Coherent strong-field coupling of a ferromagnetic nanomagnet with a photonic cavity

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ABSTRACT
We predict that strong coupling is feasible between photons and a ferromagnetic nanomagnet, due to exchange interactions that cause very large numbers of spins to coherently lock together with a significant increase in oscillator strength while still maintaining very long coherence times. The interaction of a ferromagnetic nanomagnet with a single photonic mode of a cavity is analyzed in a fully quantum-mechanical treatment. Exceptionally large quantum-coherent magnet-photon coupling with coupling terms in excess of several THz are predicted to be achievable in a spherical cavity of ~1 mm radius with a nanomagnet of ~100 nm radius and ferromagnet resonance frequency of ~200 GHz. This should substantially exceed the coupling observed in solids between orbital transitions and light. Eigenstates of the nanomagnet-photon system correspond to entangled states of spin orientation and photon number over 10^5 values of each quantum number. Initial coherent state of definite spin and photon number evolve dynamically to produce large coherent oscillations in the microwave power with exceptionally long dephasing times of few seconds. In addition to dephasing, several decoherence mechanisms including elementary excitation of magnons and crystalline magnetic anisotropy are investigated and shown to not substantially affect coherence up to room temperature. The optimal nanomagnet size is predicted to be just below the threshold for failure of the macrospin approximation.

Keywords: Strong Coupling, Coherence, Nanomagnet, Cavity Quantum Electrodynamics

1. INTRODUCTION
Modern cavity quantum electrodynamics (cavity-QED) provides the most fundamental machinery for calculating coherent properties in quantum mechanics. Even though experiments on atoms in cavities can be explained by elementary models, measurements still reveal intriguing subtleties of the influences of external couplings on coherent dynamics. These cavity-QED systems can then be used to implement schemes for quantum information technology. Further advances in experimental cavity-QED have been achieved in the past decade. Quantum nondemolition measurements with the realization of a universal quantum logic gate,^1,2 quantum nonlinear optics providing entry to a quantum regime for nonlinear optics,^3 atom-cavity microscopy resulting in sensitive measurements of an individual atom’s motion,^4,5 quantum state synthesis making the preparation of single-photon Fock states possible,^6,7 nonclassical correlations testing the theoretical models of entanglement and decoherence^8,9 are only some of the highlights of recent progress in cavity-QED.

One particularly important and useful scheme of cavity-QED is the strong coupling between light and matter. Strong coupling between light and electronic transitions^10–14 permits coherent transfer of quantum information between the two systems, as well as a host of exotic phenomena, including slow light,^15,16 lasing without population inversion,^17,18 and index enhancement via quantum coherence.^19,20 One of the most striking applications harnessing such a powerful resource of strong coupling via cavity-QED is the teleportation of an unknown qubit between two distant systems without physical transfer of the associated quantum-information carrier.^21

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Teleportation is already shown possible using coherent strong coupling between Rydberg atoms and cavity photons\textsuperscript{11, 22–26} due to its purely quantum nature. Therefore, the main idea for teleportation is indeed to harness the coherent atom-field interaction using cavity field modes.

Strong coupling between light and electronic transitions in solids has been challenging, because of shorter coherence time of orbital (electric dipole) transitions in solids compared to atoms. However, strong coupling in a single quantum dot-semiconductor microcavity system\textsuperscript{27} has been demonstrated with a coupling strength $\sim 80$ $\mu$eV, as well as some examples of strong coupling between a single exciton and a single photon in a semiconductor for several different cases.\textsuperscript{28–34} The strong coupling efforts also favor the orbital transitions over spin (magnetic) transitions, whose typical oscillator strengths are estimated\textsuperscript{35} to be smaller by a factor of the fine structure constant, $\sim 137$. However, recent investigations has revealed that paramagnetic spin systems in solids appear to be intrinsically more quantum coherent than orbital coherent states.\textsuperscript{36, 37} They also exhibit collective spin-photon effects such as superradiance\textsuperscript{38–40} including molecular magnets of $\sim 10$ spins.\textsuperscript{41}

On the other hand, strong coupling via spin transitions (magnetic) in ferromagnetic systems has been recently examined\textsuperscript{42} and it has been pointed out that the locking of the large number of constituent spins by the exchange interaction into a macrospin can cause a significant increase in coupling strength proportional to the square root of the number of exchange-locked spins in the absence of any cavity photons. Moreover, these systems still benefit from long coherence times similar to paramagnetic spin systems. They also exhibit superradiance leading to coupling strengths proportional to the total spin to the 3/2 power and therefore significantly boosting coupling when driven in the superradiance regime. Thus a single coupled ferromagnetic nanomagnet-photonic mode system provides an efficient method of strongly coupling electronic, magnetic, and photonic degrees of freedom.

We begin by describing the nanomagnet-cavity system and deriving the Hamiltonian of the system in a fully quantum mechanical treatment. Solutions for the eigenstates of the coupled system for different nanomagnet sizes are obtained by mapping the discrete system onto a continuum representation similar to a one-dimensional tight-binding model with a spatially-varying effective mass. Perturbations to the magnetic system such as magnetocrystalline anisotropy can be described as spatially-varying potentials for this one-dimensional tight-binding model. The time-evolution of coherent states is evaluated for the determination of coherent dynamics, and the source of dephasing is discussed. The optimal nanomagnet size is identified to be just below the threshold for failure of the macrospin approximation.

2. FORMALISM OF NANOMAGNET-CAVITY COUPLING

As shown schematically in Fig. 1, the oscillator is a spherical nanomagnet with a radius $r_0$ possessing a very large exchange-locked spin $S$ (consisting of $N$ electron spins) placed a distance $d$ from the center of the cavity for more efficient coupling to the cavity mode. Precession of the nanomagnet macrospin at a frequency $\omega$ resonant with the cavity is achieved by applying a uniform magnetic field $B_0$ along the $z$-axis of the cavity.

A nanomagnet acting as a macrospin, as seen experimentally in nanomagnet oscillators of roughly this size,\textsuperscript{43} has a magnetization

$$ M = \frac{\mu}{V} = -\frac{g_s \mu_B}{h V} S \Theta(r_0 - |r - d|), $$

(1)
in terms of the collective spin operator $S$ and the Heavyside step function $\Theta(x)$. The magnetization in Eq. (1) depends on the spin density of the nanomagnet. The coupling of the nanomagnet to the photonic mode (modal coupling) is the overlap of this magnetization with the cavity mode amplitude. For a nanomagnet that is small in size compared to the length scale of variations in the cavity mode strength, the coupling will be independent of the spin density and will only depend on the total spin. It is possible, however, to enhance the modal coupling through mode design, such as is common to enhance the interaction between gain media and an optical cavity in semiconductor lasers.\textsuperscript{44} For example, an optical field is strongly enhanced near a sharp metal object (used in tip-enhanced spectroscopy\textsuperscript{45}); a similar approach here could be used to strongly enhance the strength of the nanomagnet-cavity coupling.

The presence of the nanomagnet in the cavity, and its magnetization field, modifies the properties of the dynamic electromagnetic field in the cavity. The nanomagnet precesses in the static external magnetic field,
yielding a temporally-oscillating magnetization characterized by the precession frequency $\omega$. Thus the nanomagnet behaves as an oscillating source in the Maxwell equations. Introduction of the time dependence of the fields ($e^{i\omega t}$) into $H$, $E$, and $M$ produces the following Helmholtz wave equations,

\[ (\nabla^2 + k^2) (r \cdot H) = -iL \cdot (\nabla \times M), \]
\[ (\nabla^2 + k^2) (r \cdot E) = Z_0L \cdot M. \] (2)

From these the solutions of the transverse magnetic (TM) and electric modes (TE) can be obtained

\[ H = \sum_{l,m} \left[ \alpha^{(TM)}_{lm} f_l(kr)Y_{l,l,m}(\theta, \phi) - \frac{i}{k} \alpha^{(TE)}_{lm} \nabla \times g_l(kr)Y_{l,l,m}(\theta, \phi) \right], \]
\[ E = Z_0 \sum_{l,m} \left[ \frac{i}{k} \alpha^{(TM)}_{lm} \nabla \times f_l(kr)Y_{l,l,m}(\theta, \phi) + \alpha^{(TE)}_{lm} g_l(kr)Y_{l,l,m}(\theta, \phi) \right], \] (3)

where the vector spherical harmonics $Y_{l,l,m}$ are defined as $LY_{lm}(\theta, \phi)/\sqrt{l(l+1)}$, in terms of angular momentum operator of the field $L$ and spherical harmonics $Y_{lm}$. In the most general form, they are defined as

\[ Y_{j,l,m} = C(l,1;m,j,m)Y_{lm}\mathbf{e}_m \] (4)
in terms of Glebsch-Gordan coefficients and helicity basis vectors $\mathbf{e}_m$. The helicity basis vectors form a spherical tensor of rank 1, i.e. $\mathbf{e}_\pm = (\mathbf{\hat{x}} \pm i\mathbf{\hat{y}})/\sqrt{2}$, where $\mathbf{\hat{z}} = \mathbf{\hat{z}}$.

The $f_l(kr)$ and $g_l(kr)$ appearing in Eqs. (3) are the corresponding solutions for the radial part of each mode, $A^{(1)}(kr)h^{(1)}(kr) + A^{(2)}(kr)h^{(2)}(kr)$, in terms of the spherical Hankel functions. The coefficients $\alpha^{(TM)}_{lm}$ and $\alpha^{(TE)}_{lm}$, which specify the amounts of transverse magnetic and transverse electric multipole ($l, m$) field strengths, are

\[ \alpha^{(TM)}_{lm} = \frac{i k^3}{\sqrt{l(l+1)}} \int j_l(kr')Y^*_{lm}(\theta', \phi')L \cdot M d^3r', \]
\[ \alpha^{(TE)}_{lm} = \frac{-k^2}{\sqrt{l(l+1)}} \int j_l(kr')Y^*_{lm}(\theta', \phi')L \cdot (\nabla \times M) d^3r', \] (5)
where the volume integration is carried over the local sources. For a magnetic field applied in the $\hat{z}$ direction, the microwave emission of the nanomagnet is due to the oscillating components of the magnetization $M_{x,y}$ perpendicular to the radial direction (Fig. 1), and thus the cavity TE modes (magnetic field pointing in the radial direction) do not couple to the nanomagnet.

The total Hamiltonian of the system incorporates the magnetic $H$ and electric $E$ fields of the cavity and the magnetization $M$ of the nanomagnet;\textsuperscript{35}

$$\mathcal{H} = \frac{1}{2} \int \left( \mu_0 |H|^2 + \epsilon_0 |E|^2 + \mu_0 (H \cdot M) \right) \, d^3r. \quad (6)$$

The first two integrands on the right hand side of Eq. (6) correspond to the free field Hamiltonian, whereas the third integrand is the interaction Hamiltonian of the nanomagnet-cavity system.

### 3. COUPLING OF THE NANOMAGNET TO THE PHOTONIC CAVITY

The spin operators of the nanomagnet should be written in the same helicity basis as the photonic field,

$$S = \frac{1}{\sqrt{2}} (S_+ \hat{e}_+ - S_- \hat{e}_-) + S_0 \hat{e}_0, \quad (7)$$

in terms of the nanomagnet spin raising and lowering operators $S_\pm |l_s, m_s\rangle = \sqrt{(l_s \pm m_s)(l_s \pm m_s + 1)} |l_s, m_s \rangle$ in order to complete the quantization picture. Introduction of this total spin operator to Eq. (6), as well as replacing the field strength coefficients of the TM mode with the corresponding annihilation (creation) operators, yields a fully quantum Hamiltonian

$$\mathcal{H}_\gamma = \hbar \omega_\gamma \left( a_1^\dagger a_\gamma + \frac{1}{2} \right) - g \mu_B \Gamma_\gamma (a_\gamma S_+ + a_\gamma^\dagger S_-) + g \frac{\mu_B}{\hbar} B_0 S_z, \quad (8)$$

in which the spin interacts only with a single photon mode $\gamma$. Modes of higher $l$ would be out of resonance because of the cavity quantization, and energy non-conserving terms with negative helicity have been dropped (relying on the rotating wave approximation\textsuperscript{47}). The nanomagnet-photon coupling constant, $\Gamma_\gamma$, is found to be

$$\Gamma_\gamma = \frac{y_{1,\gamma}}{8 \hbar |j_{1,\gamma}|} \left[ 1 - \frac{l(l+1)}{y_{1,\gamma}^2} \right]^{-1/2} \sqrt{\frac{3 \hbar \omega_\gamma \mu_0}{\pi R^3}}, \quad (9)$$

where $y_{1,\gamma}$ is the $\gamma$-th zero of $|rj_{1}(kr)|$ satisfying the conditions for the field of TM mode at the cavity boundary. The mode frequency $\omega_\gamma$ is related to the radius of the cavity $R$ with $k_{1,\gamma} = \omega_{1,\gamma}/c = y_{1,\gamma}/R$.

The interaction with the uniform magnetic field $B_0$, introduced in Eq. (8), sets the cavity in resonance with the energy level splitting of nanomagnet spin states whenever the relation $\hbar \omega_\gamma = g \mu_B B_0$ is satisfied. Therefore, any spin flip up (down) process of the nanomagnet spins results in an absorption (emission) of a cavity photon in the case of exact resonance, e.g. an applied uniform magnetic field of $B_0 = 7$ T, corresponding to a precession of the macrospin with a frequency of $\sim 200$ GHz, will cause the nanomagnet spins to be in exact resonance with a cavity volume of $1.25$ mm$^3$. We assume the lowest TM mode of the cavity is in resonance with the spin-flip transitions of the nanomagnet, so as higher-energy modes will not be in resonance the subscript $\gamma$ will be omitted from Eq. (8) and Eq. (9).

The eigenstates of the nanomagnet, treated as a macrospin, are simultaneous eigenstates of the total spin operators $S^2$, and $S_z$ given by $|l_s, m_s\rangle$, where $|m_s\rangle \leq l_s \leq N/2$. Part of the macrospin approximation is the assumption that $l_s$ is fixed, and most likely it will be the maximal spin state $l_s = N/2$ due to additional energy requirement of any other $l_s \neq N/2$ subspace. The Hilbert space of $N$ independent spins should include the states of a macrospin corresponding to $l_s = N/2$. Therefore, the structure of these basis states is similar to those of the Dicke model\textsuperscript{48} for $N$ independent atomic spins, wherein $l_s$ is the cooperation number of the paramagnetic collection of spins. However, for a realistic nanomagnet, elements of the Hilbert space with $l_s \neq N/2$ are split off in energy due to the exchange interaction giving rise to extra mechanisms, i.e. elementary excitation of spin waves (magnons).
Since each magnon excitation reduces the total magnetic moment ($\mu \propto l_s$) of the nanomagnet in the amount of $2.21\mu_B$ for Fe, it is possible for the nanomagnet total spin angular momentum to start in a different $l_s$ subspace rather than the maximal $l_s = N/2$. This reduction in $l_s$ is less than 1% at room temperature for iron, suggesting that nanomagnet oscillators of approximately these sizes can be well-described by as having maximal spin at room temperature. The validity of the macrospin approximation relies on the effectiveness of the exchange-locking of the spins at room temperature. For the nanomagnets we consider here, consisting of up to $N = 10^9$ electron spins, the macrospin approximation is reasonable\(^{49}\) (although perhaps questionable at for the largest nanomagnet considered).

4. SOLUTIONS

The total excitation number $2\xi$, corresponding to the maximum number of photons $n$ in the cavity (when the nanomagnet is parallel to the static magnetic field), needs to be conserved by the Hamiltonian in Eq. (5). For an initial configuration of the macrospin pointing antiparallel to the static field $B_0$ and no photons in the cavity, $\xi = N/2$, the basis states of the spin-photon mode system $|n, m_s\rangle$ can be written as $|n, \xi - n\rangle$ or $|\xi - m_s, m_s\rangle$, so that the basis states are indexed either solely by photon number of the cavity ($n$), or by eigenvalue of $S_z$ ($m_s$). To proceed, we adopt the notation $|n, \xi - n\rangle$ and drop the redundant reference to the $m_s$, so the total Hamiltonian takes the form of

$$\mathcal{H} = \sum_{n=0}^{2\xi} E_0 |n\rangle\langle n| - \tau(n) [|n + 1\rangle\langle n| + |n\rangle\langle n + 1|],$$

in the Fock space, where the constant energy coefficient $E_0$ term and the coupling strength $\tau(n)$ are defined as

$$E_0 = \hbar \omega (\xi + 1)/2,$$

$$\tau(n) = \hbar \Gamma g \mu_B (n + 1) \sqrt{2\xi - n}.$$

In matrix form, the same Hamiltonian can be written as

$$\mathcal{H} = \begin{pmatrix}
E_0 & -\tau(0) & 0 & \cdots & 0 \\
-\tau(0) & E_0 & -\tau(1) & \cdots & 0 \\
0 & -\tau(1) & E_0 & \cdots & 0 \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
0 & 0 & \cdots & -\tau(2\xi - 1) & E_0
\end{pmatrix},$$

similar to the Hamiltonian matrix expected for a nearest-neighbor tight-binding model with a spatially-dependent mass. For $2\xi = N = 10^9$, the magnet-microwave mode coupling, $\tau(n)$, changes over a range of $33$ kHz - $1.3$ THz through all possible photon (spin) numbers. $\partial\tau(n)/\partial n = \tau'(n)$ acts like a driving force for a fictitious particle moving between sites labeled by photon number $n$, so $|0\rangle \rightarrow \ldots \rightarrow |n - 1\rangle \rightarrow |n\rangle \rightarrow |n + 1\rangle \rightarrow \ldots \rightarrow |2\xi\rangle$. The solutions $n_0$ of $\tau'(n)|n_0 = 0$ are equilibrium points in cavity photon number, and for this system there is one at $n_0 = (4\xi - 1)/3$. The coupling can also be expressed in terms of the collective spin number $m_s$ as $\tau(m_s) = \hbar \Gamma g \mu_B (\xi - m_s + 1) \sqrt{\xi + m_s}$, with an equilibrium point of $m_0 = (1 - \xi)/3$. For a system consisting of a very large number of spins ($\xi \gg 1$), the eigenfunctions of the Hamiltonian in Eq. (7) are expected to be centered about $n_0 = 4\xi/3$ as well as $m_0 = -\xi/3$.

For an initial state $|n, m_s\rangle$, if we are only interested in transitions which conserve energy and in which a photon is emitted, the rate of photon emission $R_n$ is proportional to $\sum_{\Psi} |\langle \Psi |a^\dagger S_- |n, m_s\rangle|^2$, where $|\Psi\rangle$ represents the possible final states of the system. Therefore, $R_n = A(n + 1)^2 (2\xi - n)$, or equivalently $R_n = A(\xi - m_s + 1)^2 (\xi + m_s)$. The factor $A$ can be identified as the Einstein $A$-coefficient by applying $R_n$ to a single spin pointing upward ($\xi = m_s = 1/2$) when the cavity has no photons ($n = 0$). Since $R_n$ reaches its maximum value of $4A(N/3)^3$ for the equilibrium point $m_0$ (or $n_0$) in the large spin limit, the equilibrium points $n_0$ and $m_0$ are the photon number and spin number, respectively where the nanomagnet-cavity system exhibits superradiance.\(^{48}\)
The eigenfunctions of the nanomagnet-cavity Hamiltonian given in Eq. (10) can be expanded as \( \psi_j = \sum_{\ell} \psi_j^{\ell} |n^{\ell}\rangle \). For a large-spin nanomagnet the eigenfunctions of the Hamiltonian can be found in the continuum limit, corresponding to replacing \( \psi_j^{\ell} \rightarrow \psi_j(x = n\xi) \) and keeping terms up to \( \mathcal{O}(\xi^3) \), yielding

\[
\tau(x) \frac{d^2 \psi_j(x)}{dx^2} + \frac{dr(x)}{dx} \frac{d\psi_j(x)}{dx} + \left(2\tau(x) - \frac{d\tau(x)}{dx} + \frac{1}{2} \frac{d^2\tau(x)}{dx^2} + E_j\right) \psi_j(x) = 0,
\]

with \( \psi_j(0) = \psi_j(2s_z) = 0 \). The lowest energy eigenvalues \( E_j \) and eigenfunctions \( \psi_j(x) \) of this differential equation, shown in Fig. 2(b), can be obtained in the WKB approximation from \( S(E_j) = (1/2\pi) \oint (E_j - V_c(x)/\tau(x))dx = j + \frac{1}{2} \), where the effective potential is \( V_c(x) = \tau'(x) - \tau'^2(x)/4\tau(x) - 2\tau(x) \). The ground state \( \psi(x) \) is extended over a broad range of \( x \) (and thus \( m_s \) and \( n \)) because \( V_c(x) \) is smoothly varying (due to large \( \xi \)); in the limit of large \( S \) and large \( \xi \), from the functional form of \( V_c(x) \), the width of the ground state can be determined from \( (\Delta n)^2/n_o \sim 1 \). Effective potential \( V_c(x) \) and the eigenfunctions calculated for a nanomagnet radius of roughly \( r_0 = 100 \text{ nm} \) consisting of \( N = 10^9 \) spins are shown in Fig. 2.

![Figure 2](image)

Figure 2. (color online) (a) The effective potential of the magnet-cavity system (for \( N = 10^9 \) spins) in the WKB approximation is shown with respect to cavity photon number \( n \) centered about the superradiance regime \( n_o \), (b) Eigenfunctions of the nanomagnet-cavity system as a function of photon number, \( n \), centered about \( n_o = 4\epsilon/3 = 6.6667 \times 10^8 \) for \( N = 10^9 \) spins: (i) ground state with a width of roughly \( 5 \times 10^3 \) photons (or equivalently spin quantum numbers \( m_s \)), (ii) \( 1^{st} \), (iii) \( 2^{nd} \), and (iv) \( 150^{th} \) excited states.

### 5. NANOMAGNET-CAVITY COHERENT DYNAMICS

#### 5.1 Coherent State Representation

A coherent state for the nanomagnet-cavity system can be written as a displaced nanomagnet-cavity ground state (by a photon number \( x_0 \) from the equilibrium point \( n_o \)) and the eigenfunctions of the nanomagnet-cavity system are complete and orthonormal, hence they serve as a suitable basis to expand any coherent state over,

\[
\phi(x,t) = \sum_{j=0}^{\infty} A_j e^{-iE_j t/\hbar} \psi_j(x).
\]

where the phase constants \( A_j \) are determined by

\[
A_j = \frac{1}{\sigma\sqrt{2\pi}} \int_{x_0}^{x_0+2\xi} \psi_j(x)e^{-(x-x_0)^2/2\sigma^2} dx.
\]

For four sizes of the nanomagnet, the coherent states shown in Fig. 3(a)(i)-(iv), are characterized by large oscillations over ranges of \( (2x_0=) \) 1780, 1.76 × 10^4, 1.76 × 10^5, and 5.34 × 10^5 photons with periods of \( T = 1.5 \text{ ms} \).
respectively, by using the same coherent state representation. Large oscillations of these quantities shown in Fig. 3(b) indicates the coherent energy exchange occurring back and forth between photons in the cavity and the spin states of the nanomagnets.

\begin{align}
\langle \Delta E_z \rangle &= \langle \phi(x,t) | \mu_z B_0 | \phi(x,t) \rangle, \\
\langle B_T \rangle &= \langle \phi(x,t) | \mathbf{H}_{TM}(d) | \phi(x,t) \rangle,
\end{align}

where $T = 150 \mu s$, $T = 15 \mu s$, and $T = 4.74 \mu s$, respectively. Summation over the first 150 eigenstates ($j_0 = 150$) extracted from WKB is sufficient enough to obtain convergence in the dynamical properties of these nanomagnets. The Zeeman energy of the nanomagnet and transverse magnetic field amplitude of the cavity at the nanomagnet’s location can also be evaluated from

Figure 3. (color online) (a) Amplitude of a coherent state for 3 different nanomagnet-photon systems consisting of (i) $N = 10^4$, (ii) $N = 10^5$, (iii) $N = 10^6$, and (iv) $N = 10^8$ spins are shown as a function of photon number $n$. The large oscillations of these coherent states occur about (i) $\eta_0 \sim 6666$ with a period of $T = 1.5 ms$, (ii) $\eta_0 \sim 6.66 \times 10^5$ with a period of $T = 150\mu s$, (iii) $\eta_0 \sim 6.66 \times 10^6$ with a period of $T = 15 \mu s$, and (iv) $\eta_0 \sim 6.66 \times 10^8$ with a period of $T = 4.74\mu s$ respectively; (b) Time evolution of the Zeeman energy of the nanomagnets (red, dashed) consisting of (i) $N = 10^4$, (ii) $N = 10^5$, (iii) $N = 10^6$, and (iv) $N = 10^8$ spins are shown in coherent state representation as well as the amplitude of the transverse magnetic mode of the cavity field (green, solid) at nanomagnet location $z = d$. 

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5.2 Dephasing of the Coherent State

The coherent properties of the nanomagnet-cavity system will also depend on the dephasing of the coherent state $\phi(x, t)$, due to inhomogeneity of the coupling $\tau(n)$ in Eq. (11). The dephasing time of the nanomagnet-cavity coherent state can be extracted by a Gaussian fit to the peak values of the autocorrelation function between a coherent state at time $t$ and its initial state at $t = 0$,

$$P(t) = \langle \phi(x, t)|\phi(x, 0) \rangle^2,$$

$$= \sum_{j=0}^{\infty} |A_j|^2 e^{iE_j t/\hbar},$$

whereas each peak (inset of Fig. 4) is representing the revival amount of the coherent state after every successful period $T$ of oscillation. Exceptionally long dephasing time of order seconds are shown in Fig. 4. As the nanomagnet size gets larger the change in $\tau(n)$ with $n$ becomes smoother and smoother, leading to longer dephasing times.

![Figure 4](image_url)

Figure 4. (color online) Dephasing time of the coherent state for nanomagnet-photon systems of (a) $N = 10^4$ and (b) $N = 10^9$ spins (or equivalently photons) obtained by a Gaussian fit to the peak values of the dephasing functions (insets) at successive time intervals. Each peak value represents the amount of correlation after every full period $T$ of oscillation.

Although this treatment is for zero temperature, the coherent properties of the nanomagnet-photon system should persist to as high a temperature (and over as long a timescale) as the macrospin description remains reliable. We have assumed an infinite $Q$ for the cavity, so the decoherence of the system is expected to be determined by photon leakage from the cavity, rather than these exceptionally long calculated times. Furthermore, the elementary spin excitations (magnons) would not directly affect the dephasing of the system, for magnons preserve the spin quantum number $m_s$, requiring an up spin to flip down for every down spin flipping up. In realistic nanomagnets, spin-lattice coupling of $m_s$ to phonons through spin-orbit coupling will cause a cutoff of the dephasing times shown in Fig. 4. For spheres of yttrium iron garnet (YIG) at low temperature this spin-lattice time is several $\mu$s. Therefore, observation of a full oscillation cycle should be possible for nanomagnets with a radius of 50 nm or larger. On the other hand, the times at room temperature in YIG ($\sim 200$) ns and iron ($\sim 20$ ns) are too small to observe a full oscillation. However, coherent dynamics corresponding to a portion of the oscillation involving $\sim 24$ photons/ns, or $\sim 470$ photons for iron and 4700 photons for YIG should be still observable for the nanomagnet with radius $r_0 = 50$ nm. The modal coupling can be boosted using approaches such as tip-enhancement of the optical field, and far stronger coupling can be achieved even for a small nanomagnet. Guided by estimates from tip-enhanced Raman spectroscopy, the intensity of the mode at the nanomagnet’s position could be increased by $10^2 - 10^3$, leading to enhancements of the oscillation frequency of order $10 - 10^3$.

5.3 Crystalline Magnetic Anisotropy

Other deviations from ideality are also investigated for the nanomagnet, such as the spin dependent cubic crystalline magnetic anisotropy (CMA). The CMA of iron is given by

$$E_{CMA} = U_1 (\kappa_1^2 \kappa_2^2 + \kappa_2^2 \kappa_3^2 + \kappa_1^2 \kappa_3^2) + U_2 \kappa_1^2 \kappa_2^2 \kappa_3^2,$$

(19)
where $U_1 = 4.2 \times 10^5 \text{erg/cm}^3$ and $U_2 = 1.5 \times 10^5 \text{erg/cm}^3$ are the cubic anisotropy constants for iron at room temperature and an arbitrary magnetization direction is defined by the directional cosines $\kappa_1, \kappa_2, \kappa_3$ referred to the cube edges. Since the nanomagnet is a sphere, shape anisotropy is not relevant. In the case of a cubic crystal whose easy axis is aligned along the body diagonal, $E_{CMA}$ energy depends on the orientation of the nanomagnet spin $S$, defined by $\kappa_i$.

The CMA of iron causes a detuning of the energy spacing for different spin orientations from the resonant frequency of the cavity, along with a dispersion in that spacing. The uniform detuning, corresponding to a shift in the precession frequency of the nanomagnet, can be compensated for with a slight adjustment in the applied magnetic field. The dispersion, however causes a variable detuning of roughly 200 neV, 13 neV, 1.3 neV, and 0.3 neV of the $E_0$ in Eq. (10) over the range of oscillation shown in Fig. 3(a)(i)-(iv), respectively. For the smallest nanomagnets the effect of CMA dominates over the coupling between the photons and the spin. For example, for a nanomagnet radius of 2 nm consisting of $10^4$ total spins, the CMA is significantly larger than the magnet-photon coupling strength $\tau(n) (~5.3 \text{neV})$ in Eq. (11). Therefore the CMA will cause the eigenstates to localize in photon and spin number, producing rapid decoherence for a coherent state. We note that this observation largely rules out the possibility of observing these coherent oscillations in a single molecular magnet, for the spins of these molecules are considerably smaller than the spin of the nanomagnet considered above. However, this detuning is much smaller than the magnet-photon coupling strength of other nanomagnet sizes (10 nm, 50 nm, and 100 nm in radii) as shown in Tab. 1 and therefore will not destroy the coherent oscillations for them, although it may still limit the dephasing times to shorter than that shown in Fig. 4(ii)-(iv).

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<thead>
<tr>
<th>$r_0$</th>
<th>2.3 nm</th>
<th>11 nm</th>
<th>50 nm</th>
<th>100 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>(N)</td>
<td>(10^4)</td>
<td>(10^6)</td>
<td>(10^8)</td>
<td>(10^9)</td>
</tr>
<tr>
<td>(E_{CMA})</td>
<td>200 neV</td>
<td>13 neV</td>
<td>1.3 neV</td>
<td>0.3 neV</td>
</tr>
<tr>
<td>(\tau(n_0))</td>
<td>5.3 neV</td>
<td>5.3 (\mu)eV</td>
<td>5.3 meV</td>
<td>0.16 eV</td>
</tr>
</tbody>
</table>

Table 1. Crystalline magnetic anisotropy energies $E_{CMA}$ for different nanomagnet sizes (with radii $r_0$ and consisting of $N$ spins) are shown in comparison with the magnet-photon coupling strength at the superradiance regime ($\tau(n_0)$).

6. CONCLUSION

We have examined the strong-field interactions between nanomagnets of radii up to 100 nm and a spherical microcavity roughly 1 mm$^3$ in volume in the presence of a static magnetic field of 7 T in magnitude. Our results demonstrate that the interaction Hamiltonian contains magnet-microwave mode coupling terms that can exceed several THz, indicating that strong-field coupling between magnets and light is possible, and should substantially exceed the coupling observed in solids between orbital transitions and light. These strong-field effects should be observable in the nanomagnet-cavity dynamics. Furthermore, the coherent states of our spin-photon coupling around the superradiance regime are characterized by large oscillations in photon number $n$ of the cavity (or equivalently the collective spin number $m_s$ of the nanomagnet) with exceptionally long dephasing times of few seconds. The effects of magnons have been also considered and shown to not substantially modify these results up to room temperature. Moreover, the coherent dynamics of a coupled photonic cavity and a nanomagnet is explored as a function of nanomagnet size. For sufficiently strong coupling, eigenstates involving highly entangled photon and spin states are found, which can be combined to create coherent states. As the size of the nanomagnet increases its coupling to the photonic mode also monotonically increases, as well as the number of photon and spin states involved in the systems eigenstates. For small nanomagnets the crystalline magnetic anisotropy of the magnet strongly localized the eigenstates in photon and spin number, quenching the potential for coherent states. For a sufficiently large nanomagnet the macrospin approximation breaks down and different domains of the nanomagnet may couple separately to the photonic mode. Thus the optimal nanomagnet size is predicted to be just below the threshold for failure of the macrospin approximation. Therefore, the coherent dynamics is expected to be observable for realistic nanomagnets with radii from 10 - 100 nm. Approaches to enhance the coupling, such as using a metal tip to enhance the optical field, have been also proposed. The realization and development of quantum information technology and understanding of quantum coherence using this concept of strong coupling between light and spins may greatly advance the field. Therefore, future work shall investigate...
how to use the strong coupling features described here to transfer coherently states of the electronic system to the photonic one, and back again. A particularly interesting direction will be to consider the effect of active nanomagnetic systems, such as those demonstrated to be coherently driven by electrical spin currents,43,54–58 on the optical state of the cavity.

ACKNOWLEDGMENTS

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REFERENCES


