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Anharmonicity of the vacuum Rabi peaks in a many-atom system

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We have experimentally observed the evolution of the vacuum Rabi doublet into a singlet in the transmission spectrum of a cavity filled with a collection of two-level atoms. For very weak excitation the peaks behave like simple harmonic oscillators, but become anharmonic as the excitation increases. The anharmonicity grows to a point where hysteresis appears in the transmission spectrum, eventually causing the two peaks to merge into one. [S1050-2947(96)50811-5]

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The interaction of a single mode of the electromagnetic field with a collection of atoms has been extensively studied in cavity quantum electrodynamics. Since the seminal work of Sánchez Mondragón *et al.* [1], where the term vacuum field Rabi splitting was coined and Agarwal's calculations of the microwave absorption by Rydberg atoms in a cavity [2] were introduced, there have been extensive investigations of the transmitted spectrum of the atoms-cavity system [3–6]. The experiments have been carried out in a regime in which the excitation is extremely weak. This means that the number of energy quanta in the system is very small, much less than 1, so it cannot sustain an appreciable population in the excited state. The weak excitation regime is characterized by the appearance of two peaks in the transmitted spectrum of the composite atoms-cavity system. This doublet is a manifestation of the degeneracy in the frequency of two oscillators being lifted once they are coupled. The rate for the exchange of energy between the two oscillators is precisely the frequency splitting. One oscillator is the single mode of the electromagnetic field, while the other is the collective polarization of the N atoms. Not surprisingly, quantum-mechanical and classical calculations predict exactly the same spectrum. Even when dissipation is taken into account, the doublet is clearly resolved as long as the two oscillators fulfill the following condition: Their decay rates should be approximately equal and smaller than the rate of energy exchange.

The purpose of this Rapid Communication is to report an experimental study of the behavior of an atom-cavity system as the excitation increases, from low to high intensities, away from the linear regime so far explored. We follow the changes in the transmitted spectrum starting with the vacuum Rabi doublet at low intensities. The presence of more energy in the system, and the possibility of coherent exchange of that energy between the atoms and the cavity, modifies the simple harmonic-oscillator structure. The transmitted spectrum presents hysteresis followed by the merging of the distorted peaks into a single peak. This work is an exploration in frequency space of the underlying structure for the dynamics of the atoms-cavity system with arbitrary excitation.

For an elementary theory we may start with the Maxwell-Bloch equations for the atom-cavity system, following the literature in optical bistability [7]. To simplify the discussion, for the present we do not take into account the transverse character of the mode in the cavity, nor the standing waves of the Fabry-Pérot interferometer. We derive the transmitted spectra for arbitrary input intensity from the steady-state solution to the Maxwell-Bloch equations when the frequency of the driving field changes by an amount Ω from the resonance condition. The purely radiative decay rate of the atoms is characterized by $\gamma_\perp$, while the cavity field decays with a rate κ. The dipole coupling between N two-level atoms and the cavity mode is $g\sqrt{N}$, where $g=(\mu^2\omega/2\hbar\epsilon_0 V)^{1/2}$; μ is the transition-dipole moment of the atom, ω the resonance frequency of both atoms and cavity, and V the cavity mode volume. The input field amplitude y and the output field amplitude x are normalized to the square root of the saturation intensity for the atomic transition. They represent the intracavity field in the absence and presence of atoms, respectively.

Guided by the behavior of the system in the low-intensity regime, where there are clearly two normal modes, we write the transmission as consisting of two parts:

$$\left|\frac{x}{y}\right|^2=\left|\frac{A}{i\Omega+\Omega_1}+\frac{B}{i\Omega+\Omega_2}\right|^2, \tag{1}$$

where

$$A=\kappa\frac{\gamma_\perp+\Omega_1}{\Omega_1-\Omega_2}, \tag{2}$$

$$B=\kappa\frac{\gamma_\perp+\Omega_2}{\Omega_2-\Omega_1}, \tag{3}$$

and

$$\Omega_{1,2}=-\frac{\kappa+\gamma_\perp}{2}\pm i\sqrt{-\left(\frac{\kappa-\gamma_\perp}{2}\right)^2+\frac{g^2N}{1+\dfrac{\gamma_\perp^2x^2}{\gamma_\perp^2+\Omega^2}}}\,. \tag{4}$$

In the limit of very low excitation, $|x|^2\ll 1$, where the system behaves like two coupled harmonic oscillators, $\Omega_{1,2}$ are the

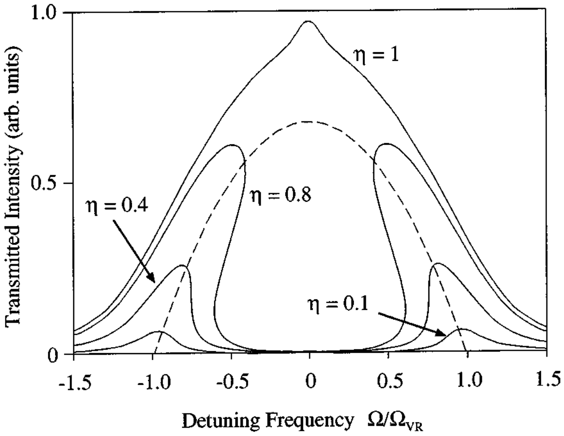

FIG. 1. Transmitted intensity of the atoms-cavity system as a function of frequency Ω (measured in units of the vacuum Rabi frequency Ω_{VR}). The curves correspond to different input intensities $\eta = I_{in}/I_{max}$, scaled to the highest peak. We show features reached by scanning Ω from below to above atomic resonance. The dashed line follows the condition for zero phase shift between the input and output fields.

eigenvalues of the linearized Maxwell-Bloch equations. As the excitation increases, the oscillators become anharmonic and their frequencies develop a dependence on the strength of the excitation. The anharmonicity grows to the point where the vacuum Rabi peaks are deformed [9] to produce regions where the resonances are three-valued functions, with one of the values unstable.

Figure 1 plots Eq. (1) for different values of the incident field. For the lowest excitation, the peaks are the vacuum Rabi doublet. As the excitation increases, the anharmonicity becomes evident. The peaks eventually touch at the atomic resonance frequency, giving rise to the single peaked transmission spectrum. There is an interesting region in the transmitted spectrum where some of the steady-state solutions of the Maxwell-Bloch equations are unstable and hysteresis occurs as the frequency of the exciting laser varies. The hysteresis creates a cave in parameter space where the lower branch of intensity optical bistability resides. As the doublet evolves into a singlet, the linewidths of the transmission peaks change from the average of the two decay rates at low excitation [3] to a broad and complex feature where the two peaks first merge [1]. Finally, for very high excitation, $|x|^2 \gg 1$, the linewidth returns to that of an empty cavity. For Fig. 1 we have chosen the cavity decay rate and the atomic decay rate to be equal. This situation is similar to the experiment.

In order to gain more intuition about the system, we follow the work of Zhu *et al.* [5] and present in Fig. 1, with the dashed line, the locus obtained by demanding that the phase of the transmitted field be equal to the phase of the incident field. This simple condition guides the position of the anharmonic peaks remarkably well and links their evolution to classical optics. From a purely phenomenological point of view, one can argue that the transmitted spectrum in the high-intensity regime should be a singlet, since the atoms are then saturated and do not contribute significantly to the dynamics of the system.

Anharmonicity is also expected from a quantum-mechanical analysis. When analyzing the interaction of one atom with a single mode of the electromagnetic field using the dressed-state formalism (see, for example, Carmichael *et al.* [8]), it is clear that the spacing of the excited states is not equal, but decreases by an amount that scales inversely with the square root of the number of energy quanta available to the system. For one atom, significant nonlinearity is achieved with relatively low numbers of quanta. The generalization to the case of many atoms follows from the results of Tavis and Cummings [10], where the spacing of the allowed excited-state transitions again decreases as the number of excitations increases; but now more photons are needed to turn on a significant nonlinearity.

We performed the measurements with our existing apparatus [11]. It consists of a high finesse Fabry-Pérot cavity intersected at 90° by a collimated beam of optically prepumped ^{85}Rb atoms. We operate on the cycling transition ($5S_{1/2}$, $F=3$, $m_F=3 \rightarrow 5P_{3/2}$, $F=4$, $m_F=4$) at 780 nm. The resonator is formed by two spherical mirrors of 7.5-cm radius with a separation of 2 mm. The finesse of the cavity is $12\,000 \pm 1500$. The exciting laser is mode matched to the TEM$_{00}$ mode of the cavity. The parameters of the experiment, $(\kappa, \gamma_\perp, g) = (3.1 \pm 0.4, 3.1, 2.5 \pm 0.3) \times 2\pi \times 10^6$ rad/s, place it in the intermediate regime, between weak and strong coupling, of cavity quantum electrodynamics.

Each data collection cycle consists of two parts. First we actively lock the cavity on resonance, with the atomic transition using an intense auxiliary beam with 12-MHz FM sidebands for 300 ms. Next a mechanical chopper blocks the auxiliary beam. 5 ms later we start to scan the frequency of a probe beam. For the duration of the scan we direct the output of the cavity into a photomultiplier tube with an acousto-optical modulator switch. We scan, increasing and decreasing the frequency of the laser to cover the region of interest six times. Simultaneously, we record a saturation spectrum from a Rb cell that serves as diagnostic and frequency marker. The cycle ends after 360 ms, and we return to actively locking the cavity. During a scan the intensity of the exciting laser is constant to better than 7%.

With a well developed hysteresis in the input-output steady state of the system [11], we proceed to study the transmitted spectrum as a function of the input intensity. Results are shown in Fig. 2. On resonance, there are on the average 0.01 photons inside the cavity at the lowest input intensity (trace j), and 11 000 at the highest (trace a). From the frequency of the vacuum Rabi splitting, the number of atoms in the cavity mode is estimated to be about 220; we therefore expect a semiclassical analysis of the experiment to be appropriate. Traces a to c correspond to the upper branch of the bistable region. At the highest power recorded (trace a), the linewidth is less than 12 MHz. Between traces c and d the system ceases to be bistable on resonance, but a bistability exists in the vicinity of the vacuum Rabi peaks. There is a significant linewidth broadening, by a factor of 2, as the input power decreases towards the switching point of the hysteresis cycle in intensity; in trace c, at the closest point before the doublet first appears, the linewidth has widened to 26 MHz. The broadening is a manifestation of the coherent interplay of the atoms and the cavity [1]. The rest of the traces (d to j) are on the lower branch of on resonance (absorptive) optical bistability [7], in a region where that is the only available state of the system. The asymmetry in the

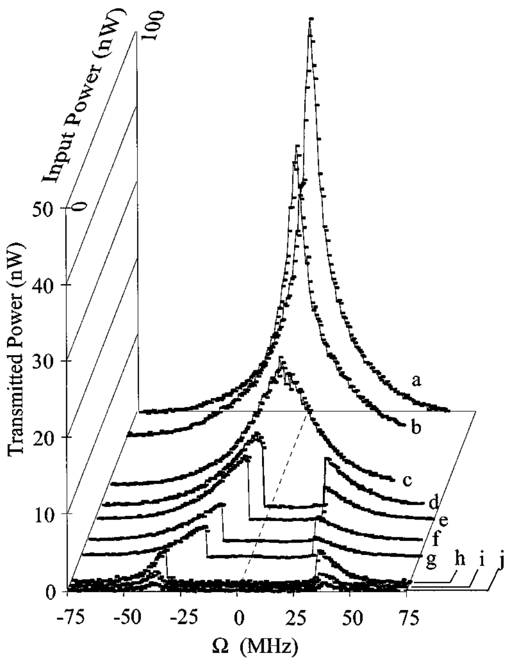

FIG. 2. Transmitted spectrum as a function of frequency for different excitation powers. The height of traces *h*, *i*, and *j* has been multiplied by a factor of 5. The frequency is scanned from below atomic resonance to above resonance.

location of the peaks and in their line shapes shows the behavior of anharmonic oscillators. The doublet in the spectrum recorded with the lowest intensity had a full width at half maximum of 7 MHz, close to the average of the two decay rates.

The theory outlined above must be modified in order to make quantitative comparisons with the data. We have carried out the calculation of the transmission function, taking into account the Gaussian transverse mode of the field as well as the standing waves in the cavity. Figure 3 presents the position of the highest point of the doublet as a function of the input power. Figure 4 shows the hysteresis observed on the transmitted power as a function of frequency, and compares it with theory. The uncertainties in the input and output power measurements, as well as the effects of residual Doppler and transit broadening, do not permit a quantitative comparison to better than a factor of 2. The calculated input and output powers for Figs. 3, 4 have thus been scaled by a factor of 1.8 to optimize the fit. It is important to note that the hysteresis is present for a lower value in the input intensity than that necessary for absorptive optical bistability. The threshold for resonant absorptive bistability is marked precisely by the appearance of a single peak in the transmitted spectrum.

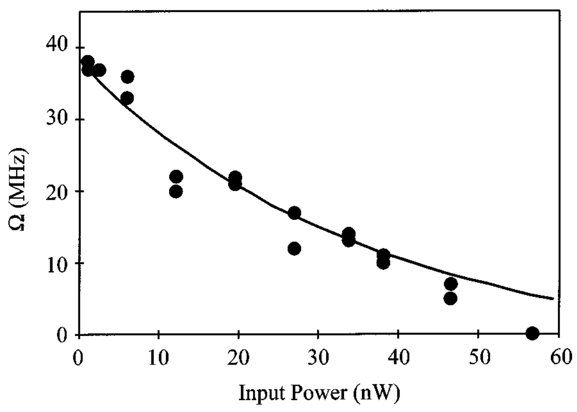

FIG. 3. Sideband frequency of the maxima in the doublet as a function of normalized input intensity. The solid line is the theoretical prediction.

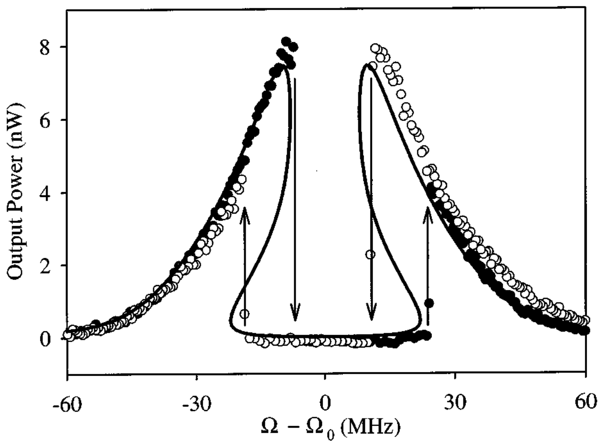

FIG. 4. Hysteresis of the transmitted light from the coupled atoms-cavity system as a function of exciting frequency. Two different scans with equal input intensities are shown. Crosses mark the scans with increasing laser frequency and circles with decreasing laser frequency. The lines are theoretical calculations.

We have analyzed the system in terms of the semiclassical Maxwell-Bloch equations, which is appropriate for a large number of atoms. It is important to contrast the observed behavior with that predicted in the fully quantum regime, where the semiclassical analysis is invalid. The quantum-mechanical formulation is made in a density-matrix approach. It establishes a one-to-one correspondence between operator expectation values and the variables of the Maxwell-Bloch equations so long as the atom-field correlations are neglected. For many atoms this decorrelation is a good approximation. For one atom it is not. A quantum-mechanical calculation for one atom [8] predicts quite different behavior from what is seen in the experiment. Figure 5 makes the comparison for $g/\kappa = g/\gamma_\perp = 10$, similar to the ratio of vacuum Rabi frequency to decay rates in Fig. 4. The frequency bistability is necessarily absent from the one-atom result since this gives the true *mean* photon number. More importantly, in the one-atom case the nonlinearity takes the form of additional two-photon resonances in between the vacuum Rabi peaks. These resonances demonstrate the existence of discrete, entangled states of the atom and cavity field, and represent direct two-photon absorption up the Jaynes-Cummings ladder. For sufficiently small linewidths many such multiphoton resonances may be resolved [8].

It is important to emphasize that currently no experiment has observed these deviations from semiclassical spectroscopy [15]. This statement is of particular relevance to work with semiconductor microcavities, where the evolution of the vacuum Rabi doublet has been the subject of recent investigations [12–14]. In semiconductor systems, although the transmitted spectrum at low intensity shows the same

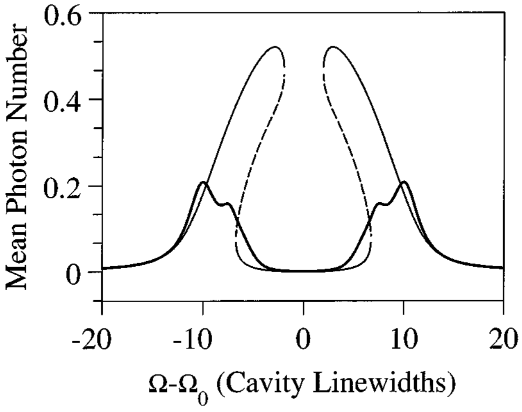

FIG. 5. Mean intracavity photon number calculated quantum mechanically for one atom (bold curve) with $g/\kappa = g/\gamma_\perp = 10$, compared with the solution to semiclassical Maxwell-Bloch equations (N atoms) with $\sqrt{N}g/\kappa = \sqrt{N}g/\gamma_\perp = 10$.

vacuum Rabi splitting, the underlying mechanism is different. Excitons replace the atoms as the second oscillator, and their bosonic character preserves harmonic behavior to very high levels of excitation. The dressed picture is of an exciton-polariton whose energy levels are approximately equally spaced. An analogy might be made with a many-atom system (many more than 220 atoms), but certainly not with one atom. There is now much interest in the high-intensity limit for the exciton-polariton [14] where the exciton-exciton interactions begin to play a role. Here the physical interpretation has been controversial, although a semiclassical nonlinear response is certainly what one would expect. Our results, together with Fig. 5, demonstrate the kind of comparison that can resolve the debate [16].

Another example where a hysteresis is linked to the change in the energy-level spacing of an oscillator is the motion of a single electron in a Penning trap [17]. The change in this case comes from the relativistic mass of the electron as it increases its cyclotron energy.

Finally, it is important to note a further connection between the semiclassical hysteresis and multiphoton processes. It is precisely these processes that can give rise to squeezing, as observed in this system [18]. Without them it would have been impossible to generate the correlated pairs necessary for reduction of the noise beyond the quantum limit.

In summary we have observed the evolution of the vacuum Rabi doublet into a singlet in a many-atom system. The mechanism for the transition is the nonlinearity brought in by the saturation of the two-level atoms. The anharmonicity observed is the semiclassical counterpart of the multiphoton processes resonances in the energy-level structure of an atom-cavity system that allow for the generation of nonclassical states of the electromagnetic field in this system.

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