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# Photon and spin dependence of the resonance line shape in the strong coupling regime

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## Abstract

We study the quantum dynamics of a spin ensemble coupled to cavity photons. Recently, related experimental results have been reported, showing the existence of the strong coupling regime in such systems. We study the eigenenergy distribution of the multi-spin system (following the Tavis–Cummings model) which shows a peculiar structure as a function of the number of cavity photons and of spins. We study how this structure causes changes in the spectrum of the admittance in the linear response theory, and also in the frequency dependence of the excited quantities in the stationary state under a probing field. In particular, we investigate how the structure of the higher excited energy levels changes the spectrum from a double-peak structure (the so-called vacuum-field Rabi splitting) to a single-peak structure. We also point out that the spin dynamics in the region of the double-peak structure corresponds to recent experiments using cavity ringing, while in the region of the single-peak structure, it corresponds to the coherent Rabi oscillation in a driving electromagnetic field. Using a standard Lindblad-type mechanism, we study the effect of dissipation on the line width and separation in the computed spectra. In particular, we study the relaxation of the total spin in the general case of a spin ensemble in which the total spin of the system is not specified. The theoretical results are correlated with experimental evidence of the strong coupling regime, achieved with a spin-1/2 ensemble.

(Some figures may appear in colour only in the online journal)

## 1. Introduction

Quantum mechanics principles provide the fundamental basis for the development of future technologies which will integrate quantum processors with quantum memories. Electromagnetic impulses, like photons, can provide the link between the

processing of quantum algorithms and temporary storage between computation steps. Namely, quantum information can be transferred by photons into a solid-state memory able to preserve the quantum aspects of the information for a certain time. Initially, the coupling between atoms and resonators was first studied in atomic physics, since in the absence of

significant atom–atom interaction, the atomic levels have large lifetimes compared to their solid-state counterparts. In such a situation, the transfer of quanta between atoms and resonators can be observed.

The coupling between atoms and a standing electromagnetic wave was introduced by Tavis and Cummings [1, 2] in the 1960s, and calculated to increase with the square root of the number of atoms. This phenomenon has been observed for an ensemble of atoms [3, 4] and single atoms [5, 6] in cavity quantum electrodynamics experiments (cQED), in which photons and the atoms interact in a cavity. In more recent years, the use of a superconducting resonator with trapped molecules above it has been proposed [7] to implement cQED experiments on a chip. These initial studies made use of the electric-field component of an electromagnetic field to coherently exchange photons between the field and the system under study.

In solid-state systems, magnetic two-level systems (like up and down spins) can have significantly longer coherence times compared to electrical analogues. It is therefore desirable to implement the cQED techniques developed for electrical coupling to magnetic coupling, although the magnetic-field component of an electromagnetic field offers much less coupling. This aspect is important for the implementation of quantum computing on a chip, using solid-state materials, in which quantum information can be rapidly lost to the neighbouring environment. Dipolar interactions between spins in such systems are a major cause of information loss, since it affects the spin coherence lifetime. But these processes can be reduced by diluting the spins in the host solid. Magnetic coupling between the electromagnetic field in the cavity and a spin ensemble has been demonstrated theoretically [8] and experimentally [9–12], in the regime called strong coupling. This case requires a coupling strength larger than both the cavity’s photon decay rate and the rate at which the spin loses its quantum state information. Aside coupling to a cavity, recent studies [13, 14] show that diluted spin systems can be coupled with and exchange information with superconducting qubits, making them suitable quantum memories.

The spin–photon coupling can be modelled by the Jaynes–Cummings (for one spin) or the Tavis–Cummings model (for multi-spin), and it has been shown that the photon absorption or emission spectrum has a pair of peaks, called ‘vacuum-field Rabi splitting’ [15]. We study how the energy structure changes with the number of photons in the cavity, investigate how the change causes the spectrum of the admittance in the linear response theory, and also study the frequency dependence of the excited quantities in the stationary state under a probing field.

When the number of photons is much smaller than the number of spins, the spectrum shows the quantum-mechanical double-peak structure, while it shows a single peak corresponding to the magnetic resonance in a driving oscillatory field when the number of photons is much larger than the number of spins.

We also point out that the spin dynamics in the region of the double-peak structure corresponds to our experiments using cavity ringing (see also [9]) showing vacuum-field Rabi

splitting. On the other hand, the spin–photon system can show coherent Rabi oscillation, representing spin dynamics under a driving classical electromagnetic field, in the region of the single-peak structure. Between these two regions, namely when the number of spins and the number of photon in the cavity are similar, chaotic spin dynamics appears.

We also study the line shape of the spectrum of photon absorption and emission in dissipative environments. We point out that the line shape is affected by several conditions: how many spins are involved, how strongly the system is driven, how strong the dissipation is, what types of dissipation exist and whether we consider the linear spectrum or the spectrum of the nonlinear steady state under a driving force.

## 2. Model

### 2.1. Hamiltonian

As the system Hamiltonian we adopt the Tavis–Cummings model (a generalized Jaynes–Cummings model)  $\mathcal{H}_0$  with a driving force  $\xi$  [1, 2, 16]:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_\xi, \quad (1)$$

where

$$\mathcal{H}_0 = \omega_0 a^\dagger a + \omega_s \left( \sum_i^N S_i^z + N/2 \right) - g \left( a^\dagger \sum_i^N S_i^- + a \sum_i^N S_i^+ \right) \quad (2)$$

$$\mathcal{H}_\xi = \xi (a^\dagger e^{-i\omega t} + a e^{i\omega t}). \quad (3)$$

Here  $\{S_i\}$  denotes a set of two-level systems, and we call them ‘spin’ hereafter. Each is represented by the operator for a spin of  $S = 1/2$ . Here,  $\omega_0$  is the eigenfrequency of the cavity,  $\omega_s$  is the energy gap of a two-level system and  $\omega$  is the frequency of the driving force. Throughout the paper, we put  $\hbar = 1$ . Here the system contains  $N$  two-level systems. The dimension of the Hilbert space of the spin system is  $2^N$ . First, we consider the case that only the uniform mode (i.e. the uniform sum of spins of the system)

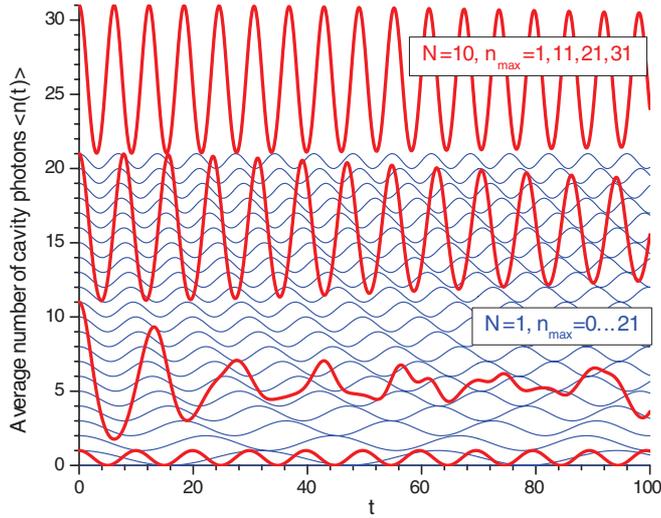
$$S = \sum_{i=1}^N S_i \quad (4)$$

couples to the cavity, and thus the total spin of the system is conserved. (We will study the case in which the total spin can be changed in section 5.) Therefore, the Hamiltonian is block-diagonalized with the total spin. The total spin is  $S = N/2$  when the number of spins is  $N$ . In the system, the sum of the photon number  $n$  and the magnetization  $M = \sum_{i=1}^N S_i^z$ ,

$$C = n + M, \quad (5)$$

is conserved. Thus, the Hamiltonian is further block-diagonalized with this quantity. To characterize the block, we introduce a quantity  $n_{\max} = C + S$  which is the number of photons when all the spins are down, i.e. the state of  $M = -S$ . The size of the sub-block increases from 1 to  $N + 1$  where  $n_{\max}$  is equal to  $2S + 1 (= N + 1)$ . For larger photon numbers,





**Figure 2.** Rabi oscillations of the average number of photons, for various values of the photon number. The thin blue curves show those for  $N = 1$  and  $n_{\max}$  from 0 to 21 (starting from the bottom). The bold red curves are for  $N = 10$  and  $n_{\max} = 1, 11, 21$  and 31, starting from the bottom.

We also give the photon number dependence of the energy and of the Rabi frequencies for the case  $N = 10$  in figure 1(b). Here, the Rabi frequencies are multiplied by 50 for increased clarity. Note that the Rabi frequencies have a larger distribution when  $N \simeq n$ .

### 2.2. Photon number dependence of the Rabi oscillation

In figure 2, we depict the Rabi oscillations versus time. The thin lines show the Rabi oscillations for the case of  $N = 1$ , i.e. the case of the Jaynes–Cummings model, where the Rabi oscillations in each sub-block are a sinusoidal curve with a single frequency  $\omega = 2g\sqrt{n_{\max}}$ . The bold lines show the Rabi oscillations for the case of  $N = 10$ . We find a simple oscillation for a small value ( $n_{\max} = 1$ ), which corresponds to the vacuum-field Rabi splitting. We also find a simple oscillation for  $n_{\max} = 21$  in which the photon degree of freedom behaves as a classical field, and the Rabi frequencies take values close to  $\omega = 2g|\langle a \rangle|$ . This oscillation corresponds to that of single spin in a driving field. If we look at the graph more carefully, we find that the amplitude decreases slowly in the case of  $n_{\max} = 21$ , which is due to a distribution of the frequencies. In the case of  $n_{\text{photon}} = 31$ , the oscillation is almost perfect sinusoidal within the observed time because the distribution is small. For larger values of  $n_{\max}$ , the oscillation looks perfect. Thus, we find two types of Rabi oscillations. The Rabi oscillation for small values of  $n_{\max}$  corresponds to the cavity ringing [3, 9], while that for large  $n_{\max}$  corresponds to the coherent Rabi oscillation which is found if the system is driven by an electromagnetic field [17, 18].

Between them, at  $n_{\text{photon}} \sim N = 10$ , we find chaotic behaviour which is attributed to the wide distribution of Rabi frequencies. This change between the two regions can be used to estimate how many spins contribute to the phenomena.

### 2.3. Dynamics in a dissipative environment

To study the dynamics of the system, we adopt a simple Lindblad-type master equation of the density matrix of the system  $\rho$  with damping terms [19–21]:

$$\begin{aligned} \frac{d}{dt}\rho = & \frac{1}{i}[\mathcal{H}, \rho] - \kappa(a^\dagger a \rho + \rho a^\dagger a - 2a\rho a^\dagger) \\ & - \gamma_{xy}(S^+ S^- \rho + \rho S^+ S^- - 2S^- \rho S^+) \\ & - \gamma_z((S^z)^2 \rho + \rho (S^z)^2 - 2S^z \rho S^z). \end{aligned} \quad (12)$$

The first term describes the coherent dynamics. In the next three terms, the relaxation effect is introduced by a conventional Lindblad form. The second term shows the cavity decay rate  $\kappa$ , and the last two terms give the spin dephasing and relaxation, respectively. In general, there is an additional pumping term due to the environment. This direct process could excite spins and thus generate emitted photons. But since the number of resonant bosons is generally low, we neglect these direct processes. Namely, we assume that the photon escapes from the system and is not created by the environment. We assume that the spin relaxation and dephasing are due only to energy relaxation processes, described here by constant values of  $\kappa$ ,  $\gamma_{xy}$  and  $\gamma_z$ .

For the electron spin resonance (ESR) line shape in the case of linear response, we need a distribution  $\rho(0)$ . The initial state, at the end of the pumping pulse, is an excited state. In order to express the degree of excitation, we used a steady-state distribution function at a temperature  $T$ :

$$\rho(0) = \frac{1}{Z} e^{-\mathcal{H}/k_B T}, \quad Z = \text{Tr} e^{-\mathcal{H}/k_B T}. \quad (13)$$

Because the system is not in equilibrium, in this approximation the temperature  $T$  can be regarded as a parameter describing the degree of freedom. A more extended treatment of the temperature effects has been formulated in Uchiyama’s paper [22], but here we take the present approximations.

### 3. ESR spectrum

The generalized susceptibility is given by the Kubo formula [19]

$$\chi(\omega) = \text{Tr} \int_0^\infty a^\dagger[a, \rho](t) e^{-i\omega t} dt = \text{Tr} a^\dagger \rho_a[\omega]. \quad (14)$$

Here, the last term is derived as follows [22]. Starting from the general form of the Kubo formula

$$\chi_{BA} = \lim_{\epsilon \rightarrow +0} \frac{i}{\hbar} \int_0^\infty dt e^{-i\omega t - \epsilon t} \text{Tr}[B(t), A]W, \quad (15)$$

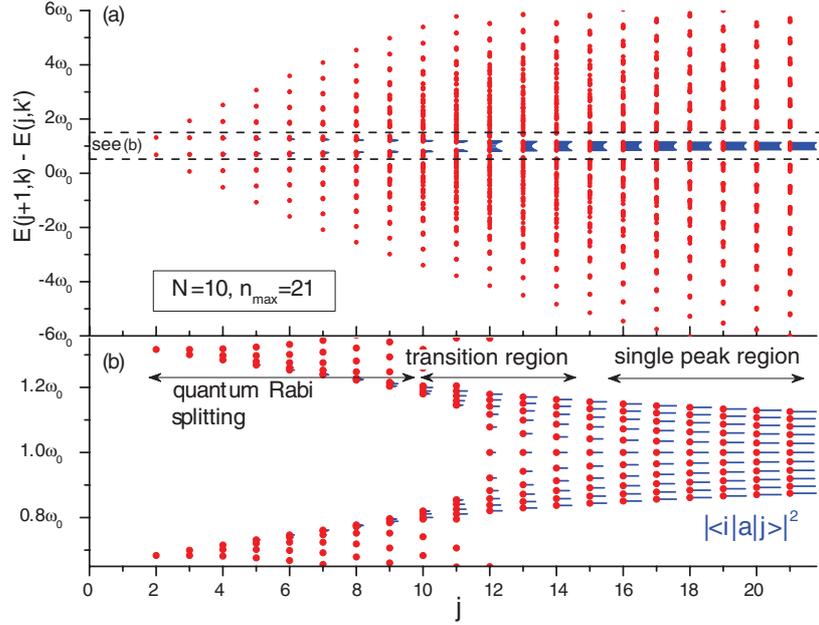
where  $W$  is the stationary density matrix of the system, we use the following identities:

$$\begin{aligned} \text{Tr}[B(t), A]W &= \text{Tr}(B(t)AW - AB(t)W) \\ \text{and } B(t) &= e^{iHt} B e^{-iHt}. \end{aligned} \quad (16)$$

Using trace properties regarding cyclic permutations, this leads to

$$\begin{aligned} \text{Tr}[B(t), A]W &= \text{Tr}(B e^{-iHt} A W e^{iHt} - B e^{-iHt} W A e^{iHt}) \\ &= \text{Tr}(B e^{-iLt} [A, W]), \end{aligned} \quad (17)$$

where  $e^{-iLt} X$  is the time evolution of operator  $X$  ( $\equiv e^{iHt} X e^{-iHt}$ ). Here we consider the driving force to be



**Figure 3.** (a) Resonance frequencies as a function of  $n_{\max}$ , for  $N = 10$ . The bars denote the strength of the matrix elements of photon absorption for the resonance  $|\langle i|a|j \rangle|^2$ . (b) Zoom for frequencies around  $\omega_0$ . Only the resonances  $\Delta E \sim \omega_0$  have visible transition probability. One distinguishes three regions: the quantum region showing Rabi splittings, the transition region and the classical region where one observes one single ESR peak.

$\xi a^\dagger e^{-i\omega t}$  and analyse the response of the operator  $a$ . Therefore, we have  $A = a$ ,  $B = a^\dagger$  and  $\rho_a[\omega]$  is the transformation of  $\text{Tr} e^{-i\mathcal{L}t}[A, W]$ . With the exception of the notation  $[a, \rho(t)](t)$ , equation (14) is the standard Kubo formula for photon absorption. In the same equation, we used the following notations:

$$[a, \rho](t) = e^{-i\mathcal{L}t}[a, \rho(0)], \quad (18)$$

where  $e^{-i\mathcal{L}t}$  denotes the time evolution of the density matrix (12) and

$$\rho_a[\omega] = \int_0^\infty [a, \rho](t) e^{-i\omega t} dt. \quad (19)$$

Here  $a^\dagger$  is a time-independent operator. The notation  $[a, \rho](t)$  means  $e^{-iHt}[a, \rho]e^{-iHt}$ , where  $a$  and  $\rho$  are the time-independent operators. Thus,  $a^\dagger$  can be put out of the integral of equation (14).

For the case of pure quantum dynamics, resonance frequencies of the linear response are given by

$$\omega_{\text{LR}}(n_{\max}, k, k') = E(n_{\max} + 1, k') - E(n_{\max}, k), \quad (20)$$

where

$$k = 1, \dots, k_{\max}(n_{\max}), \quad k = 1, \dots, k_{\max}(n_{\max} + 1),$$

at which the spectrum has delta-function peaks. The amplitude of each resonance is given by

$$I(\omega) = \frac{\pi (e^{-\beta E(n_{\max}, k)} - e^{-\beta E(n_{\max} + 1, k')})}{Z} \times |\langle n_{\max} + 1, k' | a^\dagger | n_{\max}, k \rangle|^2 \times \delta \left( \omega - \frac{E(n_{\max} + 1, k') - E(n_{\max}, k)}{\hbar} \right), \quad (21)$$

where  $Z = \sum_{n_{\max}=0} \sum_k e^{-\beta E(n_{\max}, k)}$ .

We give the distribution of the linear response resonant frequency  $\{\omega_{\text{LR}}\}$  for the case with  $N = 10$  for each value of  $n_{\max}$  in figure 3. Because the interaction is described by  $a^\dagger S^- + a S^+$ , the states with  $\Delta M = 1$  (and thus  $\Delta E \simeq \omega_0$ ) can be connected directly, while the other resonances are realized via higher order processes. Consequently, the transition with  $\omega_{\text{LR}} \simeq \omega_0$  has a large matrix element, while for the other resonances, the matrix elements are small and not visible in figure 3.

In the large  $n_{\max}$  limit, we may use the semi-classical picture for the electromagnetic field as  $a \rightarrow |a| e^{i\omega t}$ , and then the standard interaction Hamiltonian shows a rotating external field  $|a|(S^+ e^{i\omega t} + S^- e^{-i\omega t})$ . In this case, the resonance frequency is equal to  $\omega_S$ , as expected for the classical ESR case. Thus, figure 3 gives the transition from the quantum ringing (vacuum-field Rabi oscillation) region to the classical field region as a function of the photon number.

For the dissipative environments, we adopt the quantum master equation (12) instead of the pure quantum dynamics. Because  $[a, \rho](t)$  follows the equation of motion (12) [22], we use its Fourier–Laplace transformation:

$$i\omega \rho_a[\omega] - \rho_a(0) = \frac{1}{i\hbar} [\mathcal{H}_S, \rho_a[\omega]] - \kappa (Z^\dagger Z \rho_a[\omega] - 2Z \rho_a[\omega] Z^\dagger + \rho_a[\omega] Z^\dagger Z), \quad (22)$$

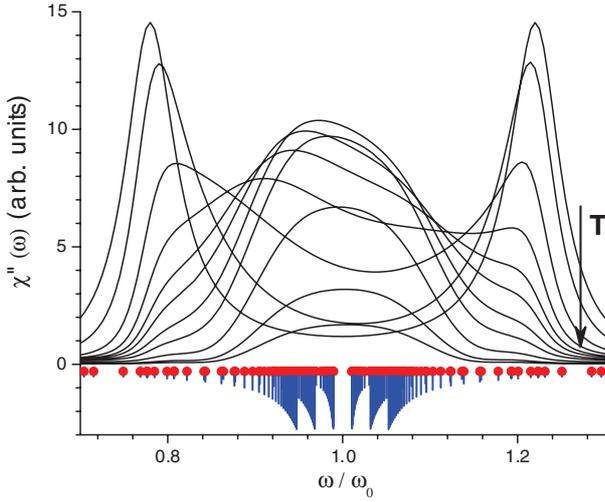
where  $Z$  denotes  $a$ ,  $S^-$  or  $S^z$  in equation (12), with

$$\rho_a(0) = [a, \rho^{\text{eq}}(T_{\text{system}})], \quad (23)$$

where  $T_{\text{system}}$  is the temperature of the system (cavity).

Equation (22) is a set of linear equations of components of  $\rho_a$  with the inhomogeneous term  $\rho_a(0)$ :

$$\rho_a(0) = i\omega \rho_a[\omega] + \frac{1}{i\hbar} [\mathcal{H}_S, \rho_a[\omega]] + \kappa (Z^\dagger Z \rho_a[\omega] - 2Z \rho_a[\omega] Z^\dagger + \rho_a[\omega] Z^\dagger Z). \quad (24)$$



**Figure 4.** Complex admittance for the case  $N = 5$  and  $n_{\max} = 21$ , with  $g = 0.1$ ,  $\kappa = 0.01$ ,  $\gamma_{xy} = 0.01$  and  $\gamma_z = 0.01$ . Here we plot data for  $T = 0.1, 1, 2, 3, 4, 5, 7, 10, 20, 50, 100$ , in increasing order as shown by the arrow. The line shape changes from a double peak to a single peak as the temperature increases. Below the axis, we plot the positions of the resonant frequencies (dots); the bar below each dot denotes the strength of the matrix element of the resonance, as in figure 3.

The explicit form of the equations for each matrix element ( $ij$ ) is given by

$$\begin{aligned} \rho_a(0)_{ij} &= i\omega\rho_a[\omega]_{ij} \\ &+ i\frac{1}{\hbar}\sum_k((\mathcal{H}_s)_{ik}\rho_a[\omega]_{kj} - \rho_a[\omega]_{ik}(\mathcal{H}_s)_{kj}) \\ &+ \kappa\left(\sum_k\sum_n Z_{ik}^\dagger Z_{kn}\rho_a[\omega]_{nj} \right. \\ &\left. - 2Z_{ik}\rho_a[\omega]_{kn}Z_{nj}^\dagger + \rho_a[\omega]_{ik}Z_{kn}^\dagger Z_{nj}\right). \end{aligned} \quad (25)$$

The ESR spectrum is obtained by the solution  $\rho_a[\omega]$  of this equation for values of  $\omega$  as

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega) = \frac{i}{\hbar}\text{Tr}a^\dagger\rho_a[\omega]. \quad (26)$$

In figure 4, we depict an example of temperature dependence of  $\chi(\omega)$  for  $N = 5$ , where the maximum value of  $n_{\max}$  is 25. The parameters  $g = 0.1$ ,  $\kappa = 0.01$ ,  $\gamma_{xy} = 0.01$  and  $\gamma_z = 0.01$  are used. There we find that  $\chi(\omega)$  has two peaks which correspond to the vacuum-field Rabi splitting [15] at a low temperature ( $T = 0.01$ ). As the temperature increases, the resonances between excited states start to contribute and a single peak is formed.

#### 4. Non-equilibrium distribution of photons

So far, we have assumed that the photons in the cavity are in equilibrium at a certain temperature. But if we excite the cavity from outside by a finite driving force  $\xi$ , the distribution of photons can have a non-equilibrium form, as studied by Bishop *et al* [16]. Also, nonlinear processes due to  $(a^\dagger)^k$ ,  $k > 1$ , can

contribute. The frequency of the nonlinear resonance is given by

$$\left| \omega_{\text{NL}}(n_{\max}, k) = \frac{E(n, k) - E(n', k)}{n - n'} \right|, \quad (27)$$

$$k = 1, \dots, k_{\max}(n_{\max}).$$

##### 4.1. Spectrum of a heterodyne measurement

Under the driving force  $\xi$ , the system reaches a stationary state  $\rho_{\text{st}}$  which is obtained as a stationary solution of (12). However, Hamiltonian (3) is time dependent and we cannot simply put  $d\rho/dt = 0$ . Thus, we use a rotating frame to remove the time dependence  $e^{\pm i\omega t}$ :

$$\mathcal{H}' = U\mathcal{H}U^{-1}, \quad U = \exp(i\omega t(a^\dagger a + S^z)). \quad (28)$$

This transforms the quantities as

$$Ua^\dagger U^{-1} = a^\dagger e^{i\omega t}, \quad US^- U^{-1} = S^- e^{-i\omega t} \quad (29)$$

and

$$\begin{aligned} \frac{dU\rho U^{-1}}{dt} &= U\frac{d\rho}{dt}U^{-1} + \frac{dU}{dt}\rho U^{-1} + dU\rho\frac{dU^{-1}}{dt} \\ &= U\frac{d\rho}{dt}U^{-1} + i\omega((a^\dagger a + S^z)U\rho U^{-1} \\ &\quad - U\rho U^{-1}(a^\dagger a + S^z)). \end{aligned} \quad (30)$$

Therefore, with this change, the equation of motion for  $\rho' = U\rho U^{-1}$  is given by (12) with the following form of the Hamiltonian:

$$\begin{aligned} \mathcal{H}' &= \omega_0 a^\dagger a + \omega_s \left( \sum_i^N S_i^z + N/2 \right) \\ &- g \left( a^\dagger \sum_i^N S_i^- + a \sum_i^N S_i^+ \right) \\ &+ \xi(a^\dagger + a) - \omega(a^\dagger a + S^z). \end{aligned} \quad (31)$$

Now, we consider the dynamics in this frame, and obtain a stationary state by setting  $d\rho/dt = 0$ :

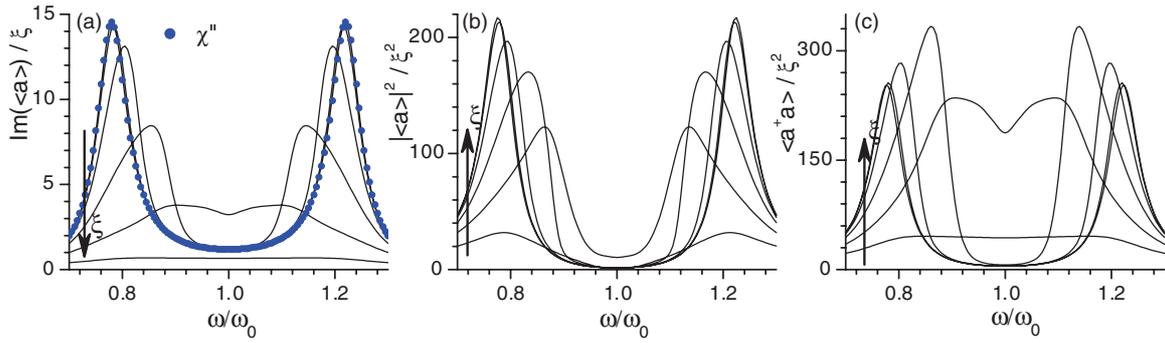
$$\begin{aligned} \frac{1}{i\hbar}[\mathcal{H}', \rho_{\text{st}}] - \kappa(a^\dagger a\rho_{\text{st}} + \rho_{\text{st}}a^\dagger a - 2a\rho_{\text{st}}a^\dagger) \\ - \gamma(S^+ S^- \rho_{\text{st}} + \rho_{\text{st}} S^+ S^- - 2S^- \rho_{\text{st}} S^+) \\ - \gamma_z((S^z)^2 \rho_{\text{st}} + \rho_{\text{st}}(S^z)^2 - 2S^z \rho_{\text{st}} S^z) = 0. \end{aligned} \quad (32)$$

This is a homogeneous equation of  $\rho_{\text{st}}$  [16]. The expectation values

$$I = \text{Tr}(a + a^\dagger)\rho_{\text{st}} \quad \text{and} \quad Q = \text{Tr}(ia^\dagger - ia)\rho_{\text{st}} \quad (33)$$

correspond to the heterodyne measurement of the field quadratures. The steady-state transmission amplitude is given by  $Q$ . For a small strength  $\xi = 0.001$ , the transmission shows a peak corresponding to the vacuum-field Rabi splitting.

In figure 5(a), we compare the line shape of the linear response  $\chi(\omega)$  ( $\circ$ ), and that of  $Q/2\xi$  for various values of  $\xi$ . We find that the spectrum of  $Q$  for small values of  $\xi$  agrees with  $\chi(\omega)$ . We plot the spectrum of  $|\langle a \rangle|^2/\xi^2$  in figure 5(b). Here we find that  $\chi(\omega)$  tends to move to a single peak, but  $|\langle a \rangle|^2$  keeps a double-peak structure. We also show the spectrum of  $\langle a^\dagger a \rangle/\xi^2$  in figure 5(c). In cases with many spins and photons,



**Figure 5.** (a) Dependence of  $Q/2\xi = \text{Im}\langle a \rangle / \xi$  on the driving frequency  $\omega$ , for different  $\xi$ . The blue dots show the linear response case (see figure 4,  $T = 0.1$ ). (b) Driving frequency  $\omega$  dependence of  $|\langle a \rangle|^2 / \xi^2$  of stationary states under various strengths of the driving force  $\xi$ . (c) Driving frequency  $\omega$  dependence of the photon number of stationary states  $\langle a^\dagger a \rangle / \xi^2$  under various strengths of the driving force  $\xi$ . The parameters are  $N = 5$  and  $n_{\max} = 25$ , with  $g = 0.1$ ,  $\kappa = 0.01$  and  $\gamma_{xy} = \gamma_z = 0.01$ . The strengths of  $\xi$  are 0.01, 0.02, 0.05, 0.1, 0.2 and 0.5, in increasing order as indicated by the arrows in each figure.

there exist many resonant modes within the vacuum-field Rabi splitting, and the double-peak structure is affected.

Under strong drive, the photon number shows a single peak, while the heterodyne signal has a double-peak shape. This can be understood from a viewpoint of the phase decoherence of the photon mode.

In the case  $\omega = \omega_0$ ,

$$\begin{aligned} |\psi(t)\rangle_{\text{ph}} &= e^{i\xi(a^\dagger + a)t} |\psi(0)\rangle_{\text{ph}} = e^{-\frac{|\xi t|^2}{2}} \sum_{n=0}^{\infty} \frac{(i\xi t)^n (a^\dagger)^n}{n!} |0\rangle \\ &= e^{-\frac{|\xi t|^2}{2}} \sum_{n=0}^{\infty} \frac{(i\xi t)^n}{\sqrt{n!}} |n\rangle, \end{aligned} \quad (34)$$

where we used the relation

$$|n\rangle = \frac{1}{\sqrt{n!}} (a^\dagger)^n |0\rangle. \quad (35)$$

In the case with  $|\psi(0)\rangle_{\text{ph}} = |0\rangle$ , the number of photons increases with time:

$$a|i\xi t\rangle = i\xi t|i\xi t\rangle \quad \text{and} \quad \langle n \rangle \propto (\xi t)^2. \quad (36)$$

The damping term causes a saturated value of the photon number in the stationary state. The dissipation term causes phase decoherence of the coherent state, due to the phase fluctuation

$$a|i\xi t\rangle = e^{i\delta} (i\xi t)|i\xi t\rangle. \quad (37)$$

By averaging over  $\delta$ ,  $|\langle a \rangle|$  is strongly reduced, while on the other hand,  $\langle a^\dagger a \rangle$  is robust against this effect. This explains the large reduction of the spectrum at its centre  $\omega \simeq \omega_0 = \omega_S$ , where the cavity photons are incoherently excited.

## 5. Dissipation of the total spin

The total spin is given by

$$S(S+1) = M_x^2 + M_y^2 + M_z^2. \quad (38)$$

So far, we studied the case where the total spin is fixed to be the maximum value ( $S = N/2$ ). This value is for the state with all the spin aligned. When the system is excited, the value of the magnetization  $M = -S + (\text{number of up spins})$  increases. In order to conserve the total spin, the  $xy$  components must

be coherent. In the model Hamiltonian (3), the total spin is conserved and the dissipation dynamics (12) does not affect the total spin either. Therefore, in the present model, when the spins are excited, namely they are polarized longitudinally, the coherence of the transverse component appears. This large coherent transverse component would cause super-radiance [23].

However, in practice, we may expect decoherence of the spin state. In this case, the total spin is reduced. While the maximum of  $S^2$  is given by  $S(S+1) = N(N+2)/4$ , the actual value can be reduced to the value corresponding to  $N$  phase-independent spins

$$\langle \phi | S^2 | \phi \rangle = \frac{3N}{4}, \quad (39)$$

where

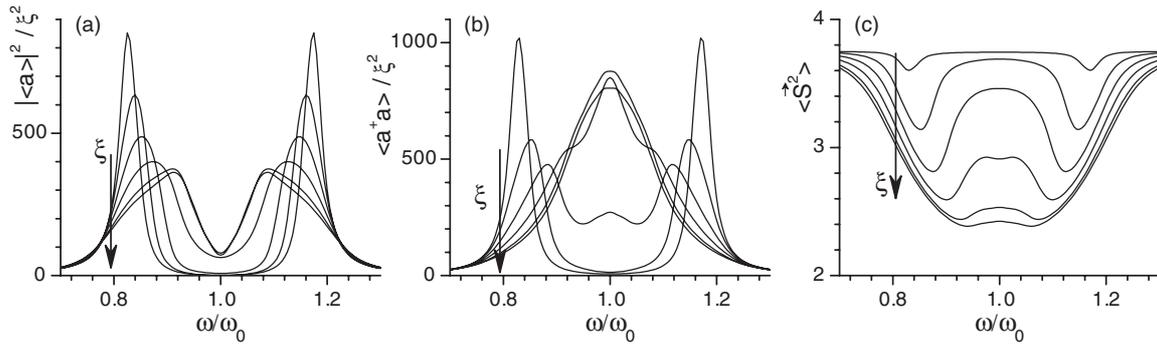
$$|\phi\rangle = \frac{1}{2^{N/2}} \sum_{\{\sigma_i = \pm 1\}} |\sigma_1, \sigma_2, \dots, \sigma_N\rangle. \quad (40)$$

This implies that we cannot use the Tavis–Cummings model with only one value of  $S$ . The effective value of  $S$  may change for each frequency. Thus, we must adopt a Hamiltonian with  $2^N$ -dimensional spin space instead of  $2S+1$ .

To take into account this fact, we adopt a new type of dissipation mechanism, in which the total spin can change. In the process given by the master equation (12), the environment couples to the total magnetization  $M_z = \sum_{i=1}^N S_i^z$ , and the dissipation effect was given by the last term in (12). In contrast, now we include the thermal contact to each spin, and we have a new master equation:

$$\begin{aligned} \frac{d}{dt} \rho &= \frac{1}{i\hbar} [\mathcal{H}, \rho] - \kappa (a^\dagger a \rho + \rho a^\dagger a - 2a \rho a^\dagger) \\ &\quad - \gamma_{xy} \sum_{i=1}^N (S_i^+ S_i^- \rho + \rho S_i^+ S_i^- - 2S_i^- \rho S_i^+) \\ &\quad - \gamma_z \sum_{i=1}^N ((S_i^z)^2 \rho + \rho (S_i^z)^2 - 2S_i^z \rho S_i^z). \end{aligned} \quad (41)$$

Using the present master equation in the extended space, we present in figure 6 the frequency dependences of  $|\langle a \rangle|^2$ ,  $\langle a^\dagger a \rangle$  (normalized to  $\xi^2$ ), and  $|\langle 2M + N \rangle|^2$  for stationary states. For  $N = 3$ , the maximum value of  $S^2$  is  $S(S+1) = 3.75$ , and



**Figure 6.** Frequency dependences of (a)  $|\langle a \rangle|^2 / \xi^2$ , (b)  $\langle a^\dagger a \rangle / \xi^2$  and (c)  $|\langle S^2 \rangle$  as a function of the frequency of the driving force with the dissipation term (equation (41)) for  $N = 3$ ,  $n_{\max} = 20$  with  $g = 0.1$ ,  $\kappa = 0.01$  and  $\gamma_{xy} = \gamma_z = 0.01$ . The values of  $\xi$  are 0.01, 0.03, 0.05, 0.07, 0.09 and 0.1, in increasing order as shown by an arrow in each figure.

its minimum value is  $3N/4 = 2.25$ .<sup>6</sup> Here we find that the total spin is largely reduced, and almost reaches the minimum value.

Thus, we expect that even if the number of spins is large in the cavity, practical values of  $S$  could be small. In the thermodynamic equilibrium state, we must estimate what number of spins in the material contribute coherently (see the following section).

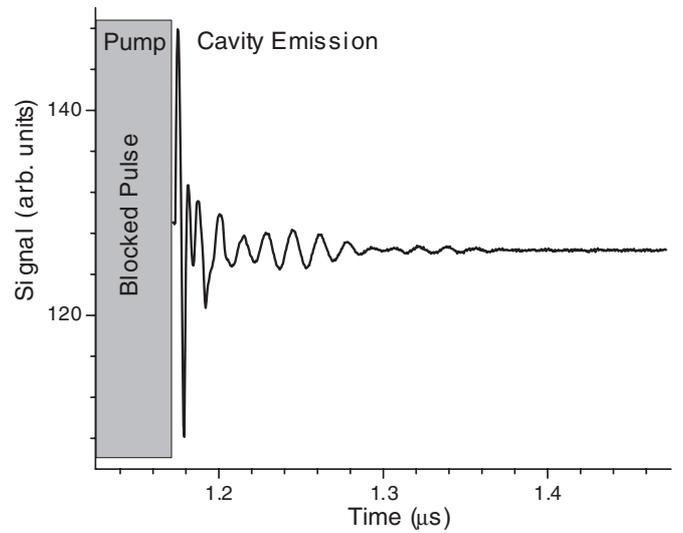
## 6. Experimental study

### 6.1. Strong coupling regime

We have carried out a spectroscopic study of a large  $N = N_{\text{spins}}$  sample, in the strong coupling regime. The system of choice is the two-level spin system of the diphenyl-picrylhydrazyl (DPPH), a well-known electronic spin resonance (ESR) standard [24]. Each DPPH molecule contains a single spin  $S = 1/2$  and therefore the spins are relatively diluted and their environment is lacking anisotropy ( $g_s \simeq 2$ ). As a consequence, the line width of the DPPH resonance is very narrow, of the order of 0.1 mT. It is only at very high Zeeman splitting (large static fields and large frequencies, of few hundreds GHz) that the anisotropy in the  $g$ -factor starts to be detectable, leading to an increased resonance linewidth. But at frequencies  $\sim 10$  GHz as in our study, DPPH is among the best candidates to study resonance splitting due to cavity photons rather than to anisotropy in the spin environment.

The sample is placed inside a cylindrical cavity operated in the  $TE_{011}$  mode, with an optimized coupling to the microwave B-field. The resonance mode is located at  $\omega_0/2\pi = 9.624$  GHz and the system is operated in reflection: a microwave pulse is pumped into the cavity and the reflected pulse is detected by a home-built heterodyne setup. More precisely, the reflected pulse is down-mixed by another pulse with a frequency close to the one used in the pump pulse (in our case, the shift is of 100 MHz and the pump pulse has a length of 1  $\mu$ s).

To study the eigenenergies of the coupled spin–photon system, we use the microwave pulse at  $\omega$  to excite the system and record the photon release after the excitation. When the

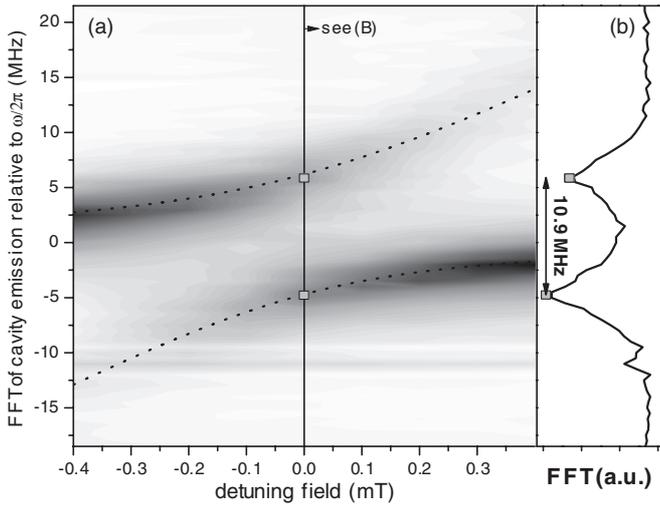


**Figure 7.** Example of cavity ringing detected following a pump pulse. The readout is blocked during pumping to avoid potentially large signals into the amplification stage. The cavity emission shows beatings which are visible as distinct peaks in the FFT trace (see figure 8).

microwave is switched off, the cavity coherently emits photons corresponding to its own eigenmodes. The time evolution of such a signal is shown in figure 7 where one can clearly see beatings of close but distinct frequencies in the emitted signal. This technique is called cavity ringing and allows us to study the energy spectrum by detecting photon frequencies in the fast Fourier transform (FFT) of decaying ringing oscillations. Obviously, most of the FFT spectrum is localized at the pump frequency. Therefore, we detune slightly the pump from the cavity resonance (by 50 MHz), in order to see the cavity-spin eigenmodes in the tail of the FFT.

Measured Fourier traces as a function of the applied static field are presented in figure 8. The FFT intensity is grey coded, the dark areas representing the eigenmodes of the spin–photon coupled system. Frequencies are relative to the sample-loaded cavity resonance, in the absence of applied field. The static field is varied around the empty cavity resonance condition  $\mu_0 H_z = \hbar\omega_0/g_s\mu_B$ , with  $g_s = 2$  by a detuning given by  $\mu_0\delta H_z = -\hbar\Delta/g_s\mu_B$ . At zero detuning, the actual FFT plot is given in figure 8 and one clearly distinguishes (that is, the peak

<sup>6</sup> We are preparing the massive calculation needed for a larger number of spins.



**Figure 8.** (a) Intensity plot of the Fourier transform of detected cavity ringing, measured at room temperature. The plot is given as a function of the magnetic field around the resonance condition (FFT frequencies are relative to the cavity resonance). One observes a level repulsion developing in the resonance window, due to the strong spin-photon coupling. The effect is particularly visible at zero detuning (vertical marking line). (b) Actual FFT trace recorded at zero detuning showing a measured Rabi splitting of 10.9 MHz. The peaks are distinguishable by a splitting larger than the peak width.

separation is larger than the peak width) two eigenenergies split by 10.9 MHz. The continuous lines are fits made considering a level repulsion  $\Delta$ .

In our experimental conditions, due to the spin-photon strong coupling, one observes two peaks separated by the characteristic Rabi splitting. The strength of the coupling can be estimated using the model of  $N$  spins coupled to a single photon, although the cavity contains a large number of  $n_{\max}$  photons. As we see in figure 3, as long as the number of photons ( $n_{\max}$ ) is much smaller than the number of spins  $N$ , the resonance frequencies are given by  $E(1, k) - E(0, 1)$ ,  $k = 1, 2$ , of equations (9). Thus, the difference of the resonance frequencies is given by that of the vacuum-field Rabi oscillation. Therefore, the cavity ringing shows coherent oscillations with the frequencies  $\omega_{e0,1}/2\pi$ , given by

$$\omega_{e0,1} - \omega = -\frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2 + \Omega_R^2}, \quad (42)$$

on which the continuous lines of figure 8(a) are based. The fit procedure leads to a Rabi splitting of  $\Omega_R/2\pi = 10.9$  MHz.

### 6.2. Photon number and the types of Rabi oscillations

As mentioned previously, it is important to compare the number of spins (or total spin size) to the number of photons, since quantum peaks require large splitting (large  $N$ ) and a smaller number of photons. In our experiment, taking into account the DPPH spin density, we estimate the number of spins to be of the order of  $10^{20}$ . Since the temperature is  $T = 300$  K, the size of the total spin will be reduced due to the Boltzmann population of the two levels. For a Zeeman splitting of  $\omega = 9.62$  GHz, the magnetization is estimated as

$$m(T = 300 \text{ K}) = \frac{N_{\text{spins}}}{2} \tanh\left(\frac{\hbar\omega}{2k_B T}\right) \simeq 10^{-3} \frac{N_{\text{spins}}}{2}, \quad (43)$$

and thus the dominant value of the total spin is of the order of

$$S = m(T = 300 \text{ K}) \sim 10^{-3} \frac{N_{\text{spins}}}{2}. \quad (44)$$

The number of photons inside the cavity can be roughly estimated by using the  $Q$  factor of the cavity ( $Q \sim 1000$ ) to obtain a photon life time of less than 100 ns and estimations of the microwave power delivered by the setup. We find a range for the number of photons of the order of  $10^{12}$ – $10^{14}$  which fulfils the condition

$$n_{\text{photon}} \ll S. \quad (45)$$

In this case, we expect indeed that the vacuum-field Rabi splitting of the resonance frequency to be clearly found. The splitting is measured to be of the order of 10 MHz:

$$\Omega_R/2\pi \simeq g\sqrt{S} = 10 \text{ MHz}, \quad (46)$$

from which we can estimate an order of magnitude for the coupling constant  $g \sim 10^{-2}$ – $10^{-1}$  Hz.

On the other hand, recent experiments demonstrate the existence of coherent Rabi oscillation of diluted spin systems driven by an electromagnetic field [17, 18]. In those experiments, the number of photons in the cavity is much larger than  $N$  as we see in figure 2. It is therefore an interesting future problem to study the transition between these two regions (i.e. the region of the vacuum-field Rabi oscillation ( $n \ll N$ ) and that of the field-driven Rabi oscillation ( $n \gg N$ )).

## 7. Summary and discussion

We studied the line shape as a function of the size of an ensemble of spins and the cavity mode photon number. We found that the line shapes change depending on the number of spins, number of photon and also types of dissipation.

We found two regions of the photon numbers. One of them is the cavity ringing region where  $N \gg n$ , while the other is that of coherent Rabi oscillation under driving electromagnetic field.

We presented experimental data of the hybridization of the cavity and of the spins which shows double peaks in the spectrum representing the vacuum-field Rabi oscillation due to the cavity ringing, and we estimated the number of spins and photons contributing to the coherent dynamics.

With our model, the role of other factors in the spread of Rabi splitting, such as dipolar or hyperfine interactions, local anisotropic crystal fields and the size of the sample versus size of a cavity (mode), can be studied as well. It is also an interesting problem to study the relation between this dissipation effect and the existence of the optical bistability [25–28]. If the dissipation of the transverse coherence does not exist, the polarization of the system is monotonic with the driving force. But the dissipation can cause a nonlinear dependence which leads to optical bistability.

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## References

- [1] Jaynes E T and Cummings F W 1963 *Proc. IEEE* **51** 89
- [2] Tavis M and Cummings F W 1968 *Phys. Rev.* **170** 379
- [3] Kaluzny Y, Goy P, Gross M, Raimond J M and Haroche S 1983 *Phys. Rev. Lett.* **51** 1175–8
- [4] Sanchez Mondragon J J, Narozhny N B and Eberly J H 1983 *Phys. Rev. Lett.* **51** 550
- [5] Thompson R J, Rempe G and Kimble H J 1992 *Phys. Rev. Lett.* **68** 1132
- [6] Brune M, Schmidt-Kaler F, Maali A, Dreyer J, Hagley E, Raimond J M and Haroche S 1996 *Phys. Rev. Lett.* **76** 1800
- [7] Rabl P *et al* 2006 *Phys. Rev. Lett.* **97** 033003
- [8] Imamoglu A 2009 *Phys. Rev. Lett.* **102** 083602
- [9] Chiorescu I, Groll N, Bertaina S, Mori T and Miyashita S 2010 *Phys. Rev. B* **82** 024413
- [10] Schuster D I *et al* 2010 *Phys. Rev. Lett.* **105** 140501
- [11] Kubo Y *et al* 2010 *Phys. Rev. Lett.* **105** 140502
- [12] Amsüss R *et al* 2011 *Phys. Rev. Lett.* **107** 060502
- [13] Zhu X *et al* 2011 *Nature* **478** 221
- [14] Kubo Y *et al* 2011 *Phys. Rev. Lett.* **107** 220501
- [15] Agarwal G S 1984 *Phys. Rev. Lett.* **53** 1732
- Houdre R, Stanley R P and Ilegems M 1996 *Phys. Rev. A* **53** 2711
- Zhu Y, Gauthier D J, Morin S E, Wu Q, Carmichael H J and Mossberg T W 1990 *Phys. Rev. Lett.* **64** 2499
- [16] Bishop L S, Chow J M, Koch J, Houck A A, Devoret M H, Thuneberg E, Girvin S M and Schoelkopf R J 2009 *Nature Phys.* **5** 105
- [17] Bertaina S, Gambarelli S, Tkachuk A, Kurkini N, Malkin B, Stepanov A and Barbara B 2007 *Nature Nanotechnol.* **2** 39
- Bertaina S, Gambarelli S, Mitra T, Tsukerblat B, Muller A and Barbara B 2008 *Nature* **453** 203
- Bertaina S, Chen L, Groll N, Van Tol J, Dalal N S and Chiorescu I 2009 *Phys. Rev. Lett.* **102** 050501
- Bertaina S, Shim J H, Gambarelli S, Malkin B Z and Barbara B 2009 *Phys. Rev. Lett.* **103** 226402
- Bertaina S, Groll N, Chen L and Chiorescu I 2011 *Phys. Rev. B* **84** 134433
- [18] De Raedt H, Barbara B, Miyashita S, Michielsen K, Bertaina S and Gambarelli S 2012 *Phys. Rev. B* **85** 014408
- [19] Kubo R, Toda M and Hashitsume N 1085 *Statistical Physics II* (New York: Springer)
- [20] Lousell W H 1973 *Quantum Statistical Properties of Radiation* (New York: Wiley)
- [21] Walls D F and Milburn G J 1995 *Quantum Optics* (Berlin: Springer)
- [22] Uchiyama C, Aihara M, Saeki M and Miyashita S 2009 *Phys. Rev. E* **80** 021128
- [23] Dicke R H 1954 *Phys. Rev.* **93** 99
- [24] Krzystek J, Sienkiewicz A, Pardi L and Brunel L C 1997 *J. Magn. Reson.* **125** 207
- [25] Lugiato L A 1984 *Progress in Optics* vol 21 ed E Wolf (Amsterdam: North Holland) pp 69–216
- [26] Drummond P D 1981 *IEEE J. Quantum Electron.* **17** 301
- [27] Gripp J, Mielke S L, Orozco L A and Carmichael H J 1996 *Phys. Rev. A* **54** R3746
- Raizen M G, Thompson R J, Brecha R J, Kimble H J and Carmichael H J 1989 *Phys. Rev. Lett.* **63** 240
- [28] Shirai T, Mori T and Miyashita S 2012 arXiv:1204.5516