comparing it with the inset of Fig. 3. It is satisfying that for reasonable values of D, this is indeed where steps occur. Strictly speaking, this is the case only at zero temperature. When at finite temperatures, partly because levels near the top of the barrier tunnel more easily [16], the semiclassical "thermally assisted resonant tunneling" picture, which is illustrated in Fig. 9, has been verified by experiments [19].

Until now everything has worked perfectly. However to go one step further one encounters the difficulty of discerning the H' term in Eq. (13), i.e., what causes tunneling? From now on we have to discriminate between Fe_8 and Mn_{12}. For Fe_8, H' is easily recognized as the transverse anisotropy energy which is well characterized experimentally [17]:

$$H' = E(S_x^2 - S_y^2), \tag{15}$$

where E is a constant. Wernsdorfer et al. [18] exactly solved the Hamiltonian Eq. (13) with Eq. (15) and gave the energy spectrum with the variation of applied field, which is depicted in Fig. 10.

At each level crossing, there is a gap opened up by the transverse anisotropy term, and to jump across the gap means tunneling not happening, and vice versa. The probability to avoid crossing the gap is given by the

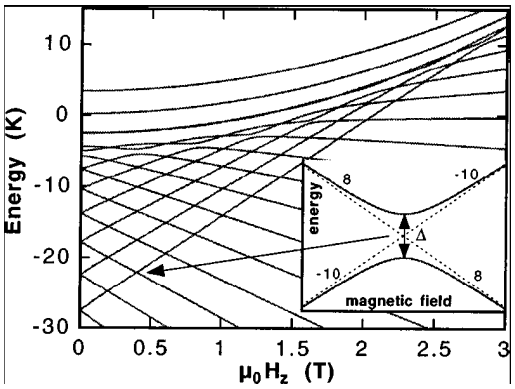

FIG. 10: Energy levels of Fe_8 as a function of applied field. The inset shows the detail at a level crossing where there is a gap opened by the transverse term.(Figure form [18])

Landau-Zener formula

$$P = 1 - exp(-\frac{\pi\Delta^2}{\beta}), \tag{16}$$

where $\beta = dH/dt$ is the rate at which the external field is swept, and Δ is the size of the gap. From this equation it is easy to see when one sweeps the field very slowly, tunneling is bound to happen.

Now let's turn to Mn_{12}, which is a more subtle case since there is no well recognized S_z symmetry breaking mechanism. One major difference between Mn_{12} and Fe_8 is that the former has tetragonal symmetry, thus any anisotropy terms must be at least of order S^4. There have been several candidate mechanisms proposed, such as transverse external field, spin-phonon interaction, dipolar interactions between neighboring molecules, hyperfine interaction with the Mn and other nuclei in the system, etc. [19]. However, the experiment data up to now cannot justify which mechanism truly exists and/or dominates.

IV. BRIDGE BETWEEN QUANTUM AND CLASSICAL

In this final section we will discuss some insufficiencies of the theories introduced in previous sections; then based on that we will propose some rough ideas for a desired mesocopic theory for nanoscale magnets, which may bridge the gap between quantum and classical descriptions.

As we mentioned in Sec. II, one main shortcoming of the LLG theory of Stoner particles is that one cannot know when the quantum influence will become too strong to be excluded, especially at mesoscopic scale. This insufficiency, to a considerable extent, diminishes the predictive power of LLG theory in this case [15]. On the other hand, the treatment of dissipation in LLG theory is somewhat empirical and phenomenological. If the microscopic nature of dissipation cannot be properly understood, how to take it into account when quantum effect are important will be a nontrivial problem.

A similar situation arises in the macrospin theory of molecular magnets in Sec. III, where the "thermally assisted resonant tunneling" at finite temperatures is still a semiclassical picture, which treats the system as a classical thermodynamic object when no tunneling happens. Moreover, dissipation is almost totally excluded in this theory, which makes macrospin theory impossible to be consistent with the essentials of LLG theory.

From the foregoing discussions we can arrive at the idea that, to construct a mesocopic theory for nanoscale magnets, the first step may be to include environment, hence dissipation, at a quantum level. In the pioneer work by Caldeira and Leggett [20] on macroscopic tunneling, dissipation is introduced as a perturbation, so that tunneling will not be totally destroyed. In their work environment is simplified to a set of independent harmonic oscillators linearly coupled to the system. Actually their treatment is in the framework of a much more mature theory today, i.e., quantum dissipation theory or dissipative quantum mechanics [21]. One can see that this framework is especially suitable for the current problem. And actually we heard that C. Hicke has already derived LLG equation in a bottom-up way using quantum dissipation theory [22].

In conclusion, we reviewed experiments and theories in the area of nanoscale magnets, which includes two categories: Stoner nanoparticles such as FePt, which are treated as classical objects using LLG theory, and molecular magnets such as Mn_{12}, which are described by quantum macrospin theory. From application point of view, in both cases the theories currently used, although very successful, suffer from some nontrivial insufficiencies. To remedy these shortcomings as well as to make a consistent bridging between classical and quantum approaches, we propose to employ quantum dissipation theory to include the influence of environment microscopically.

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