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Transverse magnetization and transient oscillations in the quantum tunneling of molecular magnets

Maxime Clusel and Timothy Ziman

1 Institut Laue-Langevin, 6 rue Jules Horowitz, B.P. 156X, F-38042 Grenoble Cedex, France,
2 LPM2C-CNRS, 25 avenue des Martyrs, B.P. 166, F-38042 Grenoble Cedex, France.

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To Yukiko, who never questioned the beauty of nanomagnetism

Abstract. We calculate the response of a molecular magnet subject to a time-varying magnetic field and coupled to a heat bath. We propose that observations of calculated oscillations transverse to the field direction may be an effective way of demonstrating quantum tunneling and thus probing the details of level repulsion. The effective model of a triangle of Heisenberg spins and weak anisotropies, as has been used to model the molecular magnets \{V_{15}\} and \{Cu_3\}, is used to illustrate this.

PACS. 75.50.Xx Molecular magnets – 75.60.Ej Magnetization curves, hysteresis, Barkhausen and related effects – 74.25.Nf Response to electromagnetic fields (nuclear magnetic resonance, surface impedance, etc.)

1 Introduction

Studies of the quantum behavior of open systems are almost as old as quantum mechanics itself. The related phenomena of relaxation and decoherence have passed from conceptual to practical questions with the improvements of experimental technique during the last decades. One can now build and manipulate physical objects at the borderline between quantum and classical worlds. In the future, materials structured on such a scale may be of use in applications such as very tiny components of magnetic memory and quantum computers. This would require detailed understanding of switching and dissipation between different quantum states. For such memories we can distinguish “dynamical” aspects, related to writing (or reading) of different states, and “relaxational” aspects, concerning the storage itself. If such memories were to be part of future quantum computers, the phase of the wavefunctions is essential and the loss of phase central to the questions of the feasibility of building such a device. It is therefore crucial to understand and control the coupling of the small magnetic object to its environment.

There is a large literature on the subject of two level systems in contact with an environment but in the magnetic systems we shall consider, there are aspects not present in, for example, “qubits” based on Josephson couplings. In particular there is the complexity of the magnetic spectrum: as we shall see, it is essential to take into account more than two levels to understand the dynamics. Furthermore the nature of the environment is unclear and new experiments should be designed that may shed light on the issue. This paper will address a multi-level problem, calculating the response of a relatively simple magnetic system to a fast-varying magnetic field, including dissipation to a simple model environment. In the context of quantum computing this corresponds to the “writing” aspect on a potential quantum memory storage device. This will be illustrated using the example of a triangle of magnetic ions, as a model of low spin molecular magnets such as \{V_{15}\} or \{Cu_3\}. We choose the triangular geometry because of recent experiments, to be noted later, and because the spectrum is sufficiently rich that it displays at least some of the features not seen in simple two-level systems.

Experimentally, the development of new magnetic coils allowing for the generation of high magnetic fields varying on very short time scales has led to experiments that can probe such interesting aspects of statistical mechanics. We shall argue that non-equilibrium measurements with pulsed magnetic fields can be more informative, if properly analysed, than equilibrium measurements for fine details of interactions between the spins. Such experiments usually involve solids, where the molecular units repeat, and are assumed to have sufficiently weak interactions that they may be considered as independent, to a first approximation. They are therefore analysed in terms of a static Hamiltonian for the individual molecule, with an...
additional time-dependent external field. The dissipation is induced by an environment, which represents contributions from coupling to phonons, dipole interactions with other molecules, hyperfine coupling to nuclear spins.

2 Molecular magnets and Landau-Zener regime

If the interactions between spins were perfectly isotropic, transitions in magnetization would occur by the crossing of eigenvalues of different spins. In real magnets anisotropies mix the levels and, on some scale, crossings are avoided. The dynamics will therefore depend strongly on the fine structure of the molecular levels of the spins. If the level repulsion is weak, as is the case if the anisotropies are small, it may be possible to enter a regime where the temperature is sufficiently low, and the coupling to the environment sufficiently strong, that we can be in a regime of pure quantum tunneling. It is for this reason that there has been great interest in the Landau-Zener tunneling régime [3].

If we consider only two levels with an avoided crossing, the transitions between the two levels, when varying parameters such as the external magnetic field, is the transitions between the two levels, when varying a tunnelling régime [2].

The advantage of these compounds compared to those of (i) is that experiments can be conducted at temperatures comparable to the splittings, thus a different régime of non-equilibrium phenomena can be studied. Furthermore Choi et al. have suggested that the extra degeneracy of different chiral states of the triangular structures may be useful for applications [6].

A problem in the interpretation of past experiments is that it is not necessarily clear whether one is really in the tunneling régime. One approach has to be to look for the characteristic dependence on the sweeping velocity of the magnetic field and compare to the predictions of Landau-Zener theory. The difficulty is that features found such as the magnetic Föehn effect [10], where extra plateaux in the magnetization occur in the non-equilibrium response, may be described either as phonon bottleneck effects [7], [11], i.e. that the bath does not relax to equilibrium on the timescale of the experiment, or by a dynamical effect of the system coupled to a bath that stays in equilibrium. In what follows we shall look for extra features which could distinguish effects intrinsic to the small quantum system from such bottleneck effects.

3 Model for the molecular magnets \{V_{15}\} or \{Cu_{3}\}

In order to describe the reaction of the magnet to the external magnetic field, we have to precise the internal Hamiltonian for the molecular magnets. For systems (ii), to which the rest of the paper is devoted, we can write the Hamiltonian for three spin $\frac{1}{2}$ moments

$$\mathcal{H}_0(t) = \sum_{i=1}^{3} (J_i S_i \cdot S_{i+1} + D_i \cdot (S_i \times S_{i+1}) + H(t) \cdot S_i).$$

The system has $2^3 = 8$ energy levels. The exchanges are chosen for triangular symmetry, i.e. the isotropic exchanges are equal and the three vectors defining DM interactions are determined by a single vector \(D\) and rotated around the three-fold symmetry axis [8]. Before discussing the dynamics, we shall discuss the static properties of $\mathcal{H}_0$. In Figure 2 we show its spectrum , i.e. with \(H(t) = H_z e_z\) constant for different values of \(H_z\). At large fields the spin $\frac{1}{2}$ components have lowest energies and cross with the two degenerate spin $\frac{3}{2}$ levels of opposite chirality. To make the degeneracies apparent a small chiral-breaking term has been added. We choose this additional term to be a Sen-Chitra interaction $\mathcal{H}_{\text{chiral}} = A_{\text{chiral}} (H \cdot e_z) S_1 \cdot S_2 \cdot S_3$. 

of equilateral triangles. In \{V_{15}\} these may be thought of as the three spin $1/2$ V$^{4+}$ ions sandwiched between two planes of six ions. As de Raedt et al have shown [9], the extra Vanadium ions renormalize the effective interactions in the plane, but for the fields of interest the out-of-plane spins can otherwise be considered as being locked in inert singlet states. The gaps are generated by Dzyaloshinsky-Moriya (DM) interactions and are typically $10^{-2}$ K so that the velocity needed for Landau-Zener tunneling would be $10^2 \text{T.s}^{-1}$, within range of recent “single-turn” magnets [15].
In current experiments and probably future applications, the magnet cannot be considered as an isolated system. It is indeed surrounded by an environment, responsible for relaxation and decoherence phenomena. In order to take into account those effects in our calculation, we have to define an environment explicitly. Unfortunately the true environment is not yet known precisely. We therefore focus here on a simple environment [17] modelled as an infinite environment is assumed to be Markovian, i.e. its correlation time is much shorter than the time scale of the dynamics. Taking these two hypotheses together implies a static thermostat at a given temperature $T$. Furthermore the environment is assumed to be Markovian, i.e. its correlation time is much shorter than the time scale of the dynamics. Taking these two hypotheses together implies a static environment, described at all times by the density matrix $\rho_{\text{env}}(t) \propto \exp(-\beta H_{\text{env}})$.

The coupling of the spin system to the bath is included with the term

$$H_{\text{coupling}} = g \int_{-\infty}^{+\infty} d\omega \, I(\omega)(b_d^+ b_\omega + b_\omega^+ b_d) X$$

The operator $X$ is some combination of spin operators $S_i$ and couples the spin system linearly to the boson operators of the bath, $b_d, b_d^+$, with dimensionless strength $g$. It has sufficiently low symmetry that it can restore equilibrium from any initial state. e.g. $X = \sum_{i=x,y,z} a_i S_i^x + b_i S_i^y + c_i S_i^z$). The coefficients $a_i, b_i, c_i$ differ from spin to spin, in order to induce transitions between states of different chiralities.

4 Environment: model and coupling to the magnet

In current experiments and probably future applications, the magnet cannot be considered as an isolated system. It is indeed surrounded by an environment, responsible for relaxation and decoherence phenomena. In order to take into account those effects in our calculation, we have to define an environment explicitly. Unfortunately the true environment is not yet known precisely. We therefore focus here on a simple environment [17] modelled as an infinite set of bosonic harmonic oscillators:

$$H_{\text{env}} = \int_{-\infty}^{+\infty} d\omega \, \omega I(\omega) \left( b_d^+ b_\omega + \frac{1}{2} \right),$$

We choose the spectral density $I(\omega)$ to be a power law,

$$I(\omega) = \omega^\alpha \Theta(\omega),$$

with $\alpha = 2$ (Super-Ohmic regime, as appropriate for a three dimensional phonon bath), and where $\Theta$ is the Heaviside distribution. Several additional hypotheses will be needed in the following. First we will supposed that the initial state of environment is a thermal state, that is to say that the environment is in thermal equilibrium with a thermostat at a given temperature $T$. Furthermore the environment is assumed to be Markovian, i.e. its correlation time is much shorter than the time scale of the dynamics. Taking these two hypotheses together implies a static environment, described at all times by the density matrix $\rho_{\text{env}}(t) \propto \exp(-\beta H_{\text{env}})$ [17].

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In contrast to the relatively featureless equilibrium magnetization it will be seen that non-equilibrium observables such as magnetization are much richer and we now turn to the question of dynamics. We remark also that in such triangular-based spin systems, switching between different states involves more than just two states at a time; because of the chirality the crossings involve three (between spin $\frac{z}{2}$ and the two chiral $\frac{1}{2}$ states) or four (between the degenerate chiral doublet at $H = 0$). At the least a generalization of the Landau-Zener form is required. We remark that in certain cases the asymptotic transition probabilities for such multi-crossing may be reduced, via an “Independent Crossing Approximation” [15] to the calculation of two-level crossings. Because of the importance of avoided crossings we need a general theory that applies for multiple crossings with the details of the anisotropy [16]. For direct comparison with experiment it is also useful to have a general form of the time-dependent external field generated by the coils.

Fig. 1. (Color online) Spectrum of Hamiltonian $\mathcal{H}_0$ and equilibrium magnetization.
5 Bloch-Redfield approach to the reduced density matrix dynamics

The system (magnet + environment) is a closed quantum system described by the Hamiltonian

$$\mathcal{H}_{\text{tot}} = \mathcal{H}_0(t) + \mathcal{H}_{\text{env}} + \mathcal{H}_{\text{coupling}}.$$  \hspace{1cm} (5)

All the physical information of this closed system can be determined by the density matrix \(\rho_{\text{tot}}(t)\). As the experimental set-up allows measurements on the magnet only, and not on the environment, we are interested in this magnet as an open quantum system, described by the reduced density matrix \(\rho(t) = \text{Tr}_{\text{env}}[\rho_{\text{tot}}(t)]\). A naïve approach to study the magnetic dynamics would be to integrate the Liouville equation, coming from first principles, to obtain the total density matrix \(\rho_{\text{tot}}(t)\) at all time, and then to perform the trace on the environment degrees of freedom. However such an approach is essentially impossible to follow in practice given the high dimension of the Hilbert space. An approach to this problem, that is by now standard, is to make the additional hypotheses on the environment presented in the previous section, to obtain a dynamical master equation for the reduced density matrix. This perturbative approach dates back to the 1950's \cite{21, 22}. We would like to stress that the equation does not come directly from first principles: it relies on additional assumptions whose validity needs to be tested for each particular system. For systems with a larger number of levels, the equations have been put in a compact form \cite{8, 23} to give the equation for the time dependence (in units where \(\hbar = 1\)) of the density matrix projected onto the Hilbert space \(\rho(t)\) of the molecular system (in our case the three coupled spins):

$$\frac{\partial \rho}{\partial t} = -i[\mathcal{H}_0(t), \rho(t)] - g \left( [X, R \rho(t)] + [X, R \rho(t)]^\dagger \right).$$  \hspace{1cm} (6)

If we define the instantaneous eigenbasis \(\{ |k, t\rangle \} \) of \(\mathcal{H}_0(t)\) by

$$H_0(t)|k, t\rangle = E_k(t)|k, t\rangle,$$  \hspace{1cm} (7)

the operator \(R(t)\) is determined by its matrix elements

$$\langle k, t|R|m, t\rangle = \left[ I(\omega_{km}(t)) - I(-\omega_{km}(t)) \right] \times n_g[\omega_{km}(t)]X_{km}(t),$$  \hspace{1cm} (8)

where \(\omega_{km}(t) = E_k(t) - E_m(t)\) and

$$n_g(\omega) = (e^{\beta \omega} - 1)^{-1}. \hspace{1cm} (9)$$

The external field \(\mathbf{H}\) is taken to have arbitrary time variation in a single direction \(e_x, e_y\) or \(e_z\).

We shall now present results based on integration of the quantum master equation \cite{8}. In practice we integrate by fourth order Runge-Kutta approximation which leads to numerically convergent results, at least for weak coupling \(g\) and for times long enough to define several hysteresis loops. The hamiltonian must be diagonalised for each time step in order to define the matrix \(R(t)\). The time variation of the external field can be arbitrary but for our purposes we shall take it as cosinusoidal:

$$\mathbf{H}(t) = H_0 \cos(\Omega t) \mathbf{e}_i, \ i = x, y, z,$$  \hspace{1cm} (10)

and as initial conditions \(\rho(t = 0) = \exp(-\beta \mathcal{H}_0(t = 0))\), i.e. the spin system in equilibrium with the heat bath at inverse temperature \(\beta = \frac{1}{k_B T}\).

6 Numerical observables

We shall now make calculations of the observables for a model case in which we take a single component of the DM vector along the \(z\)-direction, \(\mathbf{D} = D_z \mathbf{e}_z\). The purpose here is not to fit a particular experimental system, more to make general statements. We first show the three components of magnetization as a function of time. The initial condition is the equilibrium density matrix for the same temperature and the field \(H(t = 0) = -J\). It is seen that at the relatively weak couplings that can be integrated numerically (\(g = 10^{-3}\) to \(10^{-4}\)) the magnetization “lags” the equilibrium magnetization. After one full oscillation the profile (in time) of the out-of-equilibrium magnetization approximately repeats, converging slowly towards some steady-state form. We remark that the period for the sinusoidal external field is \(10^4\) in units of \(J^{-1}\). Returning to standard units, where \(\hbar\) is not unity, this corresponds to a time of \(5 \times 10^{-9}\) seconds for \(J = 1\) K.

The information in Fig. 2 can be represented in a form often used by experimentalists, i.e. as a hysteresis loop, where the magnetization is plotted against the external field. This we do in Figs. 3(a) and 4(a). The difference between the two hysteresis loops is that in Figure 3(b)
Fig. 3. (Color online) Energy spectrum and dynamics of the magnetization parallel to the applied field, for $H = H_z(t)e_z$ parallel to Dzyaloshinsky-Moriya vector $D = D_z e_z$. The parameters are $D_z = J/20$, $H_0 = J$, $\Omega = 2\pi \times 10^{-3}/\hbar$, $g = 4 \times 10^{-3}$ and $T = 0.2J$. In the right hand panel, the details of the spectrum of the four lowest energy levels around zero field of Fig. 1, showing the crossing of the two nearly degenerate doublets.

Fig. 4. (Color online) Energy spectrum and dynamics of the magnetization parallel to the applied field, for $H = H_x(t)e_x$ perpendicular to Dzyaloshinsky-Moriya vector $D = D_z e_z$. The parameters are identical to those of Figure 3: $D_z = J/20$, $H_0 = J$, $\Omega = 2\pi \times 10^{-3}/\hbar$, $g = 4 \times 10^{-3}$ and $T = 0.2J$. In the right hand panel the avoided level crossing of the doublet states at zero field.

The DM vector is parallel to the external field direction and in the second, Figure 4(a), it is perpendicular to the DM vector. In each case the magnetization is longitudinal, i.e. parallel to the applied field. The differences between the two orientations $m_z(H_z)$ and $m_x(H_x)$ are that the hysteresis loop is “fatter” when the field is perpendicular to the DM vector, but, more strikingly, that there are extra oscillations visible in $m_x(H_x)$ that are absent for $m_z(H_z)$. What is the difference between the two field orientations in terms of the spectrum of excitations? If we look at an enlargement of the spectrum of Figure 1 around zero field, as shown in Figures 3(b) and 4(b) this difference is apparent. For $m_x(H_x)$ there is an avoided level crossing at zero field with a minimum splitting whereas for $m_z(H_z)$ there is a crossing of levels. This avoided level crossing makes the hysteresis loop “thinner” as the lowest level smoothly evolves from one spin state to another. So what is the significance of the oscillations?
Fig. 5. (Color online) Details of the oscillations in the magnetisation transverse to the applied field, perpendicular to the DM vector. Inset: Wave packet on a longer time scale. The parameters are $D_1 = J/20$, $H_0 = J$, $\Omega = 2\pi \times 10^{-3}/\hbar$, $g = 8 \times 10^{-3}$ and $T = 0.5J$. The avoided crossing is centred at $t=250$.

For a two-level system formulated as a (pseudo-) spin intransverse field and time-varying longitudinal field, these transient oscillations correspond to precession around an instantaneous axis which, near the avoided crossing, is perpendicular to the applied field. In other words these are simply Rabi oscillations in the presence of a time-varying gap. These oscillations are not periodic, but the characteristic frequency range is the gap in the avoided level crossing. This mixing of longitudinal and transverse oscillation is well known in “zero-field” NMR [25]. This suggests that the oscillations may be more visible in the transverse components of magnetization, i.e. the components perpendicular to the applied field. In Figure 2 we show the transverse magnetizations, for applied field perpendicular to the DM vector. It is seen that they are small except close to avoided crossings, where there are oscillations rapid on the time scale of the driving field. We remark that they are visible at a temperature (0.5$J$) well above the gaps induced by anisotropies.

This leads to the suggestion for experiment that instead of looking exclusively at the shape of the hysteresis loop, as has been done most in the past; it may be more instructive to look at the Rabi oscillations. They may be most visible in the transverse components, as they are against a flat background. We remark that these oscillating components have been seen clearly in past numerical calculations of the longitudinal magnetization but have attracted relatively little attention. We recall however from the theory of two-level systems that the decay of longitudinal components and transverse oscillations at a fixed magnetic field determines two characteristic time scales which give different information on the spectrum of the bath: in particular the transverse oscillations are sensitive to the zero frequency component of the bath [26]. By analogy, measuring both longitudinal and transverse components one should, in principle at least, determine different properties of the spectrum of the bath.

We remark that for quantitative studies of the oscillations we should be a little more precise about what we mean by “transverse”; ideally we would take a component with vanishing mean. For the simple case we have shown here the ideal component is that which is perpendicular to both the applied field (parallel to $e_x$) and parallel to the DM vectors, i.e. $m_z (H_z)$. The component perpendicular to both the field and the DM vector $m_y (H_z)$, has small but non-zero values depending on the field. For non-collinear DM vectors on the three spins there may be no absolute separation of smooth and oscillating contributions; nevertheless the oscillations will always be stronger perpendicular rather than parallel to the applied field.

We note that these oscillations occur when the rotational symmetry is broken. This occurs if, as is the case in Figure 3, the DM vector and the external field completely break rotational symmetry, so that the phase transverse is determined by the (time-dependent) Hamiltonian [27]. Oscillations also occur, (but are not shown in the Figures), in much smaller amplitudes of oscillations for $m_y (H_z)$; that is even when the Hamiltonian $H_0$ is symmetric around the field direction. This can happen because the coupling to the bath breaks the symmetry in our simulations. Whether this is true in a physical system or not, depends on the symmetries of the environment of the magnetic molecule. To restore symmetry in our formulation, while nonetheless allowing relaxation, we could include a sum of several terms of the form written for $H_{\text{coupling}}$.

7 Conclusions

We have calculated the dynamic behaviour of weakly anisotropic triangular antiferromagnets, as could be seen in the molecular magnets $[\text{V}_{13}]$ or $[\text{Cu}_{3}]$ subject to vary fast varying magnetic fields. Our simulations model in detail effects of tunneling including transient effects in a situation where Landau-Zener expressions are not sufficient to determine dynamics at arbitrary frequency and at finite temperature. This is especially so here since the zero levels are eliminated by degeneracy of other states on the triangle. We have argued that a useful characterization of the tunneling regime, useful would be the (non-periodic) oscillations in the magnetizations and, in particular, in the transverse components close to regions of level anti-crossing. This should yield more detailed information on the anisotropies causing level repulsion in molecular magnets, and the coupling to the bath, e.g. the value of the coupling $g$ and the nature of the bath. The oscillations can persist (at least at weak coupling) up to temperatures well above the scale of the avoided level crossing. To illustrate this we have calculated hysteresis loops for a model in which there is a single component to the DM vector and the field is taken either parallel or perpendicular to this vector. We chose the simple case of a single component DM vector in order to illustrate the phenomenon. For this case there is only a single region.
of avoided crossing at zero field. In real systems such as \{Cu_3\} all three components are present. There are then avoided crossings where, in the isotropic limit, the spin $\frac{3}{2}$ cross the spin $\frac{1}{2}$ levels $H = \pm \frac{3}{4}J$, and oscillations will occur at all crossings.

We conclude that if one is interested in studying the physics at the scale of a single molecular magnet, it might be useful to study not only the traditional hysteresis loops, but also the transient oscillations, visible in the transverse component of the magnetization dynamics, as the first correction to the adiabatic limit.

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