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Observation of linewidth narrowing due to a spontaneously generated coherence effect*

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We investigate the resonance fluorescence spectrum of an atomic three-level ladder system driven by two laser fields. We show that such a system emulates to a large degree a V-type atom with parallel dipole moments—the latter being a system that exhibits spontaneously generated coherence and can display ultrasharp spectral lines. We find a suitable energy scheme in a ^{85}Rb atom and experimentally observe the narrowing of the central peak in a rubidium atomic beam. The corresponding spectrum can convincingly demonstrate the existence of spontaneously generated coherence.

Keywords: spontaneously generated coherence, resonance fluorescence, linewidth narrowing, atomic beam

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1. Introduction

Resonance fluorescence refers to the detection of an atom in open space by means of resonant absorption and re-emission of electromagnetic waves. The resonance fluorescence spectrum of a two-level atom (TLA) driven by a strong near-resonant monochromatic field was predicted[1] and observed.[2–4] On the basis of this result, resonance fluorescence from a semiconductor quantum dot in a cavity,[5] a spin-resolved quantum-dot,[6] a single molecule,[7] and a single artificial atom[8] have been reported recently. In addition, various methods have been proposed to modify the spectrum of resonance fluorescence, such as via laser fields,[9–11] the squeezed vacuum,[12] the influence of radiative decay,[13] and the magnetic fields.[14] The spectrum of resonance fluorescence can also be changed by spontaneously generated coherence (SGC).

Spontaneously generated coherence refers to the interference of two decay channels with nonorthogonal electric-dipole transition matrix elements. It gives rise to a variety of novel quantum effects, such as gain without inversion,[15–17] slowing down the light pulses,[18] enhanced Kerr nonlinearity,[19,20] two-photon correlation,[21] coherent population transfer[22] and trapping,[23] and a photonic band-gap structure.[24–26] In particular, previous results have shown that SGC may be greatly enhanced by using left-handed materials[27] and placing an atom with two closely lying levels near plasmonic nanostructures.[28] In the presence of SGC, the spectrum of resonance fluorescence has been demonstrated to exhibit narrowing,[29,30] quenching,[31] and squeezing characteristics.[32,33]

To testify the existence of SGC, an experiment of spontaneous emission in sodium dimer was reported.[34] However, another experiment[35] in a similar system failed to reproduce the same results. The truth is that it is very difficult to find a real atomic system to experimentally demonstrate the effect of SGC, because SGC exists only in atoms which have

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two close-lying levels subject to the conditions that these levels are near degenerate and the corresponding dipole matrix elements are nonorthogonal. In order to observe the phenomena based on SGC in atomic systems beyond these rigorous conditions, a few methods have been proposed to simulate this intriguing effect. Agarwal[36] has suggested working in such situations where the vacuum of the electromagnetic field is anisotropic. Ficek and Swain[37] have shown the simulation of SGC with the coupling of a DC field. SGC has also been studied in dressed states of a laser field,[38,39] and a microwave field.[40,41] Most recently, Li *et al.*[42] have suggested an experiment to test the effect of quantum interference because of energy shifts on the emission spectrum.

In this paper, we investigate the spectrum of fluorescence in a three-level ladder system driven by two laser fields. The coherent upper field can drive the coupled levels into superpositions of dressed states, and the SGC between these two close-lying levels can produce the narrowing of the spectrum. And in the dressed-state representation, this system is similar to the one studied by Zhou and Swain,[29,30] where ultrasharp spectral lines may be produced due to the SGC effect. We also explain the spectral features in terms of the dressed-atom model of the system. Fortunately we find a suitable energy scheme in ^{85}Rb atoms and carry out the experiment which can show the effect of SGC and avoid all rigorous requirements. Our observations of narrowing of the spectrum in a rubidium atomic beam are in agreement with the calculations of the fluorescence spectrum expected in our scheme and experimentally support the existence of SGC.

2. Theoretical analysis

The three-level V-type system driven by a single-mode laser field $\omega_{\rm L}$ coupled to both transitions is shown in Fig. 1(a). The system consists of two nondegenerate excited levels $|\tilde{1}\rangle$ and $|\tilde{2}\rangle$ separated from the ground level $|\tilde{0}\rangle$ by transition frequencies $\omega_{\tilde{1}}$ and $\omega_{\tilde{2}}$, and connected by the electric dipole moments $\mu_{\tilde{1}}$ and $\mu_{\tilde{2}}$, respectively. $\gamma_{\tilde{i}}$ is the spontaneous decay constant of the excited sublevel $|\tilde{i}\rangle$ $(i=1,2)$ to the ground level $|\tilde{0}\rangle$, whereas direct spontaneous transitions between the excited sublevels are dipole forbidden. The effects of quantum interference are very sensitive to the orientations of the atomic dipole polarizations. For example, if $\mu_{\tilde{1}}$ is parallel to $\mu_{\tilde{2}}$, then $\gamma_{\tilde{1}\tilde{2}}=\sqrt{\gamma_{\tilde{1}}\gamma_{\tilde{2}}}$ and the interference effect is maximal, while if $\mu_{\tilde{1}}$ is perpendicular to $\mu_{\tilde{2}}$, then $\gamma_{\tilde{1}\tilde{2}}=0$ and the quantum interference disappears. When the dipole moments are nearly parallel, ultrasharp spectral lines may be produced in the resonance fluorescence,[20] which is due to quantum interference between the two transition pathways.

As we mentioned above, it is very difficult to find the V-type system with SGC in a real atomic system. So we propose an alternative system which is experimentally attainable and has the effect of SGC. The three-level ladder system under consideration is shown in Fig. 1(b). The laser field ω_1 connects the transition of level $|2\rangle$ to level $|1\rangle$ with Rabi frequency $\varOmega_1$, while the laser field ω_2 connects the transition of level $|3\rangle$ to level $|2\rangle$ with Rabi frequency $\varOmega_2$. We define $\varDelta_1=\omega_1-\omega_{21}$ and $\varDelta_2=\omega_2-\omega_{32}$ as the frequency detunings of the two coupling fields. The decaying rates from excited level $|3\rangle$ to middle level $|2\rangle$ and from middle level $|2\rangle$ to ground level $|1\rangle$ are γ_2 and γ_1, respectively. The decaying rate from level $|3\rangle$ to ground level $|1\rangle$ is assumed to be zero. We concentrate on the fluorescence spectrum of the spontaneous emission from level $|2\rangle$ to level $|1\rangle$.

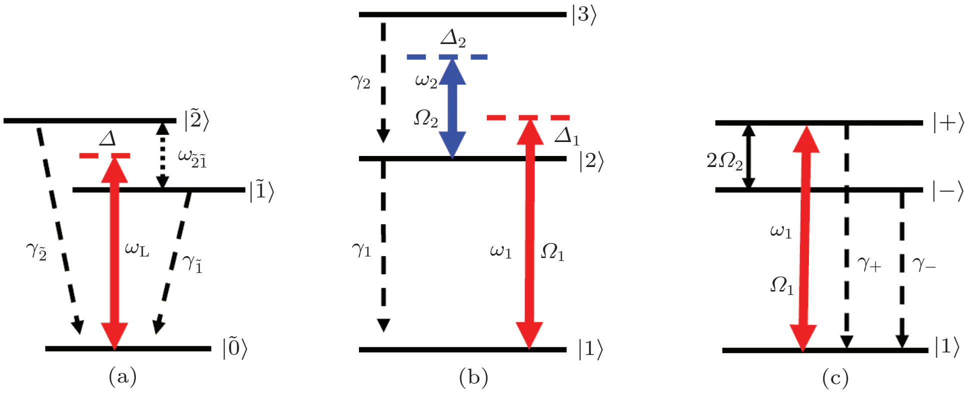

Fig. 1. (colour online) (a) Schematic diagrams of the three-level V system of SGC. (b) Schematic diagram of the three-level ladder system. (c) Dressed state representation of the three-level ladder system.

The equality between Fig. 1(b) and Fig. 1(a) can be seen in the dressed state representation of the field ω_2. With the resonant coupling field of ω_2, level $|2\rangle$ is split into two dressed sublevels, $|+\rangle = (|2\rangle + |3\rangle)/\sqrt{2}$ and $|-\rangle = (|2\rangle - |3\rangle)/\sqrt{2}$ [see Fig. 1(c)]. And the frequencies of level $|+\rangle$ and level $|-\rangle$ relative to level $|1\rangle$ are $\omega_{1\pm} = \omega_1 \pm \varOmega_2$ and the separation between $|+\rangle$ and $|-\rangle$ is $\omega_{+-} = 2\varOmega_2$. There exists quantum interference between two competing decaying channels of the spontaneous emission from the level $|+\rangle$ to level $|1\rangle$ and level $|-\rangle$ to level $|1\rangle$.

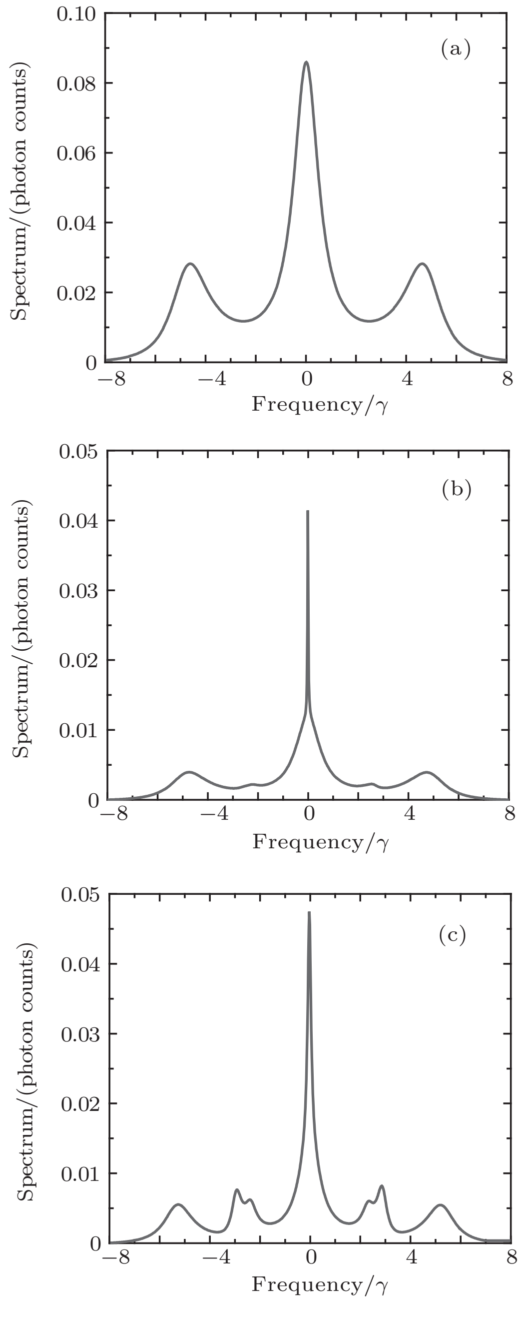

Fig. 2. Calculated fluorescence spectra of the three-level ladder atomic system driven by two laser fields. (a) $\varOmega_2 = 0$; (b) $\varOmega_2 = 0.2\gamma$; (c) $\varOmega_2 = \gamma$. Other parameters are $\varOmega_1 = 2\gamma$, $\varDelta_1 = \varOmega_2$, $\varDelta_2 = 0$, $\gamma_1 = \gamma$, and $\gamma_2 = 0.01\gamma$.

With the method described by Narducci *et al.*[43] and Manka *et al.*,[44] we calculate the spectrum of the spontaneous emission from level $|2\rangle$ to level $|1\rangle$. Figure 2 shows the results of numerical calculations of the fluorescence spectrum. We consider a few special values of $\varOmega_2$. We make $\varDelta_1 = \varOmega_2$, so that the field ω_1 can couple one of the dressed levels to the state $|1\rangle$. The purpose of this consideration is that we can obtain the strongest intensity of the fluorescence. First, when $\varOmega_2 = 0$, the system actually serves as a two-level atom, and a classic Mollow-type, three-peak resonance fluorescence is expected [see Fig. 2(a)]. Then for the case of $\varOmega_2 \neq 0$, the system serves as a three-level ladder atom with two driven fields. The spectrum demonstrates three-peaked and seven-peaked features when the Rabi frequency of ω_2 is $\varOmega_2 = 0.2\gamma$ and $\varOmega_2 = \gamma$, respectively as shown in Figs. 2(b) and 2(c). The most remarkable feature of the fluorescence spectrum is the linewidth narrowing of the center peak, which is similar to those in Figs. 2(b) and 2(d) in Ref. [29]. This narrowing is due to the quantum interference between two competing decaying channels of the spontaneous emission from level $|+\rangle$ to level $|1\rangle$ and level $|-\rangle$ to level $|1\rangle$.

3. Origin of spectral narrowing

To explore the origins of the unusual spectral features, we need to employ the dressed-state representation. First we give the form of Hamiltonian of energy-level scheme in the rotating frame, which is written as

$$\begin{aligned} H =& \varDelta_1 A_{22} + (\varDelta_1 + \varDelta_2) A_{33} \\ &+ (\varOmega_1 A_{12} + \varOmega_2 A_{23} + \mathrm{H.c.}), \end{aligned} \tag{1}$$

where we use units such that $\hbar = 1$; $A_{ij} = |i\rangle\langle j|$ represents a population operator for $i = j$ and a dipole transition operator for $i \neq j$; $\varOmega_1$ and $\varDelta_1$ represent the Rabi frequency and the detuning of ω_1; $\varOmega_2$ and $\varDelta_2$ represent the Rabi frequency and the detuning of ω_2 as we defined above.

The eigenstates of the interaction Hamiltonians is the set of three linear combinations of the energy eigenstates $|i\rangle$ $(i = 1, 2, 3)$, and they are given by the formula

$$|\varPsi_i\rangle = \cos\phi\cos\theta|1\rangle + \sin\phi|2\rangle + \cos\phi\sin\theta|3\rangle, \tag{2}$$

where

$$\tan\phi = \frac{AB}{\sqrt{A^2 + B^2}}, \tag{3a}$$

$$\tan\theta = \frac{A}{B}, \tag{3b}$$

$$A = \frac{\lambda_i}{\Omega_1}, \tag{3c}$$

$$B = \frac{\lambda_i - (\Delta_1 + \Delta_2)}{\Omega_2}, \tag{3d}$$

with λ_i being the corresponding eigenvalue.

The various components in the resonance fluorescence spectrum are associated with different transitions between neighbouring dressed state manifolds. The central peak comes from transitions between the same level of two neighbouring manifolds of the dressed states. The full width at half maximum (FWHM) of the central peak is related to the decay rate of the population $\rho_{\Psi_i\Psi_i}$ $(i = 1, 2, 3)$ in the dressed state $|\Psi_i\rangle$, which is proportional to the squared dipole moment $R_{\Psi_i\Psi_i} = |\langle\Psi_i|P|\Psi_i\rangle|^2$ $(i = 1, 2, 3)$, where $P = \mu_{12}|1\rangle\langle 2|$ is the transition dipole moment operator. In order to explain FWHM of the central peak, we plot the steady-state population of the dressed state $\rho_{\Psi_i\Psi_i}$ as a function of Ω_2 in Fig. 3(a) and the squared dipole moment $R_{\Psi_i\Psi_i}$ as a function of Ω_2 in Fig. 3(b).

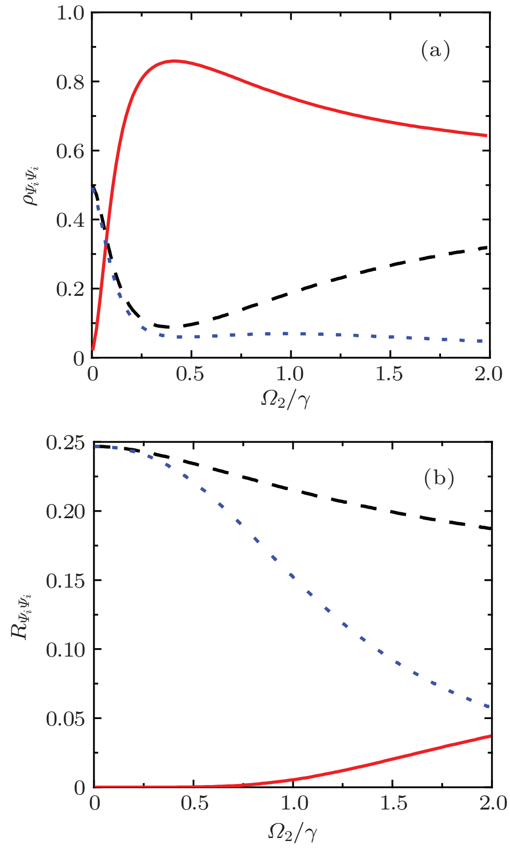

Fig. 3. (colour online) (a) The steady-state population of the dressed states $\rho_{\Psi_i\Psi_i}$ as a function of Ω_2, $\rho_{\Psi_1\Psi_1}$ (black dashed line), $\rho_{\Psi_2\Psi_2}$ (red solid line), $\rho_{\Psi_3\Psi_3}$ (blue dotted line); (b) squared dipole moment $R_{\Psi_i\Psi_i}$ as a function of Ω_2, $R_{\Psi_1\Psi_1}$ (black dashed line), $R_{\Psi_2\Psi_2}$ (red solid line), $R_{\Psi_3\Psi_3}$ (blue dotted line). The parameters are the same as those in Fig. 2.

From Fig. 3(a), we can see that when the value of Ω_2 is small ($\Omega_2 = 0.2$), the steady-state population of the three dressed states are nearly the same. So FWHM of the central peak is related to all the three squared dipole moments of the dressed level. From Fig. 3(b), the values of $R_{\Psi_1\Psi_1}$ and $R_{\Psi_3\Psi_3}$ are much larger than that of $R_{\Psi_2\Psi_2}$. So the decay rates of $\rho_{\Psi_1\Psi_1}$ and $\rho_{\Psi_3\Psi_3}$ are much larger than that of $\rho_{\Psi_2\Psi_2}$. And the small decay rate of $\rho_{\Psi_2\Psi_2}$ gives rise to a pronounced and very sharp feature at line centre. Thus the spectral feature at line centre consists of a sharp peak superimposed on a broad profile, which is the case of Fig. 2(b). And when Ω_2 is increased, the atoms initially in the state $|\Psi_1\rangle$ and $|\Psi_3\rangle$ are transferred to the state $|\Psi_2\rangle$ as shown in Fig. 3(a). As a consequence, the decay rate of $\rho_{\Psi_2\Psi_2}$ dominates the FWHM of the central peak. From Fig. 3(b), though the value of $R_{\Psi_2\Psi_2}$ is increased a little bit, it is still small enough to give rise to the narrowing of the central peak as shown in Fig. 2(c). We conclude that the spectral narrowing of the central peak is due to the slow decay of the dressed-state population $\rho_{\Psi_i\Psi_i}$ and originates from the SGC between two close-lying levels in the dressed-state picture.

4. Experimental setup

We realize a three-level ladder system with the energy scheme shown in Fig. 4(a). The experiments are conducted in the hyperfine levels of ^{85}Rb. Field ω_1 couples the $5S_{1/2}, F = 3 \rightarrow 5P_{3/2}, F = 4$ transition. The Rabi frequency and the detuning of ω_1 are Ω_1 and Δ_1, respectively. Field ω_2 couples the $5P_{3/2}, F = 4 \rightarrow 5D_{5/2}, F = 5$ transition with Rabi frequency Ω_2 and detuning Δ_2. The spontaneous decay rate γ_1 from level $|2\rangle$ to level $|1\rangle$ is about 6 MHz, the spontaneous decay rate γ_2 from level $|3\rangle$ to level $|2\rangle$ is about 0.43 MHz, and owing to the selection rule there is no spontaneous decay from level $|3\rangle$ to level $|1\rangle$.

To minimize the effect of Doppler broadening we carry out the experiment in a rubidium atomic beam. The schematic of the experimental apparatus is shown in Fig. 4(b). The coupling field ω_1 is provided by an extended cavity diode laser which runs at a wavelength of 780 nm. It works at a power of 15 mW and its linewidth is about 1 MHz. The coupling field ω_2 is provided by the coherent-899 Ti: sapphire laser which runs at a wavelength of 776 nm. It works at a power of 0 nW–480 mW with a linewidth of 0.5 MHz. The two laser beams counter-propagate nearly linearly. The laser beams, the atomic beam, and the direction for observing the fluorescence are mutually orthogonal. In the interaction region, the diameter of the atomic beam, laser beam ω_1, and laser beam ω_2 are 1 mm, 2 mm, and 3 mm, respectively. We use two apertures to select a portion of fluorescence that enters the Fabry–Perot (FP) interferometer. The selected portion is emitted in the direction nearly orthogonal to the atomic beam and originates from the interaction area of the lasers and the atomic beam. The two apertures, which are placed 20 cm away from each other, each has a diameter of 2 mm. The FP interferometer, which has a resolution of better than 2 MHz and a free spectral range ($c/4L$) of 680 MHz, gives its best performance with the mode-matching lens, and is driven to scan at a rate of about 100 MHz/min. The light transmitted through the F-P interferometer is detected by a cooled photomulipier, followed by a photon counter.

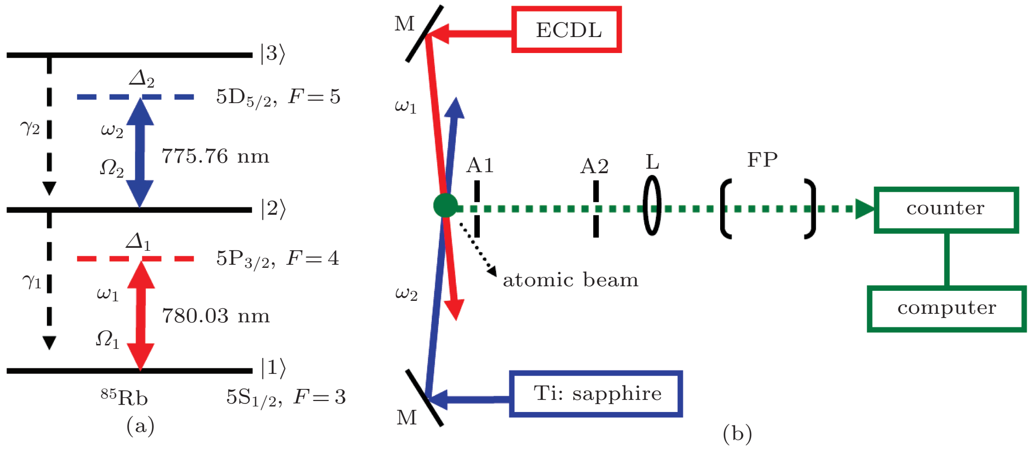

Fig. 4. (colour online) (a) Energy-level scheme for ^{85}Rb. (b) Schematic diagram of the experimental setup, where ECDL stands for external cavity diode laser; Ti:sapphire refers to coherent-899 Ti: sapphire laser; A1 and A2 denote apertures; L represnts lens; FP is the Fabry–Perot interferometer.

5. Results and discussions

We observe the fluorescence spectra of the three-level ladder system and obtain the results shown in Fig. 5. The experimental data are plotted by solid blue lines, while theoretical simulations are presented by dashed red lines. When ω_2 is not applied, a classic Mollow-type resonance fluorescence spectrum is shown up as expected [Fig. 5(a)]. The central peak has a linewidth of 11 MHz. When ω_2 is applied to the three-level system, we observe the fluorescence spectrum under the condition of $\Delta_1 = \Omega_2$ as shown in Fig. 5(b). We can see that, first, in the fluorescence spectrum there appear five peaks with one central peak and two pairs of sidebands. Second, the total intensity of the fluorescence is reduced. Third, the frequency location of the outer sideband is farther away from the central peak. And finally, the most remarkable feature is that the FWHM of the central peak is less than 10 MHz, which is smaller than that in Fig. 5(a). The linewidth narrowing is similar to the results obtained in other researches,[29] where the SGC effect can modify the fluorescence spectrum.

The experimental results are in accordance with what we predicted in the system with SGC. But they are not exactly the same. There are three main effects responsible for these differences. The first one is that the theoretical system with SGC is based on ideal atoms. However in a real atomic system, the residual Doppler broadening undoubtedly broadens the fluorescence spectrum. And second the coherent scattering of the laser field and the angle of the two laser beams in our experiment also modify the spectrum. The last one is the decay rates of $|3\rangle \rightarrow |2\rangle$, which is also responsible for the difference. We perform theoretical simulations including the above effects and the results are presented in dashed red lines in Fig. 5.

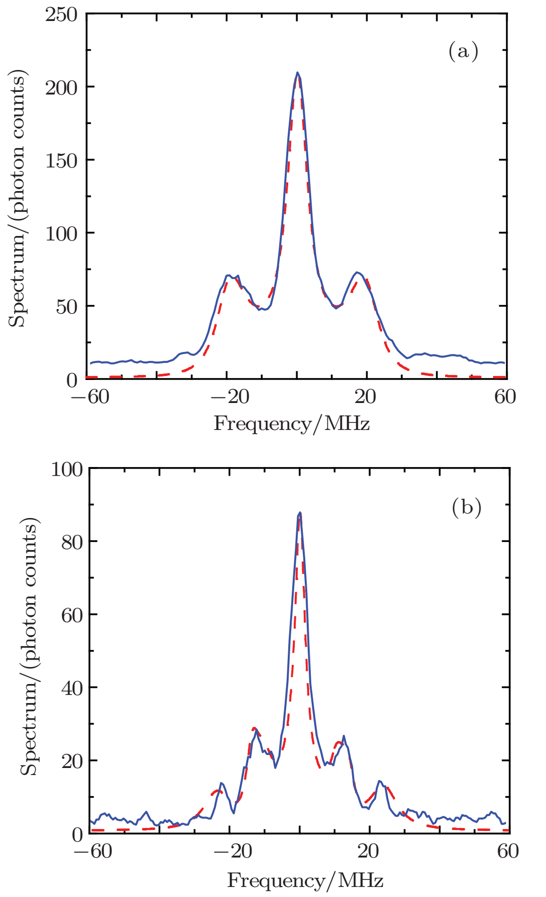

Fig. 5. (colour online) Observed spectra of fluorescence of three-level ladder system with two coupling field. Solid blue curves are for the experimental results, dotted red curves for the theoretical simulation. Panel (a) is for the case of parameters $\Omega_2 = 0$ and $\varDelta_1 = 0$; panel (b) for $\Omega_2 = 6$ MHz and $\varDelta_1 \approx 6$ MHz. Other parameters are $\Omega_1 = 12$ MHz, $\varDelta_2 \approx 0$, $\gamma_1 = 6$ MHz, and $\gamma_2 = 0.5$ MHz.

6. Conclusion

We investigated the fluorescence spectrum of a three-level ladder system and showed the equality between this system and a three-level V-type system with parallel dipole moments. We showed that the system can exhibit the features previously predicted for a system with SGC: in particular ultranarrow lines in the resonance fluorescence spectrum. We explained the spectral features in terms of the dressed-atom model of the system. We proposed a practical scheme which permits the observation of the interesting features predicted for a three-level system showing SGC, but without any requirement for two close-lying levels and parallel dipole moments. We observed the linewidth narrowing of the central peak in a rubidium atomic beam when both fields are added. The experimental measurements accord quantitatively with theoretical calculations and demonstrate the existence of SGC.

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