Magnetic Molecule on a Microcantilever: Quantum Magneto-Mechanical Oscillations

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We study the quantum dynamics of a system consisting of a magnetic molecule placed on a microcantilever. The molecule is assumed to be imbedded in a microcantilever of length \( L \). The cantilever is free at one end and firmly attached to the cantilever at the other end. The molecule is deposited on a microcantilever. The geometry of the proposed experiment is shown in Fig. 1. A magnetic molecule of spin \( S \) with a definite magnetic anisotropy axis being parallel the \( x \)-axis. A weak ac magnetic field of frequency \( \omega = \Delta/\hbar \), applied along the \( x \)-axis, will force the spin of the molecule to oscillate between the two orientations.

Conservation of the total angular momentum requires that the oscillations of the spin are accompanied by the mechanical oscillations of the cantilever (Einstein-de Haas effect [3, 4]). Consequently, if \( \omega = \Delta/\hbar \) coincides with a resonant mode of the cantilever, one should expect the effect of the ac field on the cantilever.

If the cantilever rotates by a small angle \( \delta \phi \) at the location of the spin, the Hamiltonian of the molecule \( \hat{H}_S \) becomes [5, 6]

\[
\hat{H}_S' = \hat{R}\hat{H}_S\hat{R}^{-1} = \hat{H}_S + i\sum [\hat{H}_S, \hat{S}] \cdot \delta \phi = \hbar \hat{S} \cdot \delta \phi. \tag{2}
\]

Due to strong magnetic anisotropy the molecule can be considered as a two-state system. The Hamiltonian of such a system, \( \mathcal{H}_2 \), is a projection of the Hamiltonian \( \mathcal{H}_3 \) onto the two states given by Eq. (1). These states can be viewed as the eigenstates of the Pauli matrix \( \sigma_z \).

For the geometry shown in Fig. 1, the projection can be performed by writing

\[
\mathcal{H}_2 = \sum_{i,j=\pm} \langle \Psi_i | \left( \hat{H}_S + i [\hat{H}_S, \hat{S}_x] \right) \delta \phi_x | \Psi_j \rangle | \Psi_i \rangle | \Psi_j \rangle \tag{3}
\]

with

\[
\sigma_x = |\Psi_+\rangle \langle \Psi_-| + |\Psi_-\rangle \langle \Psi_+| \tag{4}
\]

\[
\sigma_y = -i |\Psi_+\rangle \langle \Psi_-| + i |\Psi_-\rangle \langle \Psi_+| \tag{5}
\]

\[
\sigma_z = |\Psi_+\rangle \langle \Psi_-| - |\Psi_-\rangle \langle \Psi_+|. \tag{6}
\]

This gives

\[
\mathcal{H}_2 = -\frac{1}{2} \Delta \sigma_z + S \Delta \delta \phi_x \sigma_y. \tag{7}
\]

The states with a definite \( X \)-projection of the spin are eigenstates of \( \sigma_x \): \( |\pm S\rangle = \frac{1}{\sqrt{2}} (|\Psi_+\rangle \pm |\Psi_-\rangle) \). The expectation value of \( \sigma_z \) satisfies the Landau-Lifshitz equation:

\[
\hbar \dot{\sigma} = -\sigma \times \mathbf{b}_{\text{eff}}, \tag{8}
\]

with

\[
\mathbf{b}_{\text{eff}} = -\Delta \mathbf{e}_z + 2S \Delta \delta \phi_x \mathbf{e}_y. \tag{9}
\]
If the cantilever was held stationary ($\delta \phi_{x} = 0$), the solution of Eq. (9) would describe pure quantum oscillations of the spin: $\sigma_{x} = \text{const}, \sigma_{y} \propto \cos(\Delta t/h), \sigma_{y} \propto \sin(\Delta t/h)$.

We are interested in the coupled oscillations of the spin and the cantilever. Since the latter contains macroscopic number of atoms, its oscillations can be studied within a continuous elastic theory that deals with the displacement field $u(r, t)$ and the local rotation $\delta \phi$ given by [3]

$$\delta \phi(r) = \frac{1}{2} \nabla \times u(r).$$

Within such a model the spin of the molecule at a point $r_{0}$ can be replaced by the spin field

$$\Sigma(r, t) = S \sigma(t) \delta(r - r_{0}),$$

where $\sigma(t)$ satisfies Eq. (5). With account of Eq. (2) the energy of the cantilever becomes

$$H_{C} = H_{E} + \frac{1}{2} \int d^{3}r \hbar \Sigma \cdot (\nabla \times u),$$

where $H_{E}$ is the part of the elastic energy that is independent of the spin.

The dynamical equation for the displacement field is

$$\rho \frac{\partial^{2} u_{\alpha}}{\partial t^{2}} = \frac{\partial \sigma_{\alpha \beta}}{\partial x_{\beta}} ,$$

where $\sigma_{\alpha \beta} = \delta H_{C}/\delta u_{\alpha \beta}$ is the stress tensor, $\epsilon_{\alpha \beta} = \partial u_{\alpha}/\partial x_{\beta}$ is the strain tensor, and $\rho$ is the mass density of the material. This gives

$$\rho \frac{\partial^{2} u_{\alpha}}{\partial t^{2}} - \frac{\partial \sigma_{\alpha \beta}^{(E)}}{\partial x_{\beta}} = - \frac{\hbar}{2} \nabla \times \Sigma$$

where $\sigma_{\alpha \beta}^{(E)} = \delta H_{E}/\delta u_{\alpha \beta}$. It is easy to see that in the absence of the external torque, $K_{\alpha}^{(E)} = \oint dA \sum_{\beta} \rho \epsilon_{\alpha \beta} r_{\beta} \sigma_{\beta \gamma}^{(E)}$, applied to the surface of the body $A$, Eq. (14) provides conservation of the total angular momentum,

$$J = \int d^{3}r \left[ \hbar \Sigma + \rho (r \times \dot{u}) \right] , \quad dJ/dt = 0 .$$

Writing the left-hand side of Eq. (14) in the conventional form for a cantilever [4] one obtains the elastic equation that couples vertical displacements of the cantilever, $u_{y}(y, t)$, with the oscillations of the spin:

$$\rho \frac{\partial^{2} u_{z}}{\partial t^{2}} + \frac{\hbar^{2} E}{12(1 - \sigma^{2})} \frac{\partial^{4} u_{z}}{\partial y^{4}} = \hbar S \frac{\partial}{\partial y} \frac{\partial}{\partial t} \left[ \sigma_{x}(t) \Delta (y - y_{0}) \right] .$$

Here $h$ and $V$ are the thickness and the volume of the cantilever, respectively, $E$ is the Young’s modulus, and $\sigma$ is the Poisson coefficient, $-1 < \sigma < 1/2$.

It is convenient to switch in Eq. (16) to dimensionless variables $\bar{u}_{z} = u_{z}/L, \bar{y} = y/L, \bar{t} = t/V$, where

$$\nu \equiv \sqrt{\frac{Eh^{2}}{12\rho(1 - \sigma^{2})L^{4}}}$$

determines the scale of the eigenfrequencies of the oscillations of the cantilever. By order of magnitude $\nu \sim v_{s} h/L^{2}$ where $v_{s} \sim \sqrt{E/\rho}$ is the speed of sound. In terms of these variables Eq. (16) becomes

$$\frac{\partial^{2} \bar{u}_{z}}{\partial \bar{t}^{2}} + \frac{\partial^{4} \bar{u}_{z}}{\partial \bar{y}^{4}} = \frac{\epsilon}{2} \frac{\partial^{2}}{\partial \bar{y}} \left[ \sigma_{x}(t) \delta(\bar{y} - \bar{y}_{0}) \right] ,$$

where $0 < \bar{y}_{0} < 1$ and

$$\epsilon = \frac{hS}{ML^{2}v} = \frac{hS}{M} \frac{12\rho(1 - \sigma^{2})}{Eh^{2}} \frac{1}{V} \frac{1}{\nu}$$

is a dimensionless small parameter. By order of magnitude, $\epsilon \sim hS/(Mv_{s} h)$, where $M = \rho V$ is the mass of the cantilever. For, e.g., a molecule of spin $S = 10$ on a cantilever of dimensions $100 \text{nm} \times 10 \text{nm} \times 1 \text{nm}$ the parameter $\epsilon$ should be of order $10^{-7}$. Eq. (18) has to be solved with the following boundary conditions:

$$\bar{u}_{z} = 0 , \quad \frac{\partial \bar{u}_{z}}{\partial \bar{y}} = 0 \quad \text{at} \quad \bar{y} = 0 ,$$

$$\frac{\partial^{2} \bar{u}_{z}}{\partial \bar{y}^{2}} = 0 , \quad \frac{\partial^{2} \bar{u}_{z}}{\partial \bar{y}^{3}} = 0 \quad \text{at} \quad \bar{y} = 1 .$$

The first two conditions correspond to the absence of displacement and the absence of bending of the cantilever at the fixed end, while the last two conditions correspond to the absence of torque and force, respectively, at the free end.

For the free oscillations of the cantilever ($\epsilon = 0$) one writes

$$\bar{u}_{z}(\bar{y}, \bar{t}) = \bar{u}(\bar{y}) \cos(\bar{\omega} \bar{t}) .$$

Substitution into Eq. (18) with $\epsilon = 0$ then gives

$$\frac{\partial^{4} \bar{u}}{\partial \bar{y}^{4}} - \kappa^{2} \bar{u} = 0 , \quad \kappa^{2} = \bar{\nu} .$$

Solutions are

$$\bar{u}(\bar{y}) = (\cos \kappa + \cosh \kappa) \left[ \cos(\kappa \bar{y}) - \cosh(\kappa \bar{y}) \right]$$

$$+ (\sin \kappa - \sinh \kappa) \left[ \sin(\kappa \bar{y}) - \sinh(\kappa \bar{y}) \right] .$$

The third of the boundary conditions (20) provides the equation,

$$\cos \kappa \cosh \kappa + 1 = 0 ,$$

for the frequencies of the normal modes of the cantilever, $\bar{\omega}_{n} = \kappa^{2}_{n}$ (measured in the units of $\nu$ of Eq. (17)). The fundamental (minimal) frequency is $\bar{\omega}_{1} \approx 3.516$. The next two frequencies are $\bar{\omega}_{2} \approx 22.03$ and $\bar{\omega}_{3} \approx 61.70$. The profiles of the oscillations of the cantilever for three normal modes ($n = 1, 2, 3$) are shown in Fig. 2.

To consider coupled oscillations of the cantilever and the spin of the molecule we first neglect dissipation and write for the displacement

$$\bar{u}_{z}(\bar{y}, \bar{t}) = \sum_{m} R_{m}(\bar{t}) \bar{u}_{m}(\bar{y}) ,$$

for the frequencies of the normal modes of the cantilever.
where $R_m(t)$ are functions of time to be determined and $\vec{u}_m(\bar{y})$ is a normalized eigenfunction \footnote{23} that corresponds to the eigenvalue $\kappa_m$ given by Eq. \footnote{24},

$$\int_0^1 dy \vec{u}_m(\bar{y})\vec{u}_n(\bar{y}) = \delta_{mn}. \quad (26)$$

Substitution of Eq. \footnote{25} into Eq. \footnote{18} gives

$$\sum_m \left( \frac{d^2 R_m}{dt^2} + \omega_m^2 R_m \right) \bar{u}_m(\bar{y}) = \frac{\epsilon}{2} \frac{d}{dt} \frac{d}{dy} \left[ \sigma_x(t) \delta(\bar{y} - \bar{y}_0) \right], \quad (27)$$

where we have used Eq. \footnote{22}. Multiplying both parts of this equation by $\bar{u}_m(\bar{y})$ and integrating over $\bar{y}$ from 0 to 1 with account of Eq. \footnote{26}, one obtains linear second-order differential equation for $R_n(t)$,

$$\frac{d^2 R_n}{dt^2} + \omega_n^2 R_n = -\epsilon \left( \frac{d\sigma_x}{dt} \right) \bar{u}_n'(\bar{y}_0), \quad (28)$$

where $\bar{u}_n'(\bar{y}_0) \equiv (d\bar{u}_n/d\bar{y})_{\bar{y} = \bar{y}_0}$.

Coupled magneto-mechanical oscillations near the ground state correspond to small $\sigma_x, \sigma_y, R_n$, and $\sigma_z \approx 1$. This requires temperatures $k_B T \ll \Delta$. In this case Eq. \footnote{8} gives for $\sigma_x$

$$\frac{d^2 \sigma_x}{dt^2} + \Delta^2 \sigma_x = S\Delta \bar{u}_n'(\bar{y}_0) \frac{dR_n}{dt}, \quad (29)$$

where $\Delta \equiv \Delta/(\hbar v)$. Substituting into Eqs. \footnote{28} and \footnote{29} $\sigma_x(t), R_n(t) \propto \exp(i\bar{w}t)$, one obtains the following equation for the eigenfunctions of the coupled oscillations:

$$(\bar{\omega}^2 - \omega_n^2)(\bar{\omega}^2 - \Delta^2) = \frac{1}{2} \epsilon S \Delta \bar{u}_n'(\bar{y}_0) \bar{\omega}^2. \quad (30)$$

Due to the smallness of $\epsilon$, oscillations of the spin and the cantilever occur independently at frequencies $\Delta/\hbar$ and $\omega_n$, respectively, unless these two frequencies are very close to each other. The latter can be achieved by, e.g., changing $\Delta$ with the help of the dc magnetic field perpendicular to the anisotropy axis of the molecule. At $\Delta = \hbar \omega_n$ one should observe the splitting of the mechanical mode of the cantilever, $\omega_n$, into two modes

$$\omega_{n\pm} = \omega_n \left( 1 \pm \frac{\delta}{2} \right), \quad \delta = \sqrt{\frac{\epsilon S \bar{u}_n'^2(\bar{y}_0)}{2\omega_n}}. \quad (31)$$

The remarkable property of Eq. \footnote{31} is that it has no free parameters. For a chosen resonance $\Delta = \hbar \omega_n$, the relative splitting $\delta$ depends only on the position of the molecule on the cantilever. This dependence is plotted in Fig. \footnote{3}.

We shall now demonstrate that the above experiment can be performed by studying the response of the system to a weak ac magnetic field, $\mathbf{B}(t) = B_0 \mathbf{e}_x \sin(\omega t)$, applied along the anisotropy axis of the magnetic molecule. In the presence of such a field Eq. \footnote{9} becomes

$$\mathbf{b}_{\text{eff}} = -\Delta \mathbf{e}_z + 2S\Delta \delta \phi_x \mathbf{e}_y + b \mathbf{e}_x \sin(\omega t), \quad (32)$$

where $b = g \mu_B B_0$, with $g$ being the gyromagnetic factor for the spin and $\mu_B$ being the Bohr magneton. To obtain the amplitude of the forced oscillations of the cantilever we need to include dissipation in the equations of motion of the spin and the cantilever. The damping modifies the Landau-Lifshitz equation \footnote{10}:

$$\hbar \dot{\mathbf{g}} = -\mathbf{g} \times \mathbf{b}_{\text{eff}} + 2Q_s^{-1} \mathbf{g} \times (\mathbf{g} \times \mathbf{b}_{\text{eff}}). \quad (33)$$

Here $Q_s$ is the quality factor of the spin oscillations. Small oscillations ($|\sigma_{x,y}| \ll 1, \sigma_z \approx 1$) now satisfy

$$\frac{d\sigma_x}{dt} + \Delta (Q_s^{-1} \pm i) \sigma_x = S \Delta \bar{u}_n'(\bar{y}_0) R_n - ib \sin(\bar{w}t). \quad (34)$$

where $\sigma_{\pm} = \sigma_x \pm i\sigma_y$ and $b \equiv b/(\hbar v)$.
Dissipation of the mechanical motion of the cantilever can be introduced by adding the first time derivative of $R_n$ to Eq. (28):

$$\frac{d^2 R_n}{dt^2} + \frac{\bar{\omega}_n}{Q_n} \frac{dR_n}{dt} + \bar{\omega}_n^2 R_n = -\frac{\epsilon}{4} \bar{u}_n(t) \frac{d}{dt} (\sigma_+ + \sigma_-). \tag{35}$$

Here $Q_n$ is the quality factor of the oscillations of the cantilever at the eigenfrequency $\bar{\omega}_n$. One can now obtain the time dependence of $R_n$ and $\sigma_\pm$ by solving together Eq. (34) and Eq. (35). The displacement of the cantilever at a point $y$ is given by $\bar{u}_z(t, \bar{y}) = R_n(t) \bar{u}_n(\bar{y})$. The simplest way to get the solution is to replace $i \sin(\bar{\omega}t)$ in Eq. (34) with $\exp(i\bar{\omega}t)$ and solve the resulting three linear algebraic equations for $R_n, \sigma_\pm \propto \exp(i\bar{\omega}t)$. The applicability of the formulas obtained that way is limited by the range of parameters that provide the condition $|\sigma_\pm| \ll 1$ used to derive Eq. (34) from Eq. (8). It is easy to see from Eq. (34) that this requirement is violated for $\sigma_\pm$ when $\omega$ is close to $\Delta$: The strong pumping of the spin excitations by the ac field at $\omega \to \Delta$ leads to the breakdown of the linear approximation for the dynamics of the spin governed by Eq. (8).

At $\omega = \omega_n \neq \Delta$, in the practical range of the quality factors, $1 \ll Q_{n,s} \ll 1/\epsilon$, one obtains

$$|\bar{u}_z(\bar{y}_0, \bar{y})| = \frac{b Q_n \Delta |\bar{u}_n'(\bar{y}_0)| |\bar{u}_n(\bar{y})|}{2\bar{\omega}_n |\bar{\omega}_n^2 - \Delta^2|}, \quad |\sigma_\pm| = \frac{b}{|\bar{\omega}_n \pm \Delta|}. \tag{36}$$

The parameter $\Delta$ can be controlled by a dc magnetic field applied perpendicular to the anisotropy axis of the molecule. The condition $|\sigma_-| \ll 1$ determines how close to the double resonance, $\Delta = \omega_n$, one can use Eq. (36): $|\omega_n - \Delta| \gg b/2$. Substitution of $|\omega_n - \Delta| \sim b$ into the first of Eqs. (36) provides the estimate for the maximum amplitude of the oscillations of the cantilever at $\Delta \to \omega_n$:

$$\max |\bar{u}_z(\bar{y}_0, \bar{y})| \sim \frac{\epsilon Q_n}{4 \bar{\omega}_n} |\bar{u}_n'(\bar{y}_0)| |\bar{u}_n(\bar{y})|. \tag{37}$$

It is a function of the distance $y$ from the fixed end of the cantilever, parameterized by the position of the molecule $y_0$.

For, e.g., $n = 1$ the parameter $\delta$ of Eq. (34) should be generally of order $\sqrt{\epsilon S}$. A cantilever of dimensions $100\text{nm} \times 100\text{nm} \times 1\text{nm}$, carrying a magnetic molecule of spin 10, will have $\nu \sim 10^8 \text{s}^{-1}, \epsilon \sim 10^{-7}, \delta \sim 10^{-3}$, see Eqs. (37), (10), and (34). This would give $f_1 = 3.52 \nu/(2\pi) \sim 30\text{MHz}$ and the splitting, $\delta f_1$, of the first harmonic of the cantilever in the ballpark of a few kilohertz. The condition for the detection of the splitting is $Q_{n,s} \gg 1/\delta$. For a single magnetic molecule on a microcantilever the discrete character of the cantilever phonon modes and the absence of the inhomogeneous broadening of the spin mode (usually present in a bulk molecular magnet) should provide a high spin quality factor. Very high quality factors (up to $10^5$) have also been reported for microcantilevers [11, 12]. When the molecule is near the tip of the cantilever, Eq. (37) with $\epsilon \sim 10^{-7}$ and $Q_n \sim 10^5$ gives for the tip: $|\bar{u}_z| \sim 0.005$. For a 100-nm long cantilever this would correspond to the oscillations by half a nanometer. Such a displacement can be detected by tunneling or force microscopy, as well as by optical and electrical methods. Working with even smaller cantilevers would provide higher values of $\epsilon$ and would allow to relax the requirement on the quality factor.

In Conclusion, we have shown that driven quantum oscillations of the spin of a magnetic molecule can be observed by placing the molecule on a microcantilever. Since such cantilevers consist of hundreds of thousands of atoms, this would be a remarkable example of a macroscopic quantum effect. Our theory has no free parameters and, therefore, it must be helpful in designing the proposed experiment.

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