Observation of doubly dressed states in cold atoms

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We report an experimental observation of doubly dressed states in ⁸⁷Rb atoms cooled and confined in a magneto-optical trap. The doubly dressed states are produced by a strong-coupling laser and a moderate pump laser in a three-level atomic configuration. The absorption spectrum of a weak probe laser reveals a three-peaked spectral profile that can be interpreted by a dressed-state picture and agrees well with calculations based on the density-matrix equations.

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The interaction of atoms with continuous-wave, intense laser fields is of fundamental interest in laser spectroscopy and quantum optics. Theoretical and experimental studies of the spectral and dynamic features of the atoms under a variety of strong field conditions have been explored [1,2]. A double-peak absorption spectrum (Autler-Townes doublet) was observed when a weak probe field couples a strongly driven two-level system to a third atomic level [3]. A triplepeak fluorescence spectrum of the driven two-level system was first predicted [4] and then observed [5,6]. Such studies, along with the observation of the absorption and dispersion spectrum of a weak probe field in the driven two-level system [7], provide fundamental insight into radiation-matter interactions. Recent studies show that multiphoton and interference effects play important roles in the atomic response to bichromatic and/or polychromatic driving fields [8,9], and complicated spectral features are usually observed in such driven atomic systems [10,11]. Physical pictures based on the dressed states are derived to interprete the two-level atom radiation interactions and can be extended to understand interactions of multilevel atoms and multiple coherent fields [12].

Here we report an experimental observation of doubly dressed states in a three-level atomic system. The resonance fluorescence of a strongly coupled three-level atomic system has been studied before [13]. In particular, in a three-level atom containing one strong and one weak transition, quantum fluctuations can be stabilized such that the spontaneous decay from the strong transition shows a subnatural linewidth [14,15]. Our study explores a three-level system in which one transition is driven by a strong-coupling field while the other transition with a comparable spontaneous decay rate is driven by a moderate pump field. The threelevel coupled system is probed by a weak laser field connecting the system to a fourth level and the absorption spectrum of the probe is recorded. We observe a triple-peak absorption spectrum that can be interpreted by a doubly dressed-state picture. In contrast to destructive interference in a three-level system demonstrating electromagnetically induced transparency [EIT] [16], we show that constructive interference occurs between the excitation paths of the two closely spaced, doubly dressed states. Our experiment was carried out in ⁸⁷Rb atoms cooled and confined in a magneto-optical trap (MOT) and the experimental measurements agree quantitatively with theoretical calculations based on a four-level atomic system described below.

Consider a four-level atomic system coupled by three laser fields depicted in Fig. 1(a) in which $|1\rangle$ and $|2\rangle$ are two ground (hyperfine) states, and $|3\rangle$ and $|4\rangle$ are two excited states. A strong coupling laser with Rabi frequency 2Ω connects states $|2\rangle$ and $|3\rangle$ while a moderate pump laser with Rabi frequency $2\Omega'$ couples states $|2\rangle$ and $|4\rangle$. A weak probe laser with Rabi frequency 2g couples states $|1\rangle$ and $|3\rangle$. We define $\Delta_c = \omega_c - \omega_{32}$, $\Delta' = \omega_a - \omega_{42}$, and $\Delta = \omega_p - \omega_{31}$ as the frequency detuning for the coupling, the pump, and the probe field, respectively. Also Γ_3 (Γ_4) is the spontaneous decay rate of the excited states $|3\rangle$ ($|4\rangle$). For a dressed-state analysis (Ω , and $\Omega' > \Gamma_3$, Γ_4 , and g), consider states $|2\rangle$, $|3\rangle$, and $|4\rangle$ plus the coupling and pump fields as a coupled atomfield system that is probed by a weak probe field ω_p [12,13]. The three semiclassical dressed states from the three-level atom coupled by the two laser fields can be written as

$$|1,+\rangle = a_+|2\rangle + b_+|3\rangle + c_+|4\rangle, \qquad (1a)$$

$$|2,0\rangle = a_0|2\rangle + b_0|3\rangle + c_0|4\rangle, \tag{1b}$$

$$|3,-\rangle = a_{-}|2\rangle + b_{-}|3\rangle + c_{-}|4\rangle.$$
 (1c)

Here a_i , b_i , and c_i (i = +, 0, or -) are constants. For arbitrary detunings of the coupling and the pump fields, it is



FIG. 1. (a) Four-level atomic system and laser coupling scheme. (b) Intermediate picture of primary dressed states created by the coupling laser. (c) Dressed-state picture showing the primary dressed state $|-\rangle$ and the secondary dressed states (doubly dressed states) $|+, +\rangle$ and $|+, -\rangle$, which leads to a three-peaked probe absorption spectrum.

tedious to write explicitly the analytical results for the three dressed states and the corresponding energies. However, it is straightforward to derive these results when the coupling field and the pump field are both exactly on resonance, i.e., $\Delta_c = \Delta' = 0$, under which the three dressed states are given by

$$|1,+\rangle = \frac{1}{\sqrt{2}} \left(|2\rangle + \frac{\Omega}{\sqrt{\Omega^2 + \Omega'^2}} |3\rangle + \frac{\Omega'}{\sqrt{\Omega^2 + \Omega'^2}} |4\rangle \right),$$
(2a)

$$|1,0\rangle = \frac{\Omega}{\sqrt{\Omega^2 + \Omega'^2}} |4\rangle - \frac{\Omega'}{\sqrt{\Omega^2 + \Omega'^2}} |3\rangle, \qquad (2b)$$

$$|1,-\rangle = \frac{1}{\sqrt{2}} \left(|2\rangle - \frac{\Omega}{\sqrt{\Omega^2 + \Omega'^2}} |3\rangle - \frac{\Omega'}{\sqrt{\Omega^2 + \Omega'^2}} |4\rangle \right).$$
(2c)

The corresponding eigenenergies are $E_+ = \sqrt{\Omega^2 + \Omega'^2}$, $E_0 = 0$, and $E_- = -\sqrt{\Omega^2 + \Omega'^2}$. Therefore, the absorption spectrum of the probe field contains three spectral peaks corresponding to the dressed-state transitions $|1\rangle| \rightarrow |1+\rangle$, $|1\rangle \rightarrow |1,0\rangle$, and $|1\rangle \rightarrow |1,-\rangle$, with the peak positions given by $\Delta = E_+$, E_0 , and E_- , respectively. The square of the dipole moment for the probe absorption is identical for the two transitions $|1\rangle \rightarrow |1,+\rangle$, $|1\rangle \rightarrow |1,-\rangle$, which is given by $|\langle 1|d|1,+\rangle|^2 = d_{13}^2\Omega^2/2(\Omega^2 + \Omega'^2)$ while for the transition $|1\rangle \rightarrow |1,0\rangle$, it is given by $|\langle 1|d|1,+\rangle|^2 = d_{13}^2\Omega'^2/(\Omega^2 + \Omega'^2)$. Here $d_{13} = \langle 1|d|3\rangle$ is the dipole moment between the states $|1\rangle$ and $|3\rangle$. Since the absorption is proportional to the square of the transition dipole moment, the ratio of the amplitudes between the probe transitions $|1\rangle \rightarrow |1,+\rangle$

When $\Omega \ge \Omega'$, one may consider the coupled three-level system by two steps. First, the dominant interaction of the coupling field and the atom with the states $|2\rangle$ and $|3\rangle$ creates primary dressed states $|+\rangle$ and $|-\rangle$ [Fig. 1(b)] given by [12]

$$|+\rangle = \sin \theta |3\rangle + \cos \theta |2\rangle, \tag{3a}$$

$$|-\rangle = \cos \theta |3\rangle - \sin \theta |2\rangle.$$
 (3b)

Here $\tan 2\theta = -2\Omega/\Delta_C$ ($0 \le \theta \le \pi/2$). The two dressed states are separated in energy by 2Ω . Next, consider secondary dressed states created from the primary dressed states $|+\rangle$ and $|-\rangle$ by the pump field with Rabi frequency $2\Omega'$. When the pump field is tuned close to one of the primary dressed states $|+\rangle$ (or $|-\rangle$), approximately the pump field only couples the dressed state $|+\rangle$ (or $|-\rangle$) to the state $|4\rangle$ and leaves the other dressed state $|-\rangle$ (or $|+\rangle$) unperturbed. In other words, the pump field can selectively couple one of the primary dressed states and creates the doubly or secondary dressed states. For example, if the pump laser only couples the dressed state $|+\rangle$, the resulting doubly dressed state $|+,+\rangle$ and $|+,-\rangle$ are given by

$$+, -\rangle = \cos \theta' |4\rangle - \sin \theta' |+\rangle.$$
 (4b)

Here $\tan 2\theta' = -2\Omega'/(\Delta' + \Omega)$ $(0 \le \theta' \le \pi/2)$. In particular, with a resonant coupling laser $(\Delta_c = 0)$ and a pump laser with a detuning $\Delta' = -\Omega$, the doubly dressed states acquires the simplest form that is given by

$$|+,+\rangle = \frac{1}{\sqrt{2}}|4\rangle + \frac{1}{2}|2\rangle + \frac{1}{2}|3\rangle,$$
 (5a)

$$|+,-\rangle = \frac{1}{\sqrt{2}}|4\rangle - \frac{1}{2}|2\rangle - \frac{1}{2}|3\rangle.$$
 (5b)

Together with the unperturbed primary dressed state $|-\rangle$ $=(1/\sqrt{2})(|3\rangle - |2\rangle)$, the semiclassical coupled atom-field system consists of three discrete states. In order to validate the above simple analysis, we solve the Hamiltonian of the coupled atomic states $|2\rangle$, $|3\rangle$, and $|4\rangle$ under the conditions $\Omega \gg \Omega'$, $\Delta_c = 0$, and $\Delta' = -\Omega$. The eigenvalues of the three dressed states are given to the first order in Ω' as $E_{\pm} = \Omega$ $\pm \Omega'/\sqrt{2}$, and $E_0 = -\Omega$. The corresponding dressed states are $|+,+\rangle$, $|+,-\rangle$, and $|-\rangle$, which agree with the simple analysis. The absorption spectrum of the probe laser will have three spectral peaks corresponding to the probe transition from the state $|1\rangle$ to the doubly dressed states $|+,+\rangle$, $|+,-\rangle$ and the singly dressed state $|-\rangle$. Similarly, the square of the dipole moment for the probe absorption can be derived: for the transition $|1\rangle \rightarrow |+,+\rangle$ ($|+,-\rangle$), it is given by $|\langle 1|d|+,+\rangle|^2 = d_{13}^2/4$; while for the transition $|1\rangle \rightarrow |-\rangle$, it is given by $|\langle 1|d|+,+\rangle|^2 = d_{13}^2/2$. Therefore, the absorption amplitude for the two doubly dressed states will be half of the amplitude for the singly dressed state $|-\rangle$.

Coherent coupling of the atomic states creates dressed atomic states and leads to interference for the transition between the dressed states and other noncoupled states. This leads to interesting phenomenon such as electromagnetically induced transparency or coherent population trapping [16]. By coupling a single primary dressed state with a separate atomic state, the doubly dressed states are created. Interference will occur for the transition from the doubly dressed states to a noncoupled atomic state such as the one connected by the weak probe laser. For the four-level system discussed here, coherent manipulation of the three atomic states by two laser fields leads to constructive interference experienced by the probe laser at the center of the two doubly dressed states (the probe detuning $\Delta = \omega_p - \omega_{31} = \Omega$). From the dressedstate picture, the probe absorption at $\Delta = \Omega$ (from the state $|1\rangle$ to the center of the two doubly dressed states) is proportional to

$$P \propto \left| \frac{\langle 1 | \vec{d} \cdot \vec{E}_p | +, + \rangle}{-\Omega'} + \frac{\langle 1 | \vec{d} \cdot \vec{E}_p | +, - \rangle}{\Omega'} \right|^2 = \frac{g^2}{\Omega'^2}.$$
(6)

That is, the two excitation paths, $|1\rangle \rightarrow |+,+\rangle$ and $|1\rangle \rightarrow |+,-\rangle$, interfere constructively, leading to enhanced absorption. This is in contrast to a four-level system analyzed by Lukin *et al.* [17] in which the doubly dressed system exhibits sharp dark resonance due to destructive interference

$$|+,+\rangle = \sin \theta' |4\rangle + \cos \theta' |+\rangle,$$
 (4a)



FIG. 2. (a) Energy levels of ⁸⁷Rb atoms and laser coupling scheme used in the experiment. (b) Energy diagram showing detailed coupling among the magnetic sublevels and the three laser fields with appropriate polarizations.

between the secondary dressed states. The difference between the present four-level system and that of Ref. [17] is that here the state $|4\rangle$ is an excited state while it is metastable in Ref. [17]. Therefore, decoherence of the Raman coherence ρ_{14} occurs in the present four-level system but not in the four-level system of Ref. [17]. In the dressed-state picture, this transfers into constructive interference or destructive interference for the two systems, respectively.

The level structure and laser coupling scheme for ⁸⁷Rb atoms used in our experiment are depicted in Fig. 2. A coupling laser drives the $D_1 F=2 \rightarrow F'=1$ transition at 795 nm and creates dressed atomic states $|+\rangle$ and $|-\rangle$. A pump laser drives the $D_2 F=2 \rightarrow F'=3$ transition at 780 nm and generates the secondary dressed states $|+,+\rangle$ and $|+,-\rangle$. A weak probe laser couples the $D_1 F=1 \rightarrow F'=1$ transition. The coupling laser and the probe laser are linearly polarized perpendicular to each other while the pump laser is circularly polarized. Figure 2(b) shows the detailed coupling diagram among the magnetic sublevels. The induced transitions among the magnetic sublevels by the three lasers can be grouped together according to selection rules and form a manifold of four-level systems. To a good approximation,



FIG. 3. Schematic diagram of the experimental setup. The dark spot diameter in the repump beam profile is ~4 mm. At t_1 , the resonant coupling laser is turned on by an AOM, and the frequency detuning of the cooling and trapping laser (now acts as the pump laser) is suddenly shifted to $\Delta' \approx \Omega$ (or $-\Omega$). At t_2 ($t_2-t_1 = 0.15$ ms), the probe laser frequency is scanned across the D_1 $F = 1 \rightarrow F' = 1$ transition (the scan time $t_3-t_2=0.2$ ms). At t_4 the coupling laser is turned off by the AOM, and the frequency detuning of the cooling and trapping laser is shifted back to $\Delta' \approx -3\Gamma_4$ below the $D_2 F = 2 \rightarrow F' = 3$ transition. The experiment is running at a repetition rate of 10 Hz. For each period, the probe absorption measurement runs for $t_4-t_1=0.4$ ms, and the cooling and trapping lasts for $t_5-t_4=99.6$ ms.

the coupled Rb system can be viewed as equivalent to the generic four-level system depicted in Fig. 1. The validity of such a simplification has been supported by several previous studies [18,19]. The spontaneous decay rates of the states $|3\rangle$ and $|4\rangle$ are Γ_3 ($2\pi \times 5.3$ MHz) and Γ_4 ($2\pi \times 5.9$ MHz), respectively.

Our experiment was carried out in a vapor cell MOT produced in the center of a 10-port, 4,5-diam, stainless-steel vacuum chamber pumped down to a pressure $\sim 10^{-9}$ Torr. The MOT was produced with the standard six-beam configuration [20]. The rubidium vapor pressure of $\sim 10^{-8}$ Torr was maintained with three rubidium getters connected in series and placed close to the center of the vacuum chamber. The experimental scheme is depicted in Fig. 3 (the trap laser beams in the z direction are not drawn). An extended-cavity diode laser with output power of ~ 40 mW was used as the cooling and trapping laser that supplied six σ^+ and σ^- polarized beams in three perpendicular spatial directions and its frequency was locked to $\sim 3\Gamma_4$ below the D_2 $F=2\rightarrow F'$ = 3 transition. Another extended cavity diode laser with output power of ~ 10 mW was used as the repump laser and its frequency was locked to the $D_2 F = 1 \rightarrow F' = 2$ transition. A dark spot of \sim 4 mm in diameter was made inside the repump beam profile. In this dark MOT configuration [21], more atoms can be accumulated in the F=1 hyperfine states and



FIG. 4. (a) Measured probe absorption versus the probe frequency detuning Δ when the coupling laser and the pump laser are both on resonance, $\Delta_c \approx 0$ and $\Delta' \approx 0$. (b) and (c) Measured probe absorption versus Δ with the off-resonant coupling and pump lasers. The spectra are taken with $\Delta' \approx 8$ MHz and $\Delta_c \approx -21$ MHz (b), -32 MHz (c), and -42 MHz (d). Other parameters are $2\Omega = 34$ MHz, $2\Omega' = 10$ MHz, and 2g = 1.5 MHz.

more importantly, the repump laser will not interfere with the experimental measurement. The diameter of the trapping laser beams and the repumping laser beam was ~ 1 cm. The weak probe laser is provided by a third extended-cavity diode laser. It has a beam diameter ~ 0.8 mm and power attenuated to $\sim 1 \ \mu$ W. A Ti: sapphire laser (Coherent 899-21) with a beam diameter ~ 3 mm is used as the coupling laser. The probe laser and the coupling laser are overlapped with the trapped Rb cloud and the probe-laser absorption spectrum is recorded by a biased photodiode as shown in Fig. 3. The linewidth of the extended-cavity diode lasers is less than 1 MHz and the linewidth of the Ti:sapphire laser is ~ 3 MHz. The trapped ⁸⁷Rb atom cloud is ~ 1.5 mm in diameter and



FIG. 5. (a) Measured probe absorption versus the probe frequency detuning Δ with the coupling laser on resonance, $\Delta_c \approx 0$, and the pump laser far detuned ($\Delta' \approx 200$ MHz). (b) Measured probe absorption versus Δ with $\Delta_c \approx 0$ and the $\Delta' \approx -\Omega$. (c) Measured probe absorption versus Δ with $\Delta_c \approx 0$ and $\Delta' \approx \Omega$. The three-peaked spectra represent the probe transition to the three dressed states [see Fig. 1(c)]. Solid lines are experimental data and dashed lines are theoretical calculations.

contains $\sim 4 \times 10^7$ atoms (estimated from the measured single-pass absorption of $\sim 20\%$ with the weak probe laser tuned to the $D_1 F = 1 \rightarrow F' = 2$ transition). During the experiment, the trap laser beams, the dark repump laser beam, and the probe laser beam are always on, while the coupling laser beam is extracted from the first-order diffracted beam of the Ti:sapphire laser by a 110 MHz acousto-optic modulator (AOM) that is turned on and off by a pulse generator. The

diffraction efficiency of the AOM is \sim 40%. The maximum power of the Ti:sapphire laser before the AOM is $\sim 200 \text{ mW}$. The experiment is run in a sequential mode with a repetition rate of 10 Hz and the time sequence is shown in Fig. 3. For each period of $t_5 - t_1 = 100 \text{ ms}$, the time for the probe absorption measurement lasts $\sim t_4 - t_1 = 0.4 \text{ ms}$ while the rest of the time, $t_5 - t_4 = 99.6 \text{ ms}$, is used for the cooling and trapping of the Rb atoms. At t_1 (see Fig. 3), the resonant coupling laser is turned on by the AOM in 150 ns, and the cooling and trapping laser frequency is suddenly shifted to the desired value Δ' , which now acts as the pump laser for the secondary dressed-state generation. For $t_2 - t_1 = 0.1$ ms, the Rb atoms are optically pumped to the ground F=1 hyperfine state. At t_2 , the weak probe laser is scanned across the $D_1 F = 1 \rightarrow F' = 1$ transition in a time of $t_3 - t_2 \sim 0.2$ ms and the probe absorption was recorded by a digital oscilloscope. At $t_4 = t_1 + 0.4$ ms, the coupling laser is turned off by the AOM, and the cooling and trapping laser frequency is shifted back to $\sim 3\Gamma_4$ below the resonance to reestablish the MOT. Since the probe laser is very weak and frequency detuned far from the resonance in the cooling and trapping period, it does not disturb the MOT. Because the coupling laser pulse and the frequency scan time are much greater than the relaxation time of the Rb excited states, the experimental measurements are essentially carried out in the steady-state regime.

Figure 4 plots measured absorption spectra of the weak probe laser versus the probe frequency detuning Δ . The experimental data are plotted in solid lines. From the measured spectra, we derive the Rabi frequencies of the coupling laser and the pump laser, which are $2\Omega = 34$ (±2) MHz and $2\Omega' = 10 \ (\pm 2)$ MHz, respectively. We then used these parameters and the estimated Rabi frequency of the weak probe laser $(2g \approx 1.5 \text{ MHz})$ in the numerical calculations for the four-level system. The calculated results are plotted as the dashed lines and agree well with the experimental measurements. Figure 4(a) shows the probe spectrum when both the coupling laser and the pump laser are on resonance (Δc $=\Delta'=0$). We observe a three-peaked, symmetrical line profile that corresponds to the transition from the state $|1\rangle$ to the three dressed states given in Eq. (2) with equal energy splittings. The measured amplitude ratio between the two sidebands and the central peak is approximately equal to $\Omega^2/2\Omega'^2$ derived from the dressed-state analysis. The central peak at $\Delta = 0$ represents the third-order nonlinear absorption, which is enhanced by constructive quantum interference and is most pronounced at small Ω values [22,23]. Figures 4(b) and 4(c) show the probe spectra observed with off-resonant coupling and pump lasers ($\Delta c \neq 0$ and $\Delta' \neq 0$). The observed spectra show three peaks with varying amplitudes and unequal peak separations, which agree well with the theoretical calculations. As expected, when the coupling laser and the pump laser are frequency detuned from the resonance ($\Delta c \neq 0$ or $\Delta' \neq 0$), the three dressed states have different energy splittings and the mixing coefficients of the wave functions in general are all different for the three dressed states. This leads to an asymmetrical spectral profile with three spectral peaks of unequal amplitudes and unequal energy separations.

In Fig. 5 we plot measured absorption spectra of the weak probe laser versus the probe frequency detuning Δ , which corresponds to the special case discussed by Eq. (5). During the experiment, the coupling laser was locked on the atomic resonance ($\Delta c = 0$). Figure 5(a) shows the probe absorption obtained when the pump laser frequency was far detuned $(\Delta' \sim 200 \text{ MHz}$ to the blue side), which represents the EIT spectrum in a Λ -type system [16]. The two peaks in the spectrum represent the transitions from the ground state $|1\rangle$ to the two singly dressed states $|+\rangle$ and $|-\rangle$ [Eq. (3) with $\Delta c = 0$]. Figure 5(b) shows the probe spectrum with the pump detuning $\Delta' \approx -\Omega$ while Fig. 5(c) shows the probe spectrum with the pump detuning $\Delta' \approx \Omega$. The two spectra demonstrate the three-peaked spectral profile that corresponds to a primary dressed state unperturbed by the pump field and the two secondary dressed states generated by the pump field from one of the singly dressed state. The measurements agree with the dressed-state analysis. We also measured the probe absorption spectrum for other pump laser detunings (varied from $\Delta' = -100 \text{ MHz}$ to +100 MHz) and observed asymmetrical, three-peaked spectral profiles with varying amplitudes. As the pump laser is detuned away from the primary dressed states, the probe spectral profile evolves into the two-peaked EIT spectrum plotted in Fig. 5(a).

In conclusion, we have observed the doubly dressed states created by two laser fields in ⁸⁷Rb atoms cooled and confined in a MOT. The three-peaked spectrum can be interpreted by the dressed-state picture and agrees well with theoretical calculations-based on a four-level coupled system. In contrast to the destructive interference in the Λ -type EIT system, the added pump field creates constructive interference between the excitation paths to the doubly dressed states. Our experiment demonstrates the versatility of coherent manipulation of multiple atomic states with multiple laser fields in cold atoms.

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