

Rabi Spin Oscillations Generated by Ultrasound in Solids

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It is shown that ultrasound in the gigahertz range can generate space-time Rabi oscillations between spin states of molecular magnets. We compute dynamics of the magnetization generated by surface acoustic waves and discuss conditions under which this novel quantum effect can be observed.

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Quantum mechanics of a spin cluster (e.g., a magnetic molecule) embedded in a solid is determined by the crystal field. The latter depends on the symmetry of the cluster and its environment [1]. Crystal field Hamiltonians provide a good description of molecular magnets at low temperature. For instance, dynamics of a spin \mathbf{S} that prefers to look up or down along the anisotropy axis of the cluster can be described by a Hamiltonian $\hat{\mathcal{H}} = -DS_z^2$, where the anisotropy constant D arises from spin-orbit interactions. We are interested in the effect of the mechanical rotation of the crystal on the molecular spin. For a similar problem involving an orbital moment \mathbf{L} , it is well known from classical mechanics that in the reference frame rotating at an angular velocity $\boldsymbol{\Omega}$ the Hamiltonian acquires a term $-\hbar\mathbf{L} \cdot \boldsymbol{\Omega}$ (we use dimensionless L and S). The same rule is expressed by the Larmor theorem in classical electrodynamics: Rotation is equivalent to the magnetic field, leading to the effective Zeeman term, $-\mathbf{M} \cdot \mathbf{B}$, in the rotating-frame Hamiltonian, with $\mathbf{M} = \hbar\gamma\mathbf{L}$, $\mathbf{B} = \boldsymbol{\Omega}/\gamma$, and γ being the gyromagnetic ratio. The extension of the Larmor theorem to a spin is a consequence of the fact that in relativistic quantum theory the generator of rotations is $\hat{\mathbf{J}} = \hat{\mathbf{L}} + \hat{\mathbf{S}}$. Rigorous derivation of the term $-(\hat{\mathbf{L}} + \hat{\mathbf{S}}) \cdot \boldsymbol{\Omega}$ in the Hamiltonian can be obtained, e.g., from the study of the nonrelativistic limit of the Dirac equation in the rotating frame [2].

Equivalence of the rotation to the magnetic field explains Barnett effect [3]: Rotation of a body of the magnetic susceptibility χ at an angular velocity $\boldsymbol{\Omega}$ generates a magnetic moment $\mathbf{M} = \chi\boldsymbol{\Omega}/\gamma$. The “spin-rotation coupling,” $-\hbar\mathbf{S} \cdot \boldsymbol{\Omega}$, can also lead to nontrivial quantum effects. Consider, e.g., a spin cluster with the Hamiltonian $\hat{\mathcal{H}} = -D\hat{S}_z^2$ that preserves the direction of the spin along the anisotropy axis Z due to commutation of $\hat{\mathcal{H}}$ with \hat{S}_z . In the presence of the rotation about, e.g., the x axis of the crystal the Hamiltonian in the rotating frame becomes $\hat{\mathcal{H}}' = -D\hat{S}_z^2 - \hbar\hat{S}_x\Omega$. This Hamiltonian, unlike $\hat{\mathcal{H}}$, does not commute with \hat{S}_z and, therefore, allows transitions between the two orientations of \mathbf{S} along the anisotropy axis. Thus, rotation alone can induce quantum transitions between spin states that are prohibited by the Hamiltonian

of a stationary system $\hat{\mathcal{H}}$. We should emphasize that switching from the laboratory-frame Hamiltonian $\hat{\mathcal{H}}$ to the rotating-frame Hamiltonian $\hat{\mathcal{H}}' = \hat{\mathcal{H}} - \hbar\hat{\mathbf{S}} \cdot \boldsymbol{\Omega}$ does not introduce any new spin-lattice interactions in addition to the crystal field. It is just another method to obtain a solution of the problem, which, in the laboratory frame, requires introduction of the time dependence of the crystal field; e.g., $\hat{\mathcal{H}} = -D\hat{S}_z^2$, in the presence of rotation, becomes $\hat{\mathcal{H}} = -D[\mathbf{n}(t) \cdot \hat{\mathbf{S}}]^2$ with $\mathbf{n}(t)$ being the instantaneous direction of the anisotropy axis.

So far, quantum spin-rotation effects received little attention because only a very tiny magnetic field due to rotation can be produced in the rotating frame of a macroscopic body. Consequently, the corresponding quantum effects have very low probability. This Letter is based upon the observation that *local* rotations of the crystal lattice produced by high-frequency ultrasound can easily provide 10–100 G fictitious fields in the rotating frame of a rigid spin cluster in a solid. Indeed, in the presence of the phonon displacement field $\mathbf{u}(\mathbf{r}, t)$, the angle of the local rotation of the crystal lattice $\delta\phi(\mathbf{r}, t)$ and the corresponding angular velocity $\boldsymbol{\Omega}(\mathbf{r}, t)$ are given by [4]

$$\delta\phi(\mathbf{r}, t) = \frac{1}{2}\nabla \times \mathbf{u}(\mathbf{r}, t), \quad \boldsymbol{\Omega}(\mathbf{r}, t) = \frac{1}{2}\nabla \times \dot{\mathbf{u}}(\mathbf{r}, t). \quad (1)$$

For a transverse sound wave of frequency $f \sim 3$ GHz and amplitude $u_0 \sim 1$ nm, this gives $B = \Omega/\gamma \sim 10$ G in the rotating frame coupled to the local crystallographic axes. Even greater local fields can be achieved with surface acoustic waves that have been recently used in experiments on molecular magnets [5].

The equivalence of the effect of high-frequency transverse acoustic waves to the effect of high-amplitude ac magnetic field on paramagnetic spins immediately suggests that one can try to generate Rabi spin oscillations with the help of high-frequency ultrasound. Rabi effect [6] corresponds to the oscillation of the occupation numbers of two quantum levels in the presence of an ac field whose frequency is close to the distance between the levels. On resonance, the frequency of Rabi oscillations is proportional to the amplitude of the ac field. The effort to observe Rabi oscillations between quantum states of molecular

magnets in experiments employing ac magnetic fields [7–10] has been going on for some time. For such experiments to succeed, the Rabi frequency must be greater than the spin decoherence rate. This typically requires the amplitude of the ac field to be greater than 1 G, which is not easy to achieve with electromagnetic waves but, as we have seen, is possible with surface acoustic waves. Note that the condition of the validity of the elastic theory $u_0 \ll \lambda$ (where λ is the phonon wavelength) automatically provides the condition $\Omega \ll \omega = 2\pi f$, which allows one to treat local rotations classically while treating the two-level system with level separation $\hbar\omega$ quantum mechanically.

For certainty we consider a crystal of molecular magnets with the anisotropy Hamiltonian

$$\hat{\mathcal{H}}_A = -D\hat{S}_z^2 + \hat{V}, \quad (2)$$

where \hat{V} is a small term responsible for the tunnel splitting Δ of spin-up and spin-down states. The spin cluster is assumed to be more rigid than its elastic environment, so that the long-wave crystal deformations can only rotate it as a whole but cannot change its inner structure responsible for the parameters of the Hamiltonian $\hat{\mathcal{H}}_A$. This approximation should apply to many molecular magnets as they typically have a compact magnetic core inside a large unit cell of the crystal. We choose geometry in which surface acoustic waves are running along the x axis with the solid extending towards $y > 0$; see Fig. 1. Using standard formulas [4] for the displacement field in a surface acoustic wave, one obtains

$$\delta\boldsymbol{\phi}(\mathbf{r}) = \frac{1}{2} \frac{\omega}{c_t} u_0 e^{-k_t y} \cos(kx - \omega t) \mathbf{e}_z \equiv \delta\phi(x, t) \mathbf{e}_z, \quad (3)$$

where $\omega = c_t k \xi$, $k_t \equiv k\sqrt{1 - \xi^2}$, ξ is a real number between 0 and 1 satisfying

$$\xi^6 - 8\xi^4 + 8\xi^3 \left(3 - 2\frac{c_t^2}{c_l^2}\right) - 16\left(1 - \frac{c_t^2}{c_l^2}\right) = 0, \quad (4)$$

and $c_{t,l}$ are velocities of transverse and longitudinal sound.

In the presence of deformations of the crystal lattice, local anisotropy axes defined by the crystal field are rotated by the angle given by Eq. (3). In the Hamiltonian, this rotation is equivalent to the rotation of the operator $\hat{\mathbf{S}}$ in the opposite direction, which can be performed by the $(2S + 1) \times (2S + 1)$ matrix in the spin space [11],

$$\hat{\mathbf{S}} \rightarrow \hat{R}^{-1} \hat{\mathbf{S}} \hat{R}, \quad \hat{R} = e^{i\hat{\mathbf{S}} \cdot \delta\boldsymbol{\phi}}. \quad (5)$$

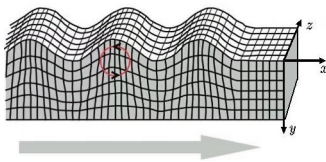


FIG. 1 (color online). Geometry of the problem.

The spin Hamiltonian in the laboratory frame becomes

$$\hat{\mathcal{H}} = e^{-i\hat{\mathbf{S}} \cdot \delta\boldsymbol{\phi}} \hat{\mathcal{H}}_A e^{i\hat{\mathbf{S}} \cdot \delta\boldsymbol{\phi}}. \quad (6)$$

In order to find the laboratory-frame wave function $|\Psi\rangle$, it is useful to introduce the *lattice-frame* wave function $|\Psi^{(\text{lat})}\rangle$, defined through the unitary transformation

$$|\Psi^{(\text{lat})}\rangle = e^{i\delta\boldsymbol{\phi} \cdot \hat{\mathbf{S}}} |\Psi\rangle. \quad (7)$$

Differentiating it on time it is easy to see that this function satisfies the Schrödinger equation with the lattice-frame Hamiltonian

$$\hat{\mathcal{H}}^{(\text{lat})} = \hat{\mathcal{H}}_A - \hbar\hat{\mathbf{S}} \cdot \boldsymbol{\Omega} \quad (8)$$

where

$$\boldsymbol{\Omega} = \frac{\partial \delta\boldsymbol{\phi}}{\partial t} = \frac{1}{2} \frac{\omega^2}{c_t} u_0 e^{-k_t y} \sin(kx - \omega t) \mathbf{e}_z. \quad (9)$$

To this point we have not made any assumptions about the magnitude of $\delta\boldsymbol{\phi}$, so that the equations (6) and (8) are exact. An interesting observation for the comparison of the effects of ultrasound and ac magnetic field is that the Hamiltonian (8) resembles the Hamiltonian of a particle of spin $\hat{\mathbf{S}}$ in the ac magnetic field which amplitude scales as the square of the frequency.

We are interested in the Rabi oscillations between the two lowest states of $\hat{\mathcal{H}}_A$:

$$|\phi_{\pm}\rangle = \frac{1}{\sqrt{2}} (|S\rangle \pm |-S\rangle), \quad (10)$$

where $|\pm S\rangle$ satisfy $\hat{S}_z |\pm S\rangle = \pm S |\pm S\rangle$. It makes sense, therefore, to project our Hamiltonian on the $|\pm S\rangle$ states, making the problem essentially a two-state problem. This gives

$$\hat{h}_{\text{eff}}^{(\text{lat})} = -\frac{\Delta}{2} \hat{\sigma}_1 - \hbar\omega_R \sin(kx - \omega t) \hat{\sigma}_3, \quad (11)$$

where Δ is the energy distance between the ground state $|\phi_+\rangle$ and the first excited state $|\phi_-\rangle$, $\hat{\sigma}_1 \equiv |S\rangle\langle -S| + |-S\rangle\langle S|$, $\hat{\sigma}_3 \equiv |S\rangle\langle S| - |-S\rangle\langle -S|$, and

$$\omega_R = \frac{1}{2c_t} \omega^2 u_0 S e^{-k_t y}. \quad (12)$$

The two-state approach will be valid if Δ and $\hbar\omega$ are small in comparison with the distances to other spin levels. Note that the tunnel splitting Δ originates from the term \hat{V} in $\hat{\mathcal{H}}_A$ that does not commute with \hat{S}_z .

It is easy to check that at $\omega \sim \Delta/\hbar$, which is our case of interest for consideration of Rabi oscillations, the second term in Eq. (11) is small compared to the first term as long as the wavelength of the acoustic wave satisfies $\lambda \gg Su_0$. For a not very large S this condition is always fulfilled by surface acoustic waves. The approximate eigenstates of the problem are then the eigenstates of σ_1 given by Eq. (10).

Their energies are $\pm\Delta/2$. The second term in Eq. (11) produces transitions between these states, resulting in the Rabi oscillations when $\hbar\omega \approx \Delta$. The standard way to obtain the evolution of the wave function is to apply the *rotating wave approximation* [6]. Note that the coordinates x and y in Eq. (11) can be viewed as parameters. Expressing the wave function as

$$|\Psi(t)^{(\text{lat})}\rangle = C_+(t)|\phi_+\rangle + C_-(t)|\phi_-\rangle, \quad (13)$$

and starting with $C_-(0) = 0$, $C_+(0) = 1$ at $t = 0$, $x = 0$, one obtains

$$C_-(t) = \frac{\omega_R}{\Omega_R} e^{-(i/2)\omega t} \sin\left(\frac{\Omega_R t}{2}\right), \quad (14)$$

$$C_+(t) = \left[\cos\left(\frac{\Omega_R t}{2}\right) + i \frac{\Delta/\hbar - \omega}{\Omega_R} \sin\left(\frac{\Omega_R t}{2}\right) \right] e^{(i/2)\omega t},$$

where

$$\Omega_R = \sqrt{(\Delta/\hbar - \omega)^2 + \omega_R^2}. \quad (15)$$

Assuming that every spin was in the ground state $|\phi_+\rangle$ before the sound wave arrived, the spatial dependence of the wave function can be obtained by making a replacement $t \rightarrow t - kx/\omega$ in Eq. (14). In the absence of spatial derivatives in the Hamiltonian (11), $|\Psi(t)^{(\text{lat})}\rangle$ is defined up to the phase factor $\exp[i\theta(x, y)]$ with θ being an arbitrary real function of coordinates. From Eq. (7) the wave function of the system in the laboratory frame is $|\Psi\rangle = e^{-i\delta\phi \cdot S} |\Psi^{(\text{lat})}\rangle$. Because of the smallness of $\delta\phi$, the dynamics of $|\Psi\rangle$ essentially coincides with the dynamics of $|\Psi^{(\text{lat})}\rangle$ and is given by Rabi oscillations between the states $|\phi_\pm\rangle$ at the frequency Ω_R . This is confirmed by numerical calculations with the lattice-frame and laboratory-frame Hamiltonians; see Fig. 2.

The expectation value of the projection of the spin onto the anisotropy axis (the Z axis) is given by

$$\begin{aligned} \langle \Psi(t) | \hat{S}_z | \Psi(t) \rangle &= 2S \frac{\omega_R}{\Omega_R^2} \left\{ \left(\omega - \frac{\Delta}{\hbar} \right) \sin(\omega t - kx) \right. \\ &\quad \times \sin^2 \left[\frac{1}{2} (\Omega_R t - K_R x) \right] \\ &\quad \left. + \frac{1}{2} \Omega_R \cos(\omega t - kx) \sin(\Omega_R t - K_R x) \right\}, \end{aligned} \quad (16)$$

where $K_R = (\Omega_R/\omega)k \ll k$ can be called the ‘‘Rabi’’ wave vector. Thus, the space-time Rabi oscillations of the occupation numbers of spin states generate space-time oscillations of the magnetization of the crystal. On resonance, when $\omega = \Delta/\hbar$, Eq. (16) simplifies to

$$\langle \Psi(t) | \hat{S}_z | \Psi(t) \rangle = S \cos(kx - \omega t) \sin(\omega_R t - k_R x), \quad (17)$$

with $k_R = (\omega_R/\omega)k \ll k$. The condition $\omega_R \ll \omega$ ($Su_0 \ll \lambda$) implies that the time dependence of $\langle \hat{S}_z \rangle$ at any point in

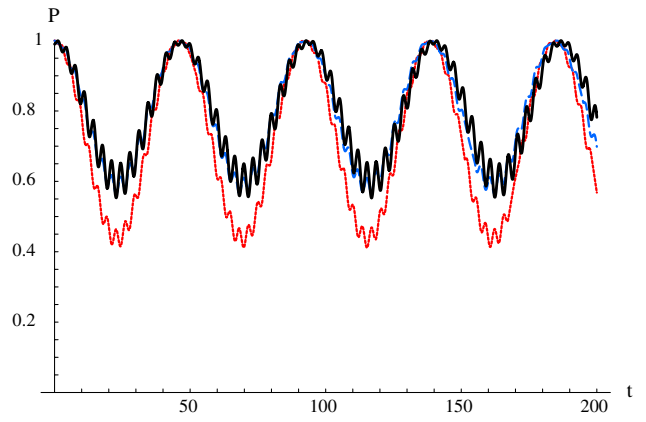


FIG. 2 (color online). Time dependence ($t' \equiv t\Delta/\hbar$) of the probability to find the spin in the state $|\phi_+\rangle$ at $x = 0$, $S = 10$, $\omega_R = 0.1\omega$, and $\omega = 0.9(\Delta/\hbar)$ in the laboratory frame. Dotted red line: Numerical result for the laboratory-frame Hamiltonian (19). Dashed blue line: Numerical result obtained with the use of Hamiltonian (11). Solid black line: Analytical result obtained with the use of Eq. (14).

space consists of the oscillations at frequency ω with beats of frequency ω_R . Similarly, $\langle \hat{S}_z \rangle$ at any moment of time oscillates in space with the wave vector k and exhibits beats with the wave vector $k_R \ll k$.

Our conclusions can be checked by obtaining the full solution of the problem in the laboratory frame in a particular case of a biaxial symmetry, when \hat{V} in Eq. (2) equals $E(\hat{S}_x^2 - \hat{S}_y^2)$. In this case Eq. (6) reduces to

$$\hat{\mathcal{H}} = -D\hat{S}_z^2 + \frac{E}{2} \{ \hat{S}_+^2 e^{-2i\delta\phi(x,t)} + \hat{S}_-^2 e^{2i\delta\phi(x,t)} \}, \quad (18)$$

where $\hat{S}_\pm = \hat{S}_x \pm i\hat{S}_y$. The second term can be treated as a

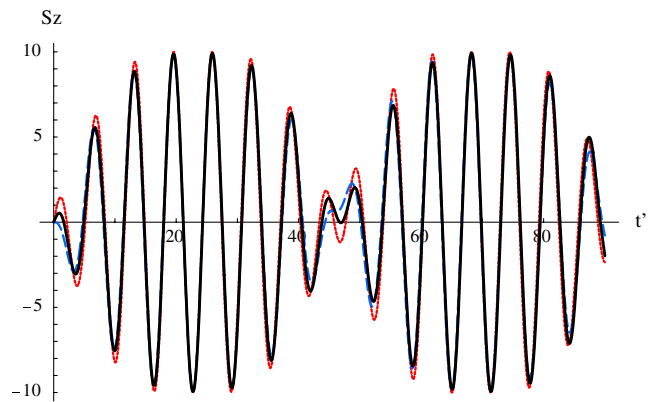


FIG. 3 (color online). Time dependence ($t' \equiv t\Delta/\hbar$) of the expectation value of the projection of the spin onto the anisotropy axis at $x = 0$, $S = 10$, $\omega_R = 0.1\omega$, and $\omega = 0.9(\Delta/\hbar)$. Dotted red line: Numerical result for the laboratory-frame Hamiltonian (19). Dashed blue line: Numerical result obtained with the use of the lattice-frame Hamiltonian (11). Solid black line: Analytical result obtained with the use of Eq. (16).

perturbation provided that $E \ll D$. At $\omega \ll (2S + 1)D/\hbar$ the dynamics of the wave function involves only a superposition of the $|\pm S\rangle$ states. As in the lattice-frame consideration, it is then convenient to project the Hamiltonian (18) onto these two states. In order to obtain such an effective two-state Hamiltonian that accounts for the tunnel splitting of the lowest energy states, one must apply perturbation theory for the degenerate states $|\pm S\rangle$ to the S th order [1]. This results in

$$\hat{h}_{\text{eff}} = -\frac{\Delta}{2} \{e^{2iS\delta\phi(x,t)}|S\rangle\langle -S| + e^{-2iS\delta\phi(x,t)}| -S\rangle\langle S|\}. \quad (19)$$

Here $\Delta = 8D(2S)![(S - 1)!]^{-2}(E/8D)^S$ is the tunnel splitting for the biaxial model in the absence of lattice distortions [12]. Numerical solution for $\langle \hat{S}_z \rangle$ that follows from Eq. (19), and its comparison with the analytical solution given by Eq. (16), are illustrated in Fig. 3. The beats discussed above are clearly seen in the figure.

We shall now discuss conditions under which the above effects can be observed. First, we notice that the Rabi oscillations of $\langle \hat{S}_z \rangle$ generated in a crystal of molecular magnets by ultrasound, contrary to the Rabi oscillations generated in a small crystal by an electromagnetic wave, will have a pronounced wave dependence on coordinates so that $\langle \hat{S}_z \rangle$ averaged over the wavelength of the sound λ will be zero. Consequently, measurements of the oscillations of $\langle \hat{S}_z \rangle$ should be done on the scale that is small compared to λ . Secondly, the decoherence rate should be lower than the Rabi frequencies involved. The lower bound on the decoherence due to spin-lattice interactions can be estimated as the spin-phonon relaxation rate. For the tunnel-split states with the splitting in the gigahertz range, it is in the kilohertz range [11]. For the rate of transitions between adjacent S_z states in molecular magnets, the prefactor in the observed Arrhenius relaxation law provides an estimate of order 10 MHz [13]. For the Rabi frequency at, e.g., $f = 3$ GHz, $u_0 = 1$ nm, $S = 10$, and $c_t = 10^3$ m/s, one obtains $\omega_R \sim 2$ GHz $\ll \omega = 2\pi f \sim 20$ GHz.

When Rabi oscillations are driven by the external acoustic wave, the latter should force the phase coherence upon the spin system. This should eliminate concern that phases of individual spins are decohered by dipolar fields. However, to provide the resonance condition, the broadening of the level splitting due to dipolar fields should be small compared to $\hbar\omega \sim \Delta$. If it is not, the tunnel splitting Δ should be increased by applying a transverse magnetic field. Mn-12 is, probably, not the best system, as it exhibits wide distribution of Δ due to large dipolar fields and internal disorder. Ni-4 where the resonant interaction of electromagnetic radiation with tunnel-split states was achieved in the gigahertz range [9] may be a promising

candidate for observing acoustic Rabi oscillations. Note that, in principle, surface acoustic waves of frequency as high as 100 GHz have been generated in experiment [14]. However, since $\omega_R \propto \omega^2$, raising ω significantly may eventually violate the condition $\omega_R \ll \omega$ under which our results were derived.

In conclusion, we have shown that transversal acoustic waves in the gigahertz range provide spin-rotation coupling that can be used to generate space-time Rabi oscillations in molecular magnets. When frequency of ultrasound ω equals the distance between tunnel-split spin states, the magnetization on the surface of the crystal oscillates as $\langle \hat{S}_z \rangle = S \cos(kx - \omega t) \sin[\omega_R t - k_R x]$, where $\omega_R = \omega^2 u_0 S / (2c_t)$ and $k_R = (\omega_R / \omega)k$, with u_0 and c_t being the amplitude and the speed of the sound, respectively.

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