Quantum Magnetopolaritons

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Abstract

This research project sought to determine the effects of an added degree of mechanical freedom to the tunnel splitting of a single molecule magnet. We consider both mechanical rotation and vibration (ie tortional oscillations) with the later consisting of an analytical and numerical portion which appear thus far to be in good agreement. The first part of this paper will discuss some background information regarding single molecule magnets. Following this will be a description of several theoretical papers proposing effects of coupling between an added degree of mechanical freedom and the macrospin of a single molecule magnet. These served as motivation to consider the effects of this coupling on the tunnel splitting and magnetopolariton formation in these molecules. This will be followed by our research, findings and possible implications of the data found.

I. BACKGROUND

A. Single-Molecule Magnets

Single-molecule magnets (SMMs) have been widely studied due to their ability to display both classical and quantum properties simultaneously. These molecules contain several transition-metal ions, have large spin numbers and possess easy axis anisotropy. They lie between microscopic (quantum) and macroscopic (classical) sized systems in the so called mesoscopic range; which accounts for their ability to bridge the quantum and classical worlds. For example, $Fe_8O_2OH_{12}tacn_6Br_8$ and $Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4$, called Fe_8 and Mn_{12} respectively, can relax by tunneling which is forbidden classically. In addition, this quantum tunneling process can often be observed at the same temperature and on the same time scale as the classical process of thermal relaxation. Another interesting aspect of SMMs is their ability to exhibit properties similar to nanomagnets which have been considered for use as quantum bits, or qubits, in quantum computing. Another possible application for SMMs lies in memory devices. To implement memory systems composed of SMMs, however, would involve the ability to write, store and read the spin information of these molecules. Each of these aspects has been, and currently is, under study [1].

In SMMs, an axial zero-field splitting leads to the division of the spin state, S, of a molecule into (2S+1) levels which correspond to different projections of the molecule's total spin on its easy anisotropy axis. These levels can be described by their spin quantum number m, where -S=m=S. The energy of each level is proportional to the square of its quantum number by a constant D. For SMMs, D is negative yielding an energy barrier between the m=S and m=-S states (See figure 1). This anisotropy barrier results in doubly degenerate ground states at zero field[2]. Also, due to strong spin-spin coupling in these types of molecules, they retain an orientation such that the spin of the entire molecule is S (ie –S or S). A Mn_{12} molecule and its corresponding double well potential are depicted in Figure 1.

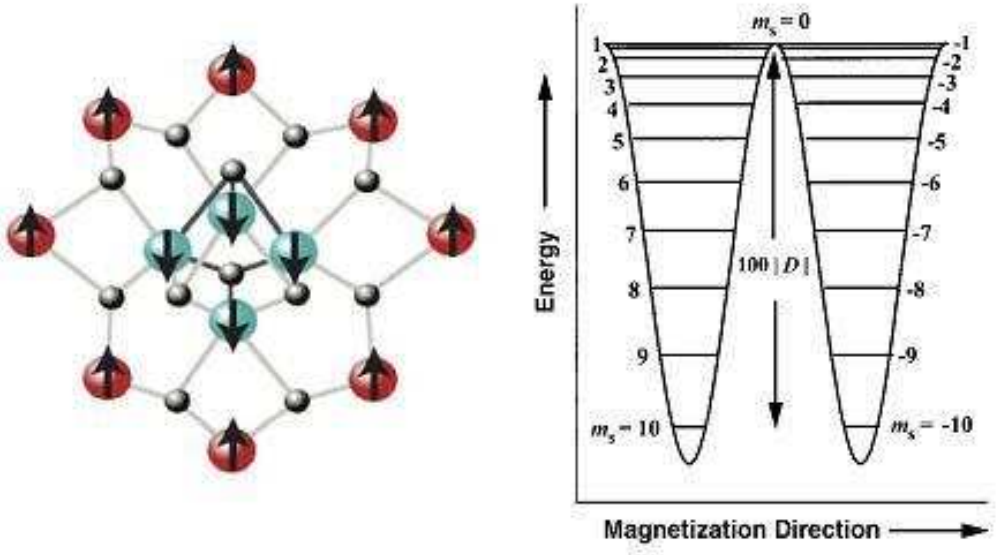

Figure 1: Left**:** Diagram of Mn_{12} (S=10) [2]. Right: Plot of the potential energy versus the magnetization direction for a SMM with an S= 10 ground state experiencing axial zero-field splitting, DS_z , where S_z is the zth component of the electron spin operator. The diagram is for zero external magnetic field[3].

B. Applied Fields and Tunneling

In the mid 1990's there were a number of experiments performed that suggested the possibility of quantum tunneling in SMMs[2]. In 1996, however, the first concrete experimental evidence of such activity was provided by Freidman et al [4]. In this experiment, as they swept a magnetic field along the easy axis, instead of finding the smooth hysteresis curves expected by what was known about thermal relaxation, they instead found plateaus at certain values of the magnetic field. This procedure was repeated at different temperatures. The results are shown in Figure 2. What they found were plots in which the hysteresis loops closed in on themselves at higher temperature indicating that the process of relaxation was assisted by thermal activation. In addition, however, they realized that the relaxation could not be entirely a thermal process due to the fact that the location of the steps, or plateaus, was independent of the temperature. The cause Friedman et al proposed was that tunneling is allowed to occur when energy levels on opposite sides of the well come into resonance. This tunneling occurs only for certain values of the magnetic field and is assisted thermally due to

the fact that, as molecules are excited, the effective barrier height is decreased. The splitting between the corresponding levels due to tunneling is referred to as the tunnel splitting. At sufficiently low temperatures the SMM can be described by the effective spin Hamiltonian as follows:

$$H \approx -D(S_z)^2 - g_z \mu_B S_z H_z - A(S_z)^4 + H', \tag{1}$$

where the first term yields the anisotropy barrier, the second is the Zeeman energy, the third is the next highest order anisotropy term and H' includes all the symmetry breaking terms that do not commute with S_z. The energy of the levels, m, as a function of the applied magnetic field are shown in Figure 3 (d and e).

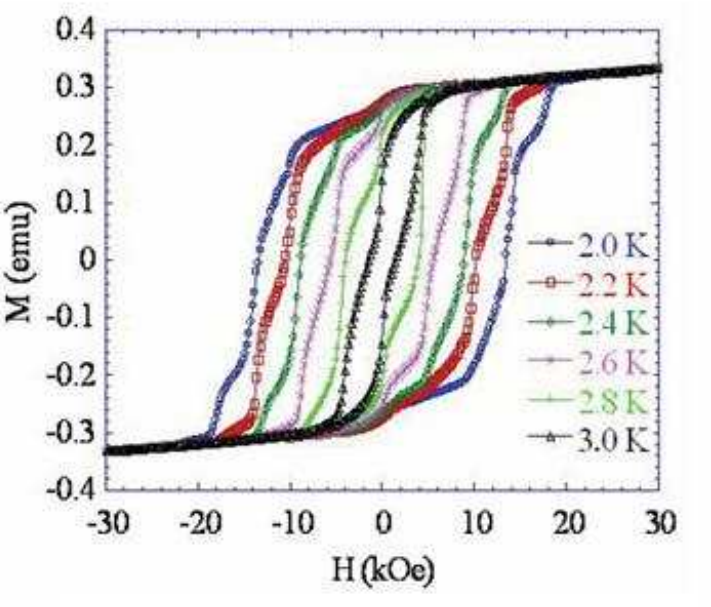

Figure 2: Magnetization as a function of the field strength forMn_{12}[4].

Another remarkable experiment depicting tunneling occurred in 1999 when Wernsdorfer and Sessoli measured the tunnel splitting in Fe_8 as a function of a magnetic field applied at several different angles in the plane perpendicular to the easy axis. Their results are shown in Figure 4. Along the so called medium axis, or $\phi = 90°$, in Figure 4, the tunnel splitting increases monotonically with magnetic field. Along the hard axis, or $\phi = 0°$, however, the tunnel splitting oscillates periodically before exhibiting monotonic growth as is expected. This is due to the destructive interference at periodic intervals of the two

dominant tunneling paths which wind around the hard axis as predicted by Anupam Garg [14].

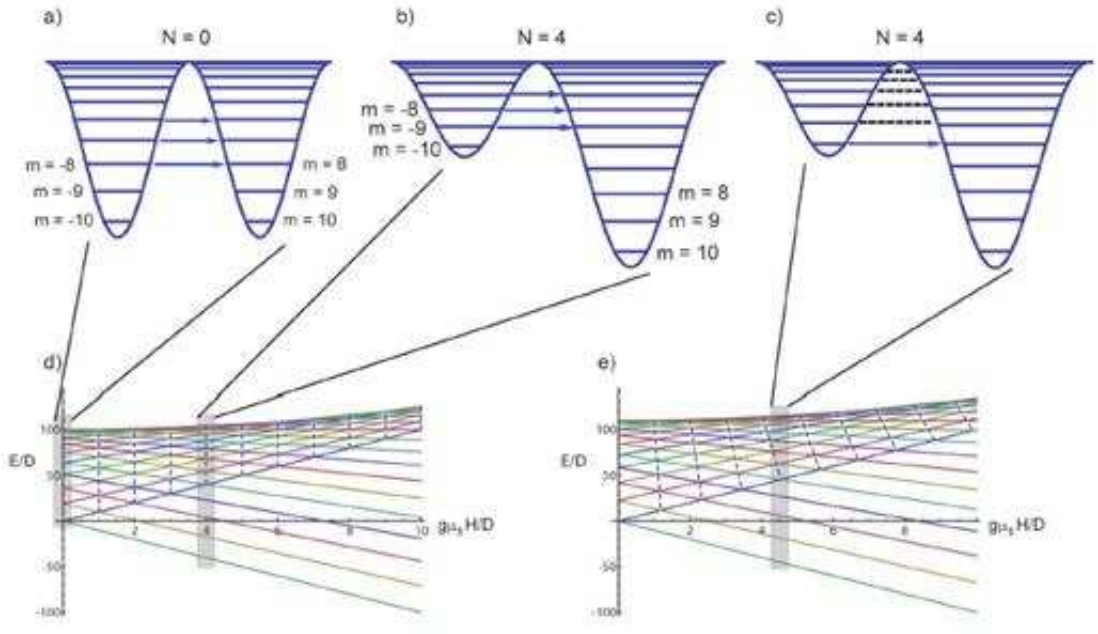

Figure 3: a) Double well potential in the absence of magnetic field. b) Double well potential for the N=4 step in the presence of a magnetic field without the fourth order term of the Hamiltonian (Eq1). c) Double well potential for the N=4 step in the presence of a magnetic field with the fourth order term of the Hamiltonian included. d) Energy crossing for states m of the Hamiltonian without the fourth order term e) Energy crossing for states m of the Hamiltonian with the fourth order term[2].

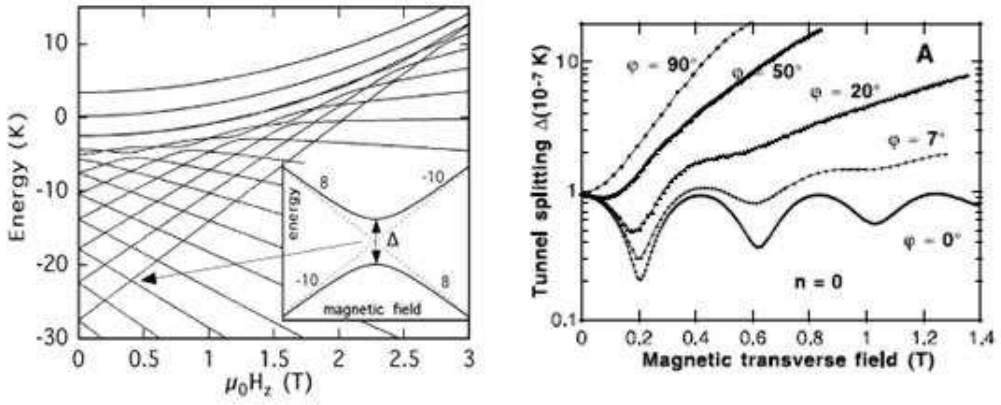

Figure 4: Left: Energy crossing for states m of Fe_8 for a field applied along the easy axis. Δ represents the tunnel splitting. Right: Tunnel splitting as a function of the field in the plane perpendicular to the easy axis. [6]

C. Mechanical Motion

Due to the magnetic properties of single-molecule magnets, rotation of these molecules induces a magnetic field[14]. In this project, we have studied the effect of mechanical rotation and vibration, along with the fields induced thereby, on the tunnel splitting of the molecule. Motivation for this research comes from several places. Recently, there have been a number of papers considering the coupling between mechanical motion and the spin precession in magnetic clusters. One such paper was published by A. A. Kovalev and G. E. W. Bauer in August of 2003 [12]. In this paper, they consider the coupling of the spins in a ferromagnetic film to the mechanical motion of a cantilever to which this film is attached. A figure of the geometry they consider is shown below.

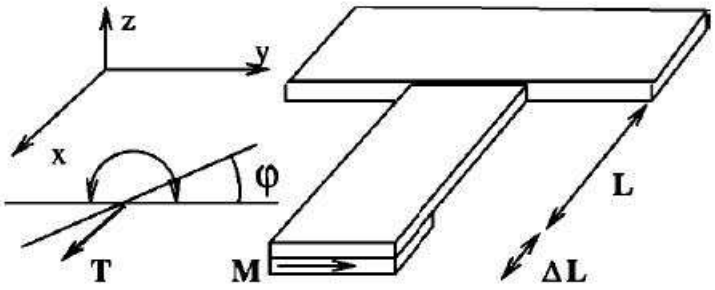

Figure 5: A nanomagneto-mechanical cantilever supporting magnetovibrational modes. On a dielectric substrate (such as Si) a single-domain ferromagnetic film is deposited at the free end[12].

There is constant magnetic field applied along the y direction and an oscillating one alond the x direction. The magnetization $\mathbf{M}$ of the ferromagnet precesses around the effective magnetic field $\mathbf{H}_{eff}$ according to the Laundau-Lifshitz-Gilbert equation (2).

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} + \alpha \mathbf{M} \times \frac{d\mathbf{M}}{dt}, \tag{2}$$

They show theoretically that the ferromagnet effectively absorbs microwaves and turns them into a precessing magnetization, which via the magnetovibrational coupling can be transformed into mechanical motion. In a similar paper in June of 2009, R. Jaafar and E.M. Chudnovsky consider a microcantilever to which a SMM has been attached[7]. By applying a weak ac magnetic field the spin of the molecule is forced to oscillate. These oscillations, then, are required to coincide with mechanical oscillations of the cantilever in order for the total angular momentum to be conserved. Therefore, when the frequency of the applied magnetic field comes into resonance with a resonant mode of the cantilever one would expect the cantilever to oscillate. In this Letter, Jafaar and Chudnovsky propose that it is possible to detect the quantum oscillations of the spin of an SMM through its resonant coupling to the mechanical modes of a microcantilever. They also suggest a possible experiment to demonstrate this effect in which tunneling or force microscopy is used to measure the displacement of the cantilever. These displacements can then be compared with their expected theoretical displacements for the first three modes of the cantilever they describe. Further motivation for our present research stems from a publication in January of 2010, by Jafaar and Chudnovsky in collaboration with D.A. Garanin[1]. In this paper, they consider

the effect of mechanical rotations on a single molecule magnet between conducting leads. The diagram of their set-up and their results are shown in Figure 5.

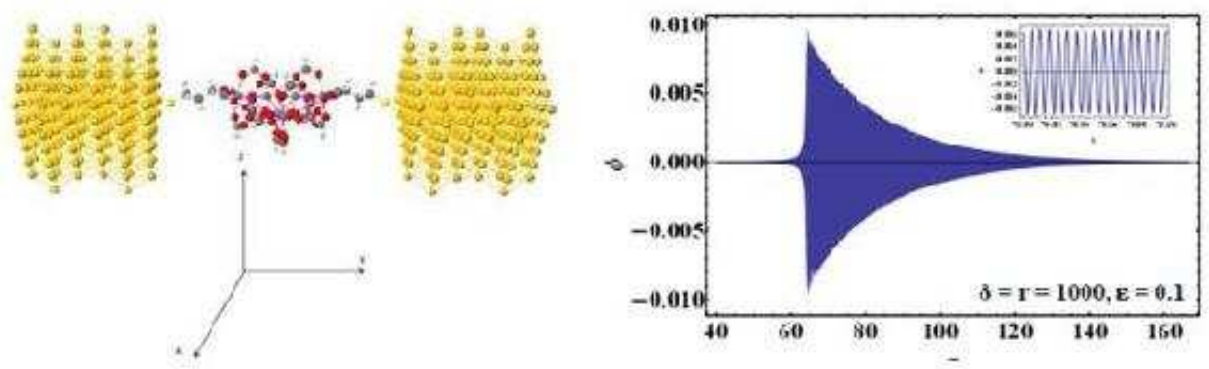

Figure 6: Left: Mn_{12} molecule bridged between two leads. Right: Typical time dependence of the rotation angle ϕ. The inset shows the fine structure of the oscillations [1]

Previously, Chudnovsky and Garanin had demonstrated that the quantum states of a SMM that is free to rotate are different than those of one fixed in a crystal[8]. This motivated them to consider an intermediate situation in which the molecule is bridged between two leads. What they found from their theoretical considerations was that oscillations of ϕ are excited when the time dependent distance between the spin levels coincides with the frequency of the oscillations of the molecule. These demonstrations of magneto-mechanical coupling are what prompted us to consider the possibility of using the mechanical motion of the molecule to affect its tunnel splitting. For small tunnel splitting, the oscillations between spin states is slow, allowing time for decoherence to occur and the spin to localize. This is of interest because, if the effect of rotation can be made large enough in practical setting, such mechanical motion could be used to increase the tunnel splitting, effectively rewriting the spin information of the molecule. This, then, would enable the implementation of SMMs in high-density magnetic storage.

II. RESEARCH METHODS AND FINDINGS

A. Mechanical Rotation

Due to the magnetic properties of single-molecule magnets, rotation of these molecules is equivalent to applying a magnetic field. In this project, we have studied the effect of this mechanical rotation, and the field induced thereby, on the tunnel splitting of the molecule. What we found was that, for a significant change in tunnel splitting to occur, the frequency of rotation must be very large. In fact, a field induced by rotation of just 0.1 Tesla requires frequency of rotation of 3 GHz. In a paper entitled "Rotational Actuators based on Carbon Nanotubes" by A. M. Fennimore et al [9], Fennimore and his colleagues describe the construction and successful operation of a nanoscale electromechanical actuator. This actuator is depicted in Figure 6 and consists primarily of a rotatable metal plate that is attached to a multi-walled carbon nanotube. After either end of the nanotube is attached to a surface, depicted by A1 and A2 in Figure 6, the outer nanotube wall is severed on either side of the rotor which allows the center section to spin freely. Small voltages are then applied at positions S1, S2 and S3 in Figure 6 which capacitively attract the rotor and drive the nanomotor.

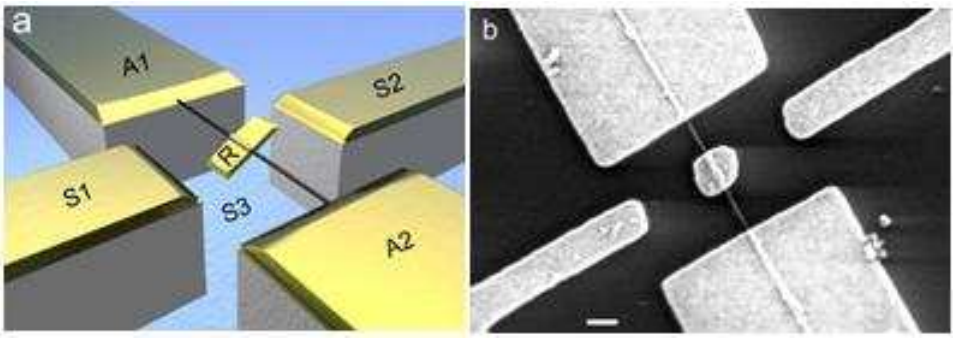

Figure 7: Left: Diagram of the nanoscale electromechanical actuator produced by Fennimore et al. Right: SEM image of the same actuator[9].

According to Fennimore et al, such a nanomotor can be operated in the GHz range. Thus, such a high frequency requirement is not entirely impracticable.

B. Mechanical Vibration

We then considered the effect of mechanical vibration of the molecule. There have been two portions to this research so far. One has been analytical and the other numerical. Our general Hamiltonian for each case is as follows:

$$\hat{H} = (\hat{a}^{\dagger}\hat{a} + \frac{1}{2})\hbar\omega_r + e^{-i\hat{S}_y\hat{\phi}}\hat{H}_A e^{i\hat{S}_y\hat{\phi}} + \gamma\hbar\hat{\mathbf{S}}\mathbf{B}, \tag{3}$$

where $\hat{a}^{\dagger} = \frac{1}{\sqrt{2}}(-\frac{d}{d\phi} + \phi)$ denotes the creation operator and $\hat{a} = \frac{1}{\sqrt{2}}(-\frac{d}{d\phi} - \phi)$ denotes the annihilation operator. The first term represents the motion of the oscillator, the second, the anisotropy of the molecule rotated by an angle, ϕ, and the third, the external magnetic field. In single molecule magnets, there is a much larger distance between the levels of the molecule than between the different mechanical modes of a given level. Because of this, to consider the crossings between mechanical modes we consider the Hamiltonian to be projected onto the two lowest states of the molecule such that H_A describes a two level system considering only the lowest two levels of the molecule and taking the effects of the higher levels (which are comparatively far away) to be negligibly small.

The general form of H_A from equation 2 can be expressed as:

$$\hat{H}_A = \hat{H}_{\parallel} + \hat{H}_{\perp}, \tag{4}$$

where $\hat{H}_{\parallel}$ commutes with S_z and $\hat{H}_{\perp}$is a perturbation that does not commute with S_z. The eigenstates, $|\pm S>$, of S_z are degenerate ground states of $\hat{H}_{\parallel}$. $\hat{H}_{\perp}$, however, slightly perturbs the $|\pm S>$states. Calling the normalized perturbed states $|\psi_{\pm S}>$, the ground state and the first excited state become:

$$\Psi_{\pm} = \frac{1}{\sqrt{2}}(|\psi_S> \pm |\psi_{-S}>), \tag{5}$$

These two lowest states of the molecule, therefore, correspond to two eigenfunctions of the molecule recast as the symmetric and antisymmetric combinations of some other states which are approximately the states which point in one direction of minimum energy or the other.

The crossings of mechanical modes we consider are very similar to the points of resonance discussed earlier and shown in Figure 3. The difference is that now we are considering states of different phonon number rather than states of different spin number. In the ana-

lytical portion, we consider the points of anti-crossing for states of different phonon number individually. Our projected Hamiltonian is as follows:

$$\hat{H} = \begin{pmatrix} \hat{a}^\dagger \hat{a} + \frac{1}{2} + W & \Delta D(i\alpha) \\ \Delta D^\dagger(i\alpha) & \hat{a}^\dagger \hat{a} + \frac{1}{2} - W \end{pmatrix}, \tag{6}$$

where W denotes the magnetic field, Δ denotes the tunnel splitting and $D(i\alpha) = e^{i\alpha(\hat{a}^\dagger)+i\alpha(\hat{a})}$ denotes the displacement operator. Implementing first order perturbation theory to obtain the Hamiltonian in the basis of the two degenerate states yields:

$$H_{n,m} = \begin{pmatrix} n + \frac{1}{2} + W & E(n,m)\Delta \\ E*(n,m)\Delta & m + \frac{1}{2} - W \end{pmatrix}, \tag{7}$$

The two Fock states are denoted by n and m; the numerical value of n, m denoting the phonon number. In addition, we use the matrix elements of the displacement operator with respect to Fock states implemented by Perez-Leija et al [10] shown below:

$$E(n,m) = (-i\alpha)^{m-n} e^{-\frac{\alpha^2}{2}} \sqrt{\frac{n!}{m!}} L_n^{n-m}(\alpha^2), \tag{8}$$

$L_n^{n-m}(\alpha^2)$ denotes the Laguerre polynomial. Two examples of the results we obtained are displayed below:

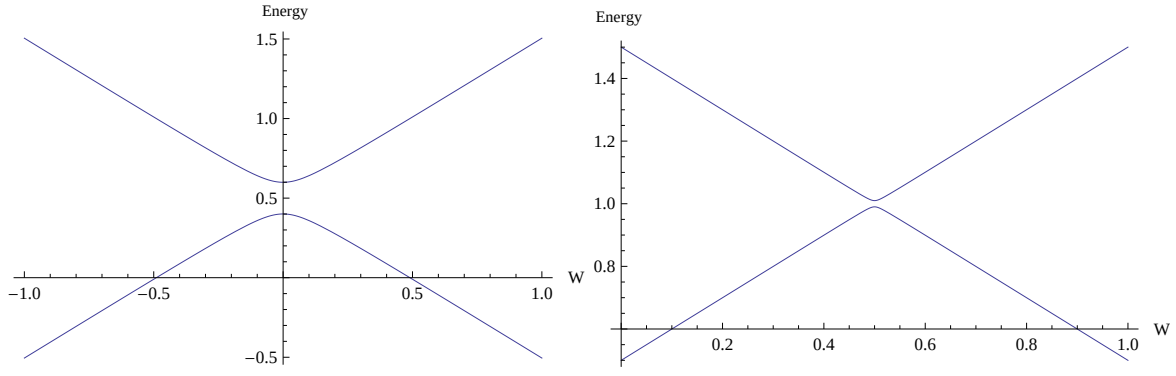

Figure 8: Left: Graph of the eigenvalue solutions for $n = 0$, $m = 0$ as a function of the magnetic field, W, where $\alpha = 0.1$ and $\Delta = 0.1$. Right: Graph of the eigenvalue solutions for $n = 0$, $m = 1$ as a function of the magnetic field. Again $\alpha = 0.1$ and $\Delta = 0.1$.

Also of note, is the dependence of the tunnel splitting on the parameter α. Alpha is defined as twice the spin multiplied by the value of the delocalization of zero point oscillations

(See equation 8):

$$\alpha = 2\mathbf{S}x, \tag{9}$$

where $x = \sqrt{\frac{\hbar}{2\omega_r I_z}}$ and $\omega_r = \sqrt{\frac{k}{I_z}}$; x represents the delocalization of zero point oscillations, ω_r represents the frequency, I_z the moment of in inertia and k the tortional spring constant. The value of x corresponds to the angle for which quantum mechanical effect should be seen, ie to see quantum mechanical effects you must be able to measure the angle with this precision. The proportionality of alpha to S shows that we can probe quantum mechanical effects with more sensitivity. There is an interesting quantum property that arises in our analytical data in that the tunnel splitting oscillates with an increasing value of alpha.

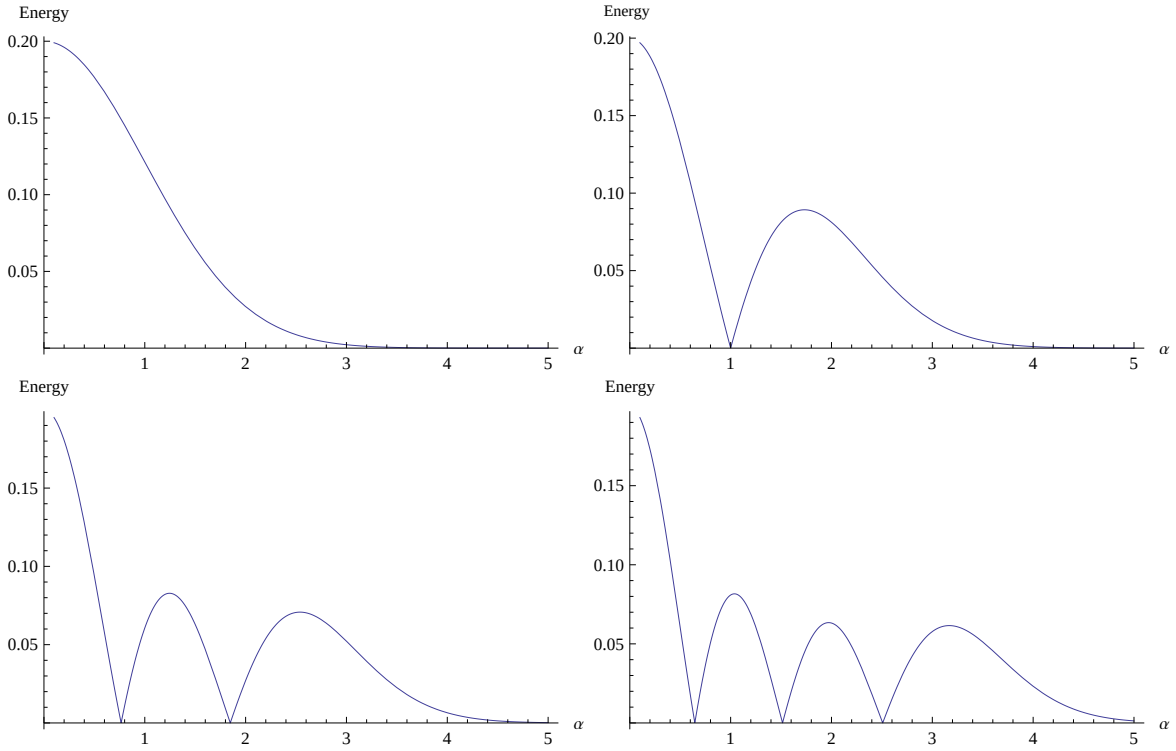

Figure 9: Graph of the dependence of the tunnel splitting on alpha for Left/Top: $n = m = 0$, Right/Top: $n = m = 1$, Left/Bottom: $n = m = 2$, and Right/Bottom: $n = m = 3$.

This dependence on alpha allows for the tunnel splitting to be tuned by a judicious choice in alpha. Following our analytical considerations, we looked at the effect of mechanical vibrations through a numerical model. This numerical calculation enabled us to consider the crossings of all states with different phonon number simultaneously. A sample of our

results is shown below:

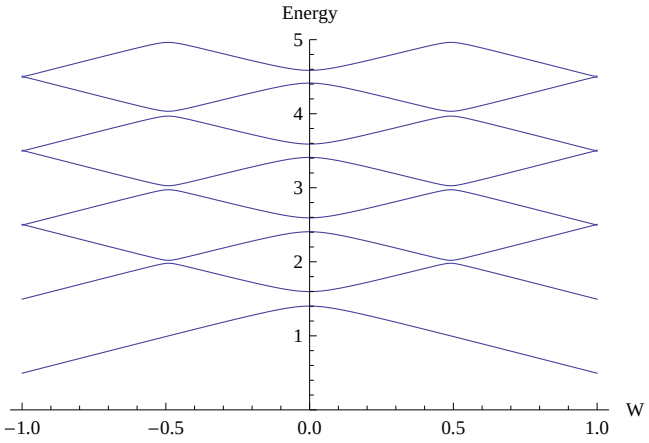

Figure 10: Numerical results depicting the energy levels (and thus tunnel splittings) as a function of the magnetic field for $\alpha = 0.1$ and $\Delta = 0.1$.

Tunnel splitting behavior can be observed at zero magnetic field and magnetopolariton behavior at crossings for crossings in the presence of magnetic field. Upon comparison of our numerical and analytical results we obtained good agreement. A plot for the crossing of a state containing one phonon with one containing no phonons is shown in Figure 9. This plot superimposes the analytical data with the numerical calculations enabling a comparison at the crossing point. In each case anti-crossing, or polariton, behavior occurs.

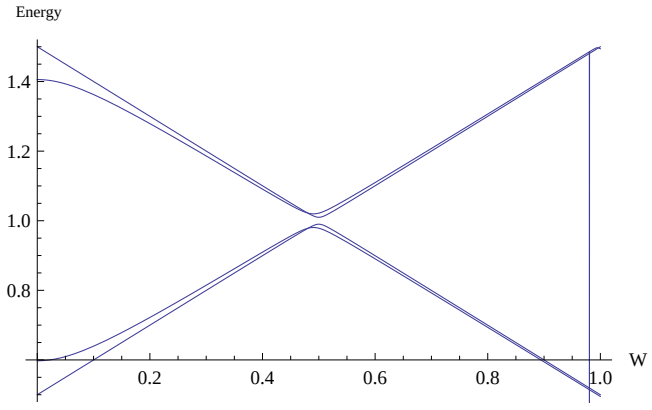

Figure 11: Superposition of the analytical and numerical results as a function of the magnetic field for $n = 0$ and $m = 1$ where $\alpha = 0.1$ and $\Delta = 0.1$.

C. Future Study and Conclusions

In a related study published in Nature in 2010 by A.D. O'Connel et al, it is shown that it is possible to cool a mechanical mode to its quantum ground state by using a microwave frequency mechanical oscillator coupled to a quantum bit. They also show that you can controllably create single quantum excitations, or phonons, in the resonator. Figure 11 depicts the change in the frequency of the qubit as it is tuned through the resonator frequency. Here there is a splitting depicted similar to what we expect to see from the coupling of mechanical oscillations to the macrospin of an SMM. In both the experiment shown here and our theoretical considerations the coupling of mechanical motion to a two-states system is considered. The fact that such behavior was experimentally found in this very similar system serves as further motivation to look for it in the case we describe.

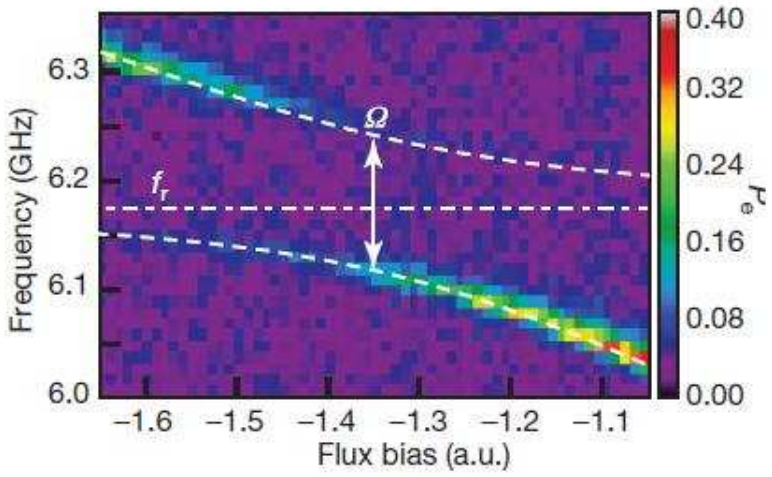

Figure 12: Qubit spectroscopy, showing P_e as a function of qubit frequency and microwave frequency. The qubit frequency behaves as expected, with a prominent splitting as the qubit is tuned through the resonator frequency.

For the case of a SMM attached to a carbon nanotube nanomotor, we find $\alpha \sim 10^{-2}$. To obtain this value we take $k \sim 10^{-18} N * m$ and $I_z \sim 10^{-34} kg * m^2$. This value is not large enough to observe supression of tunneling by the tuning of alpha. It is, however, large enough to observe the effect of magnetopolariton formation in these molecules. In previous experiments, such as that performed by Wernsdorfer and Sessoli in 1999, small splittings have been observed by Landau-Zener transitions [11]. We believe such techniques can also

be employed to detect the splitting in this case. In the case of the molecule bridged between conducting leads, we find $\alpha \sim 10$. To find this we take $\omega_r \sim 10^9 s^{-1}$and $I_z \sim 10^{-41} kg * m^2$. This large of a value for alpha would enable the experimental observation of the supression of tunneling if alpha was tuned accordingly.

In conclusion, we have been able to show theoretically that magnetopolariton behavior occurs in single molecule magnets when their magnetic moment becomes coupled to an extra degree of mechanical freedom. This effect could be applicable in a number of ways. One possible implementation would be to use the knowledge of the frequency and tunnel splitting to make precise mass measurements. In addition, if the effect of rotation can be made large enough in practical setting, such mechanical motion could be used to increase the tunnel splitting, effectively rewriting the spin information of the molecule. This, then, would enable the implementation of SMMs in high-density magnetic storage. Also, the implementation of tortional oscillations could be useful for quantum spin manipulation. The next step in the research will be to consider the molecule Hamiltonian in its full form instead of projecting it onto the two lowest states. This will enable us to determine the corrections needed to account for the interference of higher energy levels.

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