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Decay of bichromatically driven atoms in a cavity

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The decay characteristics of two-state Rydberg atoms are investigated in a microwave cavity driven simultaneously by two strong rf fields. Tuning one of the driving fields leads to sharp resonances in the decay rate of the upper atomic level. Their width may become smaller than the cavity linewidth and transit time broadening. We present a theoretical model, interpreting the resonances as cavity-induced decay of Floquet states of the composite atom-field system. The observed phenomena arise from the interplay of cavity-modified Bloch-Siegert shifts and dynamic enhancement of spontaneous emission at single-photon and multiphoton resonances.

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Atoms driven by a strong electromagnetic field have played a major role in the study of the interaction of light and matter. A prominent example is the fluorescence spectrum of two-level atoms subjected to an intense laser field. It is characterized by the well-known triplet structure first predicted by Mollow [1] and observed experimentally [2,3]. An intriguing extension of single-field resonance fluorescence is the interaction of atoms with a strong bichromatic driving field. Several recent experiments have demonstrated new physical effects in such a system. The total fluorescence rate displays resonances at subharmonics of the Rabi frequency [4,5], and also the spectral structure of the scattered light fundamentally differs from the monochromatic case [6–8]. The observations are most elegantly interpreted in terms of transitions between Floquet states [9] of the driven atom. These states provide a description where the two-level system with time-dependent fields is transformed to an infinite set of stationary *quasienergy* levels contributing to the atomic transition.

Previous experiments on bichromatic driving were performed with atoms in free space. Major modifications of the system behavior are expected when the atom is placed in a cavity. Under monochromatic driving, the resonant structure of the density of states in a cavity was shown to cause dynamic effects such as vacuum field dressed state pumping [10] and suppression of spontaneous emission [11,12]. In this paper we present an experimental and theoretical investigation of cavity-modified dynamics of two-level atoms driven by a strong bichromatic field. Such a system exhibits new phenomena due to the interplay of cavity enhancement of radiative transitions and intensity-induced modification of the atomic levels close to the cavity resonance.

The experiment uses Rydberg atoms in a low order microwave cavity with a moderate quality factor Q. The system behavior is well described in terms of cavity-modified decay. We demonstrate that the spontaneous transition rate intricately depends on the intensity and detuning of the two components of the microwave field injected into the cavity. Enhanced decay appears in narrow resonance zones in parameter space. The width of these resonances is essentially determined by the strong frequency dependence of the spectral density of the cavity and can become much smaller than the cavity linewidth itself. Structures narrower than the cavity mode have not been observed with cavity-induced transitions up till now.

A theoretical model of our experiment describes the atom-field system in terms of a Floquet expansion [9] and a suitably adopted master equation. The numerically determined cavity-induced transition rates between Floquet states agree well with the behavior observed experimentally.

The experimental apparatus is shown in Fig. 1. It is based on the setup used to investigate cavity effects under monochromatic driving [12]. A collimated beam of rubidium atoms is prepared in the $53\,^2P_{3/2}$ state of ^{85}Rb by three-step excitation using diode lasers. Subsequently, the atoms traverse a cylindrical niobium cavity along its axis. The mean thermal speed of the atoms leads to a cavity transit time $\tau = 97$ μs.

The TM_{020} mode of the cavity is tuned near the $53\,^2P_{3/2} \rightarrow 53\,^2S_{1/2}$ transition at $\omega_0 = 2\pi \times 25.59003$ GHz. As only transitions with $\Delta m = 0$ couple to the linearly polarized cavity mode, the two-level atom approximation is applicable. The atom-field coupling constant between the $|m| = 1/2$ magnetic substates is $g = 2\pi \times 17 \times 10^3$ s^{-1}.

At a temperature of 4.2 K, the superconducting cavity has a loaded quality factor $Q = 1.9 \times 10^6$, corresponding to a cav-

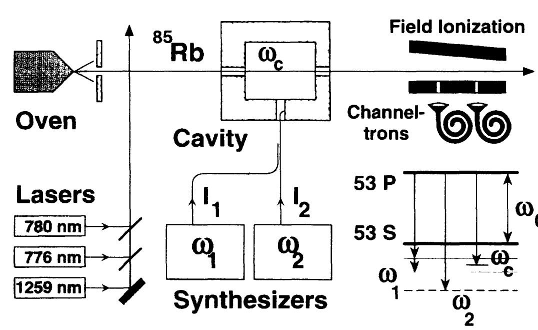

FIG. 1. Experimental setup. The inset shows the unperturbed atomic levels involved. The driving field at frequency ω_2 has fixed detuning. The cavity ω_c is tuned halfway between ω_2 and the atomic frequency ω_0. Spectra are taken by sweeping the field ω_1 across ω_c (shaded region).

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ity decay rate $\kappa = 2\pi \times 6.7 \times 10^3\ \mathrm{s}^{-1}$. It is thus of the same order as the coupling constant g. The cavity field is linearly polarized with the electric field vector parallel to the direction of the atomic beam. There are $\bar{n} = 2.9$ blackbody photons in the cavity mode, causing thermally induced transitions in addition to spontaneous decay.

The bichromatic driving field is generated by two microwave synthesizers with linewidths of 1 kHz and 2 Hz, respectively. It should be noted that, in contrast to many optical experiments, the tuning and intensity of the two injected fields may be controlled independently. The fields are coupled to the cavity through a hole in the sidewall, equidistant from the two end caps.

A fundamental difference between the setup described here and fluorescence of atoms in optical cavities is that the microwave resonator we employ is closed on all sides, so that atoms interact exclusively with a single cavity mode. Therefore, cavity-related effects may be observed unperturbed by decay into side modes. The closed-cavity geometry makes it necessary to employ a special scheme for spectral analysis of the system since the fluorescence intensity emitted to the side, which is usually monitored in optical experiments, is absent and atomic fluorescence into the cavity mode is much too weak to be observed directly. In the setup presented here, information on the atom-field interaction is obtained by measuring the occupation of the atomic levels after the Rydberg atoms have left the cavity. This is accomplished by state-selective field ionization using two secondary electron multipliers with an efficiency of about 10%.

The spectral structure of the system is investigated by varying the parameters of the external driving fields, thus changing the ac Stark shifts of the levels. Spectra are recorded by scanning the frequency of only one field (called ω_1), while all other parameters are kept fixed, specifically the frequency of the second field (ω_2), the injected intensities of both external fields, and the cavity tuning. Note, however, that there is an inherent variation of the *intracavity* amplitude of the scanned field, since coupling to the cavity mode depends on detuning and follows the Lorentzian cavity line shape. Atoms are injected into the cavity in the upper Rydberg state ($53\,P$). Atomic decay induced by the resonator mode serves as a fixed probe of the atom-cavity system. It is monitored by counting the number of atoms leaving the cavity in the lower state ($53\,S$).

A typical spectrum recorded in this way is shown in Fig. 2. The baseline corresponds to 5% of the atoms leaving the cavity in the lower level. This is an order of magnitude less than expected for cavity-enhanced transitions with no external fields applied. Such an inhibition of decay in the presence of strong driving is due to dynamically suppressed emission reported previously [12].

A striking new feature in the spectra obtained with a bichromatic driving field is pronounced peaks in the number of atoms leaving the cavity in the lower state. By analogy with the dynamically suppressed emission outside these resonance zones, they must be attributed to dynamically enhanced decay. It is noteworthy that the transition from inhibited to enhanced decay happens on a scale much smaller than the cavity linewidth κ.

Some properties of the resonances in Fig. 2 should be pointed out. First, the lines appear in pairs, approximately

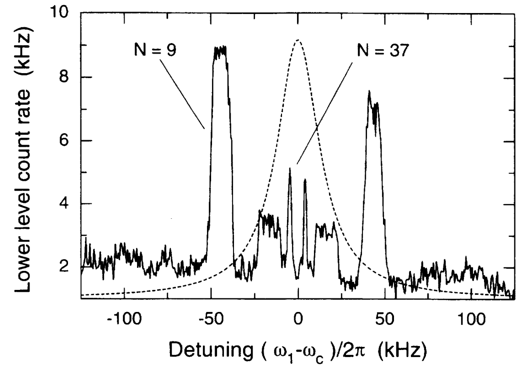

FIG. 2. Count rate in the lower level ($53\,S$) as a function of the driving frequency ω_1. The peaks correspond to cavity-enhanced decay at multiphoton resonances. The dashed curve marks the cavity resonance. The experimental parameters are: $\omega_c - \omega_0 = 2\pi \times 937$ kHz, $\omega_2 - \omega_0 = 2\pi \times 1300$ kHz, $I_1 = -30$ dBm, $I_2 = -11.6$ dBm. The order N of the multiphoton transition, obtained from numerical simulations, is indicated.

symmetric to the cavity resonance frequency ω_c. The inner pair of resonances has the smallest linewidth. Next to them are some weaker resonances which are not completely resolved. Finally, the two strong outer lines have the largest width. Here, the occupation of upper and lower level reaches thermal equilibrium, indicating that thermalization by cavity-induced transitions is faster than the transit time.

Scans of ω_1 were repeated at different intensities of the driving field components at ω_1 and ω_2, and the resonance frequencies were evaluated. The observed shift of the peaks is the cavity-modified analogue of the Bloch-Siegert shift known from magnetic resonance [13]. Results for the strongest line, observed over a range of two orders of magnitude of the injected intensity, are compiled in Fig. 3. Increasing the intensity of the scanned field (ω_1) shifts the resonance peaks away from the cavity frequency. On the other hand, when the intensity of the fixed field (ω_2) is increased, a reduction of the splitting between the peaks is observed. The fact that the peak positions depend on the power of both injected fields indicates that the dynamics is governed by

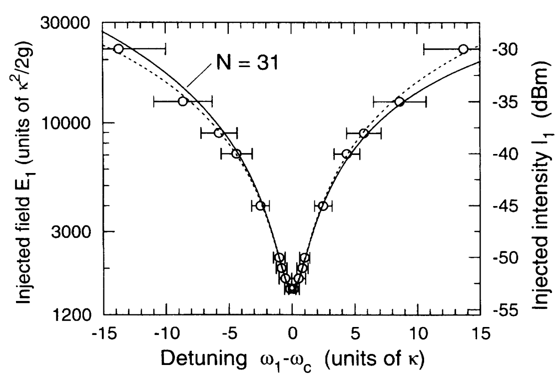

FIG. 3. Observed shift of the strongest resonance as a function of the intensity of the driving field ω_1. The solid line is the theoretical result obtained from the Floquet analysis of the system. The dashed line corresponds to the empirical condition (1). Parameters: $\omega_c - \omega_0 = 75.5\kappa$, $\omega_2 - \omega_0 = 189\kappa$, $2gE_2/\kappa = 1.05 \times 10^5 \kappa$.

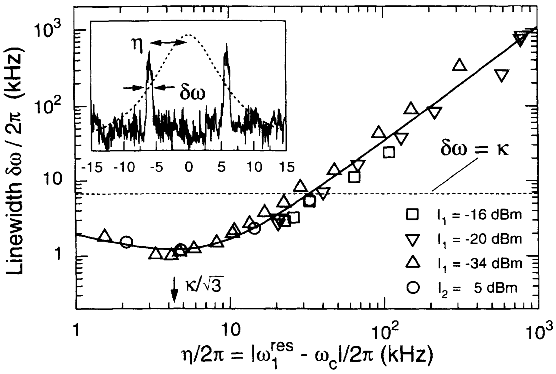

FIG. 4. Observed linewidth $\delta\omega$ as a function of the detuning from the cavity frequency. For all parameters investigated, the width is proportional to the square root of the inverse slope of the Lorentzian cavity response function (solid line) and can drop below the cavity width κ (dashed line).

bichromatic effects and none of the fields may be regarded as weak. The dashed line in Fig. 3 represents an empirical fit to the data. We found that the resonances lie on a curve well described by the condition that the Rabi frequencies Ω_1 and Ω_2 of the two fields inside the cavity have a constant ratio:

$$\Omega_1/\Omega_2=f. \tag{1}$$

The parameter f is of the order of unity and the intracavity Rabi frequencies are given by

$$\Omega_k=\left|\frac{2gE_k}{\kappa+i(\omega_c-\omega_k)}\right|, \quad k=1,2, \tag{2}$$

where E_1 and E_2 denote the strengths of the injected fields, and the Lorentzian line shape of the cavity mode is taken into account.

Another remarkable feature of the resonances is their sharpness. In Fig. 4, the width of the peaks is evaluated as a function of their detuning from the cavity frequency for a large range of different experimental parameters. All data lie close to a unique curve which is given by the square root of the inverse slope of the Lorentzian cavity response function. When the cavity slope is steepest, we observe the narrowest lines with a width of only 1 kHz, as shown in the inset of Fig. 4. This is a factor of almost 10 below the cavity decay rate κ and the transit time broadening τ^{-1}.

We are able to explain the essential experimental results in the frame of a theoretical model based on the Floquet analysis of the atom-field system. In the following, we sketch a theory of cavity-induced transitions between atomic Floquet states in a bichromatic field.

The Hamiltonian of an atom interacting with two classical fields ω_1 and ω_2 in a frame rotating at the average frequency $\omega_l=(\omega_1+\omega_2)/2$ is

$$H(t)=\hbar\Delta_l S^z+\frac{\hbar}{2}\,e^{-i\nu t}(S^+\Omega_2+S^-\Omega_1^*)+\text{H.c.}, \tag{3}$$

where $\Delta_l=\omega_0-\omega_l$, $\nu=(\omega_2-\omega_1)/2$, and S is the spin-1/2 angular momentum operator describing the two-level atom.

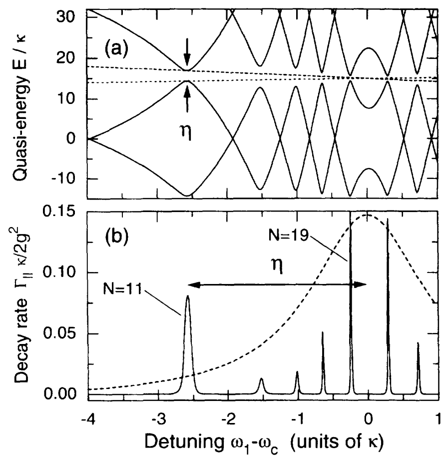

FIG. 5. (a) Part of the quasienergy spectrum of a two-level atom in a cavity subjected to bichromatic driving. Close to the dashed line, transitions to other Floquet levels are resonant with the cavity. Parameters: $\omega_c-\omega_0=140\kappa$, $\omega_2-\omega_0=200\kappa$, $2gE_1/\kappa=500\kappa$, $2gE_2/\kappa=15972\kappa$. (b) Theoretical decay rate of the population inversion in the Floquet basis. When the cavity is at resonance with a Floquet transition at an avoided crossing ($|\omega_1^{\text{res}}-\omega_c|=\eta$), strong decay occurs.

Since the Hamiltonian $H(t)$ is periodic in time, Floquet's theorem [9] applies. It states that the solution $|\Psi(t)\rangle$ of the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle=H(t)|\Psi(t)\rangle \tag{4}$$

may be expanded in a series

$$|\Psi(t)\rangle=e^{-iEt}\sum_{n=-\infty}^{\infty}(\alpha_{1n}|1\rangle+\alpha_{2n}|2\rangle)e^{-in\nu t} \tag{5}$$

with stationary coefficients α_{kn}, $|k\rangle$ being the unperturbed atomic states. Using expansion (5), we numerically solved Eq. (4) by standard matrix continued fraction methods. The spectrum of eigenvalues, denoted *quasienergy levels*, is

$$E=\pm\epsilon+n\nu, \quad n=0,\pm1,\pm2,\ldots, \tag{6}$$

ϵ being the smallest positive eigenvalue. Part of a typical Floquet spectrum is shown in Fig. 5(a) as a function of the detuning of the first field. Note the avoided crossings of the Floquet levels when ϵ approaches $\nu/2$, and the varying slope of the levels as ω_1 is tuned across the cavity line.

Concentrating on the first Brillouin zone $[-\nu/2,\nu/2]$, we can use the eigenfunctions $|\Psi_\pm(t=0)\rangle$ of the eigenvalues $\pm\epsilon$ as basis states of the system. They depend on the intensities and detunings of the driving fields. The quantity relevant to the interpretation of our experimental results is the cavity-induced rate of decay of the population difference between $|\Psi_+\rangle$ and $|\Psi_-\rangle$, which, by analogy with longitudinal Bloch vector relaxation, we call $\Gamma_\parallel$.

We derive an expression for $\Gamma_\parallel$ by starting from the master equation of an atom in a lossy single-mode cavity at a finite temperature. Subsequently, we apply the Born approximation to second order in g to adiabatically eliminate the cavity mode variables, which is possible in the moderate Q

regime [14]. The method is analogous to the treatment of a monochromatically driven cavity employed in Ref [15]. The final result obtained is

$$\Gamma_{\parallel}=2g^2(1+2\bar{n})\ \mathrm{Re}\sum_{n,m,p,q}\left(\frac{\alpha_{2n-}\alpha^*_{1m+}\alpha^*_{2p-}\alpha_{1q+}}{\kappa+i\delta-2i\epsilon+i\nu(p-q)}+\frac{\alpha_{2n+}\alpha^*_{1m-}\alpha^*_{2p+}\alpha_{1q-}}{\kappa+i\delta+2i\epsilon+i\nu(p-q)}\right)\delta_{n-m,p-q}, \tag{7}$$

where $\alpha_{kn\pm}$ are the eigenvector components corresponding to the quasienergies $\pm\epsilon$ and $\delta=\omega_c-\omega_l$. Through ϵ and $\alpha_{kn\pm}$, the decay rate $\Gamma_{\parallel}$ strongly depends on intensity and detuning of the driving fields. This can be regarded as a consequence of non-Markovian dynamics, induced by the finite response time of the cavity mode.

Evaluating $\Gamma_{\parallel}$ numerically, we obtain agreement with all essential features of the experimental spectra. In particular, from Eq. (7), strong decay to the lower level is expected when the coupling between $|\Psi_+\rangle$ and $|\Psi_-\rangle$, given by the product of eigenvector components in the numerator, is large. From the theory of multiple-quantum resonances in free space [13] it is known that the transition probability peaks when the slope of the quasilevels E becomes zero, that is, when the levels undergo an avoided crossing. In the limit of weak driving fields, the corresponding resonances are found at $\Delta_l=N\nu$, where N is an odd integer.

In the strong driving situation considered here, the resonances are subject to large Bloch-Siegert-like shifts [13,16], which are strongly increased by the presence of the cavity. Still, the peaks may be labeled by the number N of driving field photons contributing to the transition, as indicated in the figures. In order to compare the Floquet theory with the experimentally observed shifts, the maxima in the decay rate $\Gamma_{\parallel}$ were calculated. The result obtained is represented by the solid line in Fig. 3, showing good agreement with the empirical condition (1).

However, not only does the cavity shift the position of the observed resonances, a more dramatic effect of the cavity is that it determines which multiphoton resonances lead to atomic decay. The stringent condition for decay to occur is that a frequency component of the $|\Psi_+\rangle$ to $|\Psi_-\rangle$ transition must coincide with the cavity frequency. Therefore

$$\delta=\nu P\mp2\epsilon \quad \text{with} \quad P=p-q=0,\pm2,\pm4\ldots \tag{8}$$

must hold, ensuring that the resonance denominator in Eq. (7) becomes minimal. In the Floquet diagram in Fig. 5(a), this condition is represented by straight dashed lines. Evidently, only a few multiphoton resonances meet condition (8) and are dynamically enhanced, while all other transitions are suppressed by the cavity. This is illustrated in Fig. 5(b), where $\Gamma_{\parallel}$ is shown below the respective Floquet energies. The resulting spectrum is in keeping with the experimental data presented in Fig. 2.

If ω_1 is tuned close to the cavity, enhanced decay occurs mainly at $\delta=2\epsilon$ and $\delta=2(\nu-\epsilon)$. In this case a simple relation between the resonance frequencies and the quasienergies holds: The level splitting $\eta=\nu-2\epsilon$ at the avoided crossing is equal to the detuning $|\omega_1^{\mathrm{res}}-\omega_c|$, so that η may be determined directly from the ω_1 spectra (c.f. Fig. 5).

The observed width of the resonances, too, has a natural interpretation in terms of Floquet transitions. The cavity modifies the structure of the Floquet levels. As a consequence, the width of an avoided crossing as a function of the control parameter ω_1 is found to be inversely proportional to the square root of the slope of the cavity line. Since the linewidth of the resonances is determined by the width of the avoided crossing regions, the peaks narrow when the local slope of the cavity response function increases. The minimum width should occur when ω_1^{res} is detuned from ω_c by $\kappa/\sqrt{3}$, in accordance with the experimental observation (Fig. 4). When higher-order multiphoton transitions contribute to a resonance, a further reduction of the linewidth is expected, even below the resolution of the microwave source we employ.

In closing the paper, it must be stressed that the composite system of atom, cavity, and driving fields analyzed here is very different from an atom in free space. The observed resonance frequencies are governed by Bloch-Siegert shifts and depend only weakly on the atomic transition frequency. In fact, varying the atomic frequency ω_0 in our theoretical calculations by as much as 50κ has a much smaller effect on the positions of the resonances than changing the multiphoton order N.

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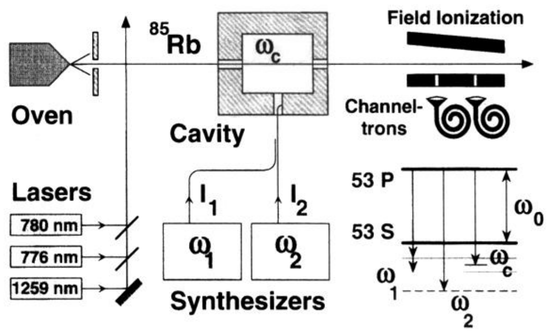

FIG. 1. Experimental setup. The inset shows the unperturbed atomic levels involved. The driving field at frequency ω_2 has fixed detuning. The cavity ω_c is tuned halfway between ω_2 and the atomic frequency ω_0. Spectra are taken by sweeping the field ω_1 across ω_c (shaded region).