Nonlinear spectroscopy in the strong-coupling regime of cavity QED

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Abstract

Nonlinear spectroscopic techniques, such as two-photon absorption and four-wave-mixing, on atoms in high-Q cavities (strong coupling limit) provide a means to probe higher-order transitions in the Jaynes-Cummings ladder of energy states of the atom+cavity system. It can be used for exploring and improving our understanding of the rich structure of the atom+cavity "molecule". The dressing of the atomic levels by the quantized cavity field alters the nonlinear susceptibility of the atoms leading to new resonances. These resonances exhibit super-radiant character. The resonances also exhibit dependences on the number of atoms present in the cavity. The nonlinearity of the radiation-matter interaction along with the coupled atom+field oscillator model has to be used to explain the new resonances.

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I. INTRODUCTION

Cavity Quantum Electrodynamics (QED) is an exciting frontier of quantum optics research where new technology is now making it possible to experimentally test the validity of concepts of light matter interactions in the quantum limit. The radiative properties of atoms such as the spectrum and spatial distribution of electromagnetic noise are modified when they are confined to a cavity of dimensions comparable to the wavelength. This was demonstrated by Purcell [1] half a century ago.

In free space the atom decays spontaneously by coupling to a large number of vacuum modes. This decay is exponential in time and is irreversible. The transition rate is governed by Fermi's Golden rule. But when an atom is placed in a high–Q cavity, it couples to the eigenmodes of the cavity. The radiation from the atom can survive for long in the cavity and is able to re-excite the atom. Hence spontaneous emission in a high–Q cavity is oscillatory and non-perturbative [2].

The strong coupling limit is defined as the regime where the atom–cavity field interactions evolve faster than the dissipation due to spontaneous emission and cavity losses, ie. $g_0 \geq \gamma, \kappa$ where g_0 is the atom–cavity coupling, γ is the atomic decay rate and κ is the cavity decay rate. New technology makes it possible to fabricate extremely high–Q cavities suitable for the study of the atom–cavity system in the strong coupling limit. The atom and cavity form a joint system with new eigenstates belonging to the atom + cavity "molecule". Hence the absorption and emission spectra are modified. Under these conditions the number of photons required to saturate an intracavity atom is [2] $n_0 \sim \gamma^2/g_0^2 \leq 1$. The number of atoms required to have an appreciable effect on the cavity field is [2] $n_0 \sim \kappa\gamma/g_0^2 < 1$.

Cavity QED is being used to test our fundamental concepts on the quantum theory of light and matter. For example, Haroche et. al., have shown that the strong entanglement between the atom and the cavity field can be used to generate "Schrödinger cat" states and to study nonlocal correlations which would shed light on Bell's inequalities issues. Walther et. al., have used a one–atom maser (micromaser) to generate nonclassical states of light displaying sub-Poissonian photon statistics with photon number fluctuations much below the standard quantum limit. A good review of these and other interesting cavity QED experiments can be found in Ref. [2].

In this paper we study how the nonlinear response of the atom + cavity "molecule" is modified. Pump–probe techniques are used to study the two-photon absorption (2PA) and four-wave-mixing (FWM) behavior of the system. These nonlinear spectroscopic studies shed light on the rich level structure of the higher order transitions in the "molecule". The appearance of interesting effects such as the dependence of the transition frequency on the number of atoms and superradiant behavior have been predicted.

II. THEORY

In the strong coupling limit, the atom and the cavity field form a system of coupled oscillators. The case of a single atom interacting with a single mode of a high–Q cavity was studied by Jaynes and Cummings [3]. The generalization to multiple atoms was given by Tavis and Cummings [4]. In the rotating wave approximation and for the dissipationless case, the total Hamiltonian for N atoms in a cavity is given by

$$H = H_0 + H_1 \qquad \text{where}$$
$$H_0 = \hbar\omega_0\sigma^z + \hbar\omega_c\hat{a}^\dagger\hat{a}$$
$$H_1 = \hbar\sum_{l=1}^{N}\left[g_0\hat{a}^\dagger\hat{\sigma}_l^- + g_0^*\hat{a}\hat{\sigma}_l^+\right]$$

where $g_0 = \Omega_{Rabi}/2$, $\hat{\sigma}_l^+$ and $\hat{\sigma}_l^-$ are the raising and lowering operators for the lth atom and $\hat{a}, \hat{a}^\dagger$ are the annihilation and creation operators of the cavity field mode satisfying $[\hat{a}, \hat{a}^\dagger] = 1$. The operator $\frac{\hat{\sigma}^z}{2} + \hat{a}^\dagger\hat{a}$ is a conserved quantity for the above Hamiltonian. Physically this means that the total number of excitations of the atom+cavity field system is conserved. Hence the dressed states of the system can be classified according to the eigenvalues of $\frac{\hat{\sigma}^z}{2} + \hat{a}^\dagger\hat{a}$.

In general the atom–cavity coupling gives rise to an infinite hierarchy of states. We shall concentrate on the limit of vanishing excitation. In $\chi^{(3)}$ processes such as 2PA and FWM, the atoms absorb at most two photons from the external field. So the cavity field excitation can change by a maximum number of two photons. Hence it is sufficient to look at dressed states corresponding to the eigenvalues of $\frac{\hat{\sigma}^z}{2} + \hat{a}^\dagger \hat{a}$ equal to

$$-\frac{N}{2}, -\frac{N}{2}+1, -\frac{N}{2}+2$$

corresponding to 0, 1 and 2 excitations respectively [5].

The ground state $|0\rangle$ corresponds to zero excitations of both the atoms and the cavity field. The energy of the state is $E_0 = -\frac{N}{2}\,\omega_0$.

For a single quanta of excitation of the atom+cavity system, the basis states are

$$|\psi_1\rangle = |1\rangle_c|0\rangle_A, \qquad |\psi_2\rangle = |0\rangle_c|1\rangle_A$$

where $|n\rangle_c$ is the state of the cavity field with n photons,
$|1\rangle_A \sim \sum_{l=1}^{N} |0\rangle_1|0\rangle_2 \ldots |1\rangle_l \ldots |0\rangle_N$ and
$|0\rangle_A \Rightarrow$ all atoms are in the ground state.
The eigenstates containing a single excitation are given by

$$|\pm\rangle = \frac{1}{\sqrt{2}}\left[|\psi_2\rangle \pm |\psi_1\rangle\right].$$

The energies are $E_\pm = \left(-\frac{N}{2}+1\right)\omega_0 \pm g_0\sqrt{N}$.

For states with two quanta of excitation, ie. $\langle\frac{\hat{\sigma}^z}{2} + \hat{a}^\dagger\hat{a}\rangle = -\frac{N}{2}+2$, the basis states are $|\phi_1\rangle = |0\rangle_c|2\rangle_A$, $|\phi_2\rangle = |1\rangle_c|1\rangle_A$, $|\phi_3\rangle = |2\rangle_c|0\rangle_A$
where $|2\rangle_A \sim \sum_{l\neq p}^{N} |0\rangle_1 \ldots |1\rangle_l \ldots |1\rangle_p \ldots |0\rangle_N$.
The eigenstates are linear combinations of the $|\phi_i\rangle$ and are given by

$$|1\rangle = \left(\frac{N-1}{4N-2}\right)^{1/2}|\phi_1\rangle + \frac{1}{\sqrt{2}}|\phi_2\rangle + \left(\frac{N}{4N-2}\right)^{1/2}|\phi_3\rangle$$
$$|2\rangle = -\left(\frac{N}{2N-1}\right)^{1/2}|\phi_1\rangle + 0\times|\phi_2\rangle + \left(\frac{N-1}{2N-1}\right)^{1/2}|\phi_3\rangle$$
$$|3\rangle = \left(\frac{N-1}{4N-2}\right)^{1/2}|\phi_1\rangle - \frac{1}{\sqrt{2}}|\phi_2\rangle + \left(\frac{N}{4N-2}\right)^{1/2}|\phi_3\rangle.$$

The eigenenergies are given by

$$E_{1,3} = \left(-\frac{N}{2} + 2\right)\omega_0 \pm g_0(4N-2)^{1/2}, \qquad E_2 = \left(-\frac{N}{2} + 2\right)\omega_0 \ .$$

The first two excited state energy levels for the Jaynes-Cummings (single atom case) and Tavis-Cummings (multiple atoms case) energy ladders are shown in Fig.1(a) and (b) respectively.

So far we have discussed the ideal case of a dissipationless system with no incident fields. Our aim is to study the dynamics of the atom+cavity system when it is excited with pump and probe fields. In reality the atom+cavity system decays due to spontaneous emission (to modes other than the cavity modes, $\gamma_{||}$), transverse decay ($\gamma_\perp = \gamma_{||}/2$ for pure radiative decay) and cavity field decay (κ). The evolution of the system under these various influences is governed by the Heisenberg's equations of motion which can be derived from the quantum master equation [6]. In the semiclassical approximation where the atomic and cavity field wavefunctions are considered disentangled (thereby making approximations such as $\langle\hat{\sigma}^z\hat{a}\rangle \rightarrow \langle\hat{\sigma}^z\rangle\langle\hat{a}\rangle$, $\langle\hat{a}\hat{\sigma}_l^-\rangle \rightarrow \langle\hat{a}\rangle\langle\hat{\sigma}_l^-\rangle$, and $\langle\hat{a}^\dagger\hat{\sigma}_l^+\rangle \rightarrow \langle\hat{a}^\dagger\rangle\langle\hat{\sigma}_l^+\rangle$ possible), the Heisenberg's equations of motion for the cavity field mode $\langle\hat{a}\rangle$, atomic polarization $\langle\hat{\sigma}_l^-\rangle$ and inversion $\langle\hat{\sigma}^z\rangle$ for the l th atom in a sample of N atoms is given by [7]

$$\dot{\langle\hat{a}\rangle} = -(\kappa + i\Theta)\langle\hat{a}\rangle + \sum_l^N g\langle\hat{\sigma}_l^-\rangle + \epsilon \tag{1}$$

$$\dot{\langle\hat{\sigma}_l^-\rangle} = -(\gamma_\perp + i\Delta)\langle\hat{\sigma}_l^-\rangle + g\langle\hat{a}\rangle\langle\hat{\sigma}^z\rangle \tag{2}$$

$$\dot{\langle\hat{\sigma}^z\rangle} = -\gamma_{||}(\langle\hat{\sigma}^z\rangle + 1) - 2g^*(\langle\hat{a}^\dagger\rangle\langle\hat{\sigma}_l^-\rangle + \langle\hat{a}\rangle\langle\hat{\sigma}_l^+\rangle) \tag{3}$$

where $\Theta = (\omega_c - \omega_p)/\kappa$ is the cavity detuning, $\Delta = (\omega_a - \omega_p)/\gamma_{||}$ is the atomic detuning and $\epsilon = \epsilon_p + \epsilon' e^{-i\nu t}$ is the pump plus probe driving fields. The pump is at frequency ω_p and the probe is at a frequency detuning ν from ω_p. The equations tell us that the evolution of the atoms and the cavity field is entangled. From these equations it is possible to derive the the absorption coefficient and other related quantities of interest.

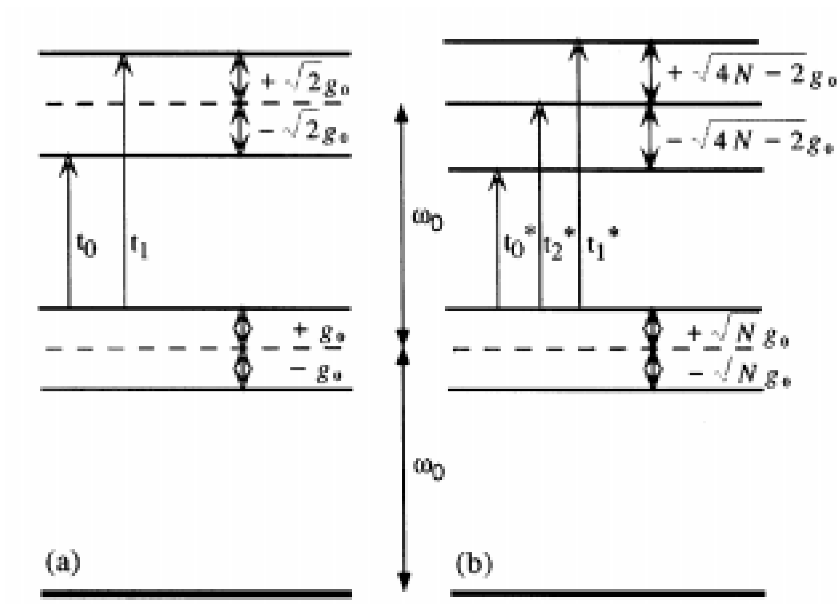

FIG. 1. Comparison of the level structure of the first two excited states of a coupled atom–cavity system for the one atom case (left) and the many atom case (right). In fig. (b) the ground state is $|0\rangle$, the one excitation state has levels $|+\rangle$ (higher energy) and $|-\rangle$ (lower energy) and the two excitations state is made up of levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ in descending order of energy. The various t 's in the figure indicate some of the possible transitions.

III. TRANSITIONS IN THE ATOM–CAVITY MOLECULE

A. Linear absorption

We first look at the case of linear absorption by the atom+cavity system. The two channels for a single photon absorption are $|0\rangle \rightarrow |\pm\rangle$, with resonances at $\omega = \omega_0 \pm g_0\sqrt{N}$ in the absence of dissipation (cf. Fig.1). In the $N = 1$ limit, the splitting is $\pm g_0$. The interaction of the atom with the cavity field mode gives rise to the single atom vacuum-Rabi splitting which has been observed in spectroscopy experiments [8,2].

In the presence of dissipation, the transmission of a weak probe beam for the case of zero detuning between cavity and atom (i.e. $\omega_c = \omega_0$) would be [7]

$$t_{lin}(\omega_p) = \frac{\kappa(\gamma_\perp - i\omega_p)}{(\lambda_+ - i\omega_p)(\lambda_- - i\omega_p)}$$

where $\lambda_\pm = -\frac{\kappa+\gamma_\perp}{2} \pm \left[\left(\frac{\kappa-\gamma_\perp}{2}\right)^2 - g_0^2 N\right]^{1/2}$, i.e. the resonances are shifted. The condition $\frac{\kappa+\gamma_\perp}{2} < g_0\sqrt{N}$ (strong coupling limit) is essential for observing the split in resonance so that it is not smeared out by the dissipation.

B. Nonlinear interactions

We now turn our attention to nonlinear interactions in the atom+cavity system. Pump-probe experiments where a pump beam in tuned to one of the resonances and the absorption of a weak tunable probe beam is monitored, are used to study the nonlinear spectroscopy of the system. For example, if we consider the following sequence of absorption $|0\rangle \xrightarrow{\omega_{pump}} |+\rangle \xrightarrow{\omega_{probe}} |1\rangle$, resonance occurs at

$$\begin{aligned}
\omega_{probe} &= \omega_0 + g_0(\sqrt{4N-2} - \sqrt{N}) \\
&\to \omega_0 + g_0\sqrt{N} \qquad \text{for large N} \\
&\to \omega_0 + g_0(\sqrt{2} - 1) \qquad \text{for N=1}
\end{aligned}$$

For 2PA with $\omega_{pump} = \omega_{probe} = \omega$, resonances occur at $\omega = \omega_0 \pm g_0\sqrt{N - 1/2}$.

Four-wave-mixing susceptibilities can be calculated using standard perturbative methods. For example, the nonlinear interaction arising due to $\chi^{(3)}(\omega_1, \omega_1, -\omega_2)$ has resonances at [5] $\omega_2 = \omega_0 \pm g_0\sqrt{N}$, and $\omega_2 = 2\omega_1 - \omega_0 \pm g_0\sqrt{N}$.

Another interesting effect is that of atomic coherence. By tuning the pump to ω_0, the paths $|0\rangle \to |-\rangle \to |1\rangle$ and $|0\rangle \to |+\rangle \to |1\rangle$ can be made equivalent. The destructive interference between the two paths can give rise to a interference minimum for two photon absorption [9]. This is similar to the creation of dark states where the phase relation between the wavefunctions of intermediate states results in destructive interference for the process of absorption to a higher state.

In all the examples given above, we see that each free space resonance of the nonlinear absorption spectra is split by the interaction with the cavity modes into a doublet. The resonance frequency depends on the *number* of atoms in the cavity. Also the strength of these transitions depend on the dipole matrix elements which depend on N. For example [9,5],

$$d_{\pm 0} = d\sqrt{N/2}, \qquad d_{\pm 1} = \frac{d}{2}\left(\frac{(2N-2)}{[2(2N-1)]^{1/2}} \pm N^{1/2}\right).$$

where d is the free atomic dipole moment and d_{ij} are the dipole moments for transitions from the i to the j states. In the limit of large N,

$$d_{3+}, d_{1-} \rightarrow 0, \qquad d_{1+} \rightarrow dN^{1/2}, \qquad d_{2\pm} \rightarrow \mp d(N/2)^{1/2}.$$

Hence depending on the number of atoms present, some transitions are enhanced while others are destroyed. The radiation due to each transition varies as the square of the dipole matrix elements and so we see that certain transitions, for example the transition $|0\rangle \rightarrow |+\rangle \rightarrow |1\rangle$, will exhibit a N^2 dependence, ie. superradiant character. Also since $d_{1-} \sim 0$ in the large N limit, there would no longer be an interference minimum for 2PA mentioned above.

IV. EXPERIMENT

I shall now briefly describe the experimental efforts by Kimble et. al. [7] towards the study of nonlinear cavity QED. The experiments are sensitive to the slightest of fluctuations and are difficult to perform. The whole experimental apparatus has to be actively controlled and stabilized. The main experimental problems arise from the fact that the number of atoms in the cavity and their positions fluctuate leading to a change in the coupling constants and in the energy level structure.

A high finesse cavity ($F \sim 10^5$) of length 346 μm formed by two high reflecting curved mirrors (transmission $\sim 10^{-6}$, scattering losses $\sim 10^{-6}$) is used. As mentioned earlier, such high quality cavities are essential to achieve the strong-coupling condition for these experiments. The cavity length is actively controlled. An optically prepared beam of cesium atoms intersects the cavity axis at 90^o. In this experiment the $(6S_{1/2}F = 4, m_F = 4) \rightarrow (6P_{3/2}, F' = 5, m_F' = 5)$ transition was investigated at 852 nm.

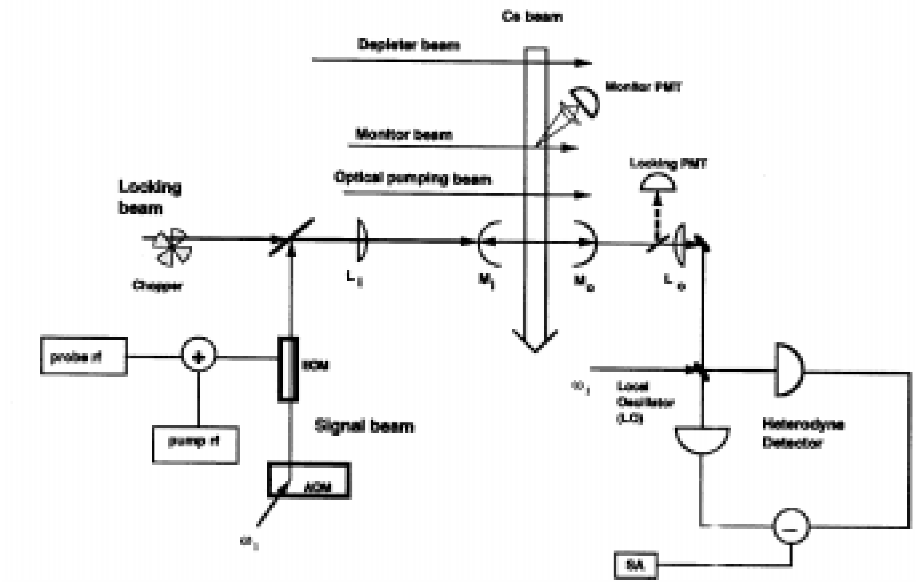

FIG. 2. Experimental setup (from Ref. [7]).

The probe field is generated by modulating the output of a frequency stabilized titanium-sapphire laser with an acousto-optic modulator (AOM) and an electro-optic modulator (EOM). The pump field required for pump-probe experiments is generated by adding an additional constant-frequency, variable strength rf signal to the probe. Figure 2 shows a schematic of the setup. Figure 3 shows the transmission spectrum of the probe beam in the absence of the pump field. The observed doublet structure with peaks at $\omega = \pm g_0$ arising due to the $|0\rangle \xrightarrow{\omega_{probe}} |\pm\rangle$ transitions is a direct spectroscopic measurement of the vacuum-Rabi splitting.

To study the multiphoton quantum transitions, the average number of atoms in the cavity is increased to $\bar{N} = 4.2$ so that semiclassical approximations are valid. The sequence of transitions $|0\rangle \xrightarrow{\omega_{pump}} |+\rangle \xrightarrow{\omega_{probe}} |1\rangle$ is probed. One expects the peak of the transmission to decrease from $g_0\sqrt{N}$ (about ω_0) for no pump to $g_0(\sqrt{4N-2} - \sqrt{N})$ with the pump. Fig.4 shows the transmission of the probe beam as the pump intensity is gradually increased from zero. For zero pump intensity, we see the vacuum-Rabi splitting talked about before. As the pump intensity is increased, the strength of the "unpumped" peak decreases while that of the pumped peak increases. At the same time the pumped peak migrates inwards towards the common atom-cavity frequency. In addition, the width of the peak decreases. For a linear system, the probe response would have been independent of the pump beam. Hence fig.4 is a manifestation of the nonlinear dynamics of the atom–cavity system.

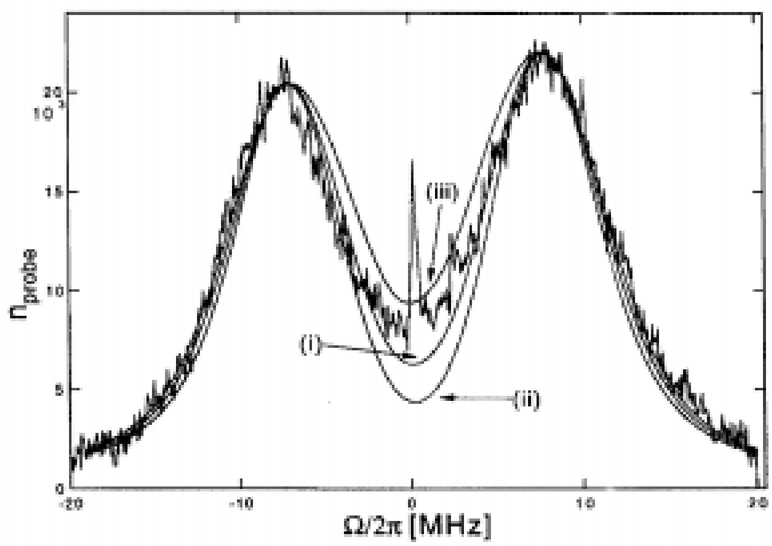

FIG. 3. Linear spectrum for $\bar{N} \approx 1.1$ atoms measured by heterodyne detection shows the vacuum-Rabi splitting at $\omega = \pm g_0$ (from Ref. [7]).

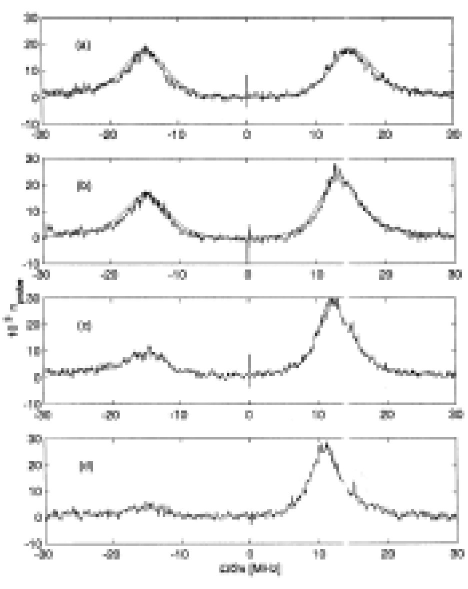

FIG. 4. Sequence of probe spectra for $\bar{N} \approx= 4.2$ atoms with $\omega_a = \omega_c \equiv 0$. The frequency Ω of a constant amplitude probe beam is swept and the transmission is recorded. Trace (a) is without a pump beam. From (b) to (d) the pump power is increased (from Ref. [7]).

V. CONCLUSIONS

The interaction of the atoms with the cavity modes in the strong coupling regime has been discussed theoretically and experimentally. The interaction leads to a modification of the energy level structure. The free space resonances are split into doublets by the interaction. The energy levels and transition matrix elements depend strongly on the number of atoms in the cavity. Nonlinear spectroscopy experiments towards these studies have been described briefly.

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