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Dynamical quenching of tunneling in molecular magnets

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ABSTRACT

It is shown that a single molecular magnet placed in a rapidly oscillating magnetic field displays the phenomenon of quenching of tunneling processes. The results open a way to manipulate the quantum states of molecular magnets by means of radiation in the terahertz range. Our analysis separates the time evolution into slow and fast components thereby obtaining an effective theory for the slow dynamics. This effective theory presents quenching of the tunnel effect, in particular, stands out its difference with the so-called coherent destruction of tunneling. We support our prediction with numerical evidence based on an exact solution of Schrödinger's equation.

1. Introduction

Since a few decades molecular magnetic materials have arisen as a new test ground for several phenomena in quantum behavior of finite size magnetic systems [1–5]. Molecular magnets have attracted attention due to their potential in the implementation of several molecular spintronic devices [6–8]. The long spin coherence times displayed turn them into promising candidates in the context of quantum computing, where the molecule spin is used to encode q-bits [10,5,9].

The quantum mechanical degrees of freedom associated with molecular magnets can be manipulated and controlled with great accuracy by the application of external magnetic fields [2–5]. It has been proposed [10] that suitable manipulation of molecular magnets with time dependent magnetic fields can be used to control the population of their quantum states thereby paving the road toward an implementation of a quantum computation scheme known as Grover's algorithm. Within the same framework in this work we propose a way to control the quantum mechanical state of a molecular magnet by means of a rapidly varying magnetic field. We will show that radiation within the range of terahertz frequencies can be used to quench the quantum state of a molecular magnet thereby providing a useful tool for potential applications. There is previous work relating radiation in the terahertz range with magnetic properties of materials, for example terahertz radiation has been used to control the spin waves of antiferromagnets [11,12] and in Ref. [13] hybrid magnetic structures have been used to generate radiation in the terahertz range. Our predictions allow an extension of such control into the subject of molecular magnets. We study the dynamics of the magnetization of a molecular magnet exposed to circularly polarized terahertz radiation. In response to such perturbations the system can display a quenching of the tunneling rate between energetically equivalent states giving rise to a trapping of the quantum mechanical state. This effect is analogous to an interesting effect in classical mechanics the trapping of a classical particle that can be achieved by introducing a rapidly oscillating potential $V(x, t) = V_0(x) + V_1(x, t)$ [14,15]. Under the action of such forces the slow dynamics of the particle is trapped by an effective potential $V_{\text{eff}} = V_0(x) + \overline{F^2}/(2m\omega^2)$ where F is the force associated with the oscillating potential, $F = -\partial_x V_1(x, t)$, ω its frequency and the bar represents an average over an entire cycle of the oscillating force. Based on those ideas it was proposed and demonstrated by Kapitza [15] that a pendulum with a rapidly vibrating point of suspension would be stabilized in the upward position. Once stabilized the pendulum was shown to display small oscillations around its new equilibrium configuration. The main result of this paper is that a similar result holds for the quantum mechanical state associated with the spin of a molecular magnet. In this sense

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we can say that the prediction corresponds to a *Kapitza effect in Hilbert space*. Instead of promoting transitions between states, the high frequency radiation traps the state of the spin in a given configuration keeping it from describing tunnel transitions into other configurations. A quantitative statement of this effect is encoded in the tunneling time (the time that it takes for the system to tunnel from one minima to another) which is seen to diverge in certain circumstances. In response to the oscillatory disturbance the tunnel effect is suppressed and the states are frozen in a given configuration. A similar effect of suppression of tunneling has been reported in the literature concerning spins where the effect is attributed to interference of Berry's phases of different paths associated with tunneling. In this context the effect has been dubbed quenching of the tunnel amplitude [26,3].

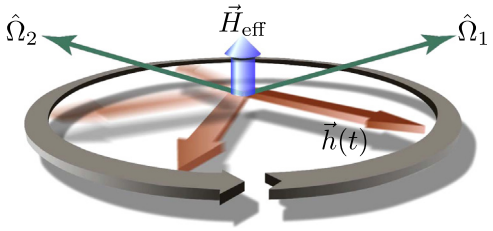

Fig. 1. Schematics of the proposed arrangement. A rapidly rotating magnetic field, $\vec{h}(t)$, in the plane induces a slow dynamics characterized by a stationary effective field, $\vec{H}_{\rm eff}$. The direction of the effective magnetic field is perpendicular to the plane. The slow dynamics is characterized by two degenerate classical ground states $\hat{\Omega}_1$ and $\hat{\Omega}_2$. In general, there are oscillations between those states mediated by quantum tunneling. However, for certain values of the parameters the quantum mechanical oscillations between those two states are quenched.

2. Effective slow dynamics

As is common in the theoretical studies of molecular magnets our study is based on the reduction of the electronic degrees of freedom, to some effective low-energy Hamiltonian, based entirely on localized spin degrees of freedom of the magnetic ions within the molecule [16]. In this context, the total energy has two contributions $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1$, one arising from the intrinsic anisotropy [2], in the form

$$\mathcal{H}_0 = -\mathcal{D}S_x^2+\mathcal{E}(S_z^2-S_y^2). \tag{1}$$

For $\mathcal{D}>\mathcal{E}$ this Hamiltonian represents a quantum spin with easy axis along the x direction and a hard axis along the z direction. The case $\mathcal{E}=0$ has been previously studied in Ref. [17]. This Hamiltonian can be used as a model for describing the magnetic degrees of freedom of several single molecule nanomagnets [2]. Among them the most widely used ones are the Mn_{12}–*ac* molecule [5] (with $S=10$, $\mathcal{D}=0.55$ K and $\mathcal{E}=0.02$ K), the molecular complex Fe_8 molecular magnet [3] (with $S=10$, $\mathcal{D}=0.29$ K and $\mathcal{E}=0.05$ K) and Ni_4 [23] (with $S=4$, $\mathcal{D}=0.75$ K and vanishingly small $\mathcal{E}$). The spin is perturbed by a circularly polarized time dependent external field [12]:

$$\mathcal{H}_1 = -h(\cos\omega t\, S_x+\sin\omega t\, S_y) \tag{2}$$

Here ω is the frequency of the oscillation of the magnetic field in the x, y plane and $h=g\mu_B H$ with H being the amplitude of the oscillating magnetic field and g the gyromagnetic ratio (of order 1 in the examples given). Regarding the geometry we can say that it is dominated by the anisotropy terms in the Hamiltonian. To apply our theory in the experimental setting the incident radiation must be polarized in the plane perpendicular to the hard axis. In a molecular magnet based crystal this can be achieved by selecting the crystal orientation with respect to the incident light. In the remaining parts of this paper we will address the problem of how the spin responds to the perturbation in the limit of large ω.

In the special case of spin 1/2 all the anisotropic contributions reduce to the identity. The resulting behavior has the characteristic form of Rabi oscillations [21]. For higher spin the interaction between the magnetic moment and a radiation field is more complex and has been addressed in several references. For instance in Ref. [22] the effect of photon assisted tunneling events was reported in Fe_8 samples irradiated with circularly polarized light. In Ref. [23] microwave spectroscopy was used to reveal quantum superpositions of high spin states ($S=4$) in Ni_4.

It is possible to derive a general treatment of a quantum system driven by rapidly varying potentials [24]. In general, the goal is to find an effective equation for the dynamics spanned by the Schrödinger equation:

$$i\hbar\partial_t|\Psi\rangle=\mathcal{H}|\Psi\rangle \tag{3}$$

As in the classical case [25], the dynamics of the wave vector can be separated into two components, a fast one that varies in a period of the potential and a slow one that evolves at a slower pace. The method consists in an extension of the method of multiple scales from classical mechanics into quantum mechanics. We separate the slow and fast dynamics, by means of a unitary transformation, and proceed to write an effective theory that involves only the slow variables. The slow dynamics is affected by the rapid motion and is described by an effective Hamiltonian. We start from the time dependent Hamiltonian $\mathcal{H}$ and perform a time dependent unitary transformation $\exp(i\mathcal{F}(t))$. The basic idea is to absorb the time dependency of the Hamiltonian into the operator $\mathcal{F}(t)$ and to obtain an effective time independent Hamiltonian given by

$$\mathcal{H}_{\rm eff}=e^{i\mathcal{F}}\mathcal{H}e^{-i\mathcal{F}}+i\hbar\left(\frac{\partial e^{i\mathcal{F}}}{\partial t}\right)e^{-i\mathcal{F}}. \tag{4}$$

To find the specific representations of $\mathcal{H}_{\rm eff}$ and $\mathcal{F}$ we write them as power series in $1/\omega$:

$$\mathcal{H}_{\rm eff}=\mathcal{H}_{\rm eff}^{(0)}+\frac{1}{\hbar\omega}\mathcal{H}_{\rm eff}^{(1)}+\frac{1}{(\hbar\omega)^2}\mathcal{H}_{\rm eff}^{(2)}+\cdots \tag{5}$$

$$\mathcal{F}=\frac{1}{\hbar\omega}\mathcal{F}^{(1)}+\frac{1}{(\hbar\omega)^2}\mathcal{F}^{(2)}+\cdots \tag{6}$$

Expanding Eq. (4) and equating both sides order by order we found recursive relations between $\mathcal{F}^{(i)}$ and $\mathcal{H}_{\rm eff}^{(i)}$. The expression for $\mathcal{F}^{(i)}$ is found by enforcing that every term in the expansion of the effective Hamiltonian be time independent. A lengthy, but straightforward calculation leads to the following first contributions in the expansion for $\mathcal{F}$:

$$\mathcal{F}^{(1)}=-h\left(\sin\omega t S_x-\cos\omega t S_y\right) \tag{7}$$

and

$$\mathcal{F}^{(2)}=-ih\left(\cos\omega t\,[S_x,\mathcal{H}_0]+\sin\omega t\,[S_y,\mathcal{H}_0]\right), \tag{8}$$

while for the Hamiltonian we obtain

$$\mathcal{H}_{\rm eff}^{(1)}=-\frac{h^2}{2}S_z \tag{9}$$

and

$$\mathcal{H}_{\rm eff}^{(2)}=\frac{h^2}{4}\left([[S_x,\mathcal{H}_0],S_x]+[[S_y,\mathcal{H}_0],S_y]\right) \tag{10}$$

Collecting the contributions up to second order we obtain an effective Hamiltonian: