Fluorescence properties in a single driven four-level atomic system*

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The spontaneous emission decay dynamics of a tripod configuration four-level atom driven by a single laser field is studied. Under different initial conditions, we discuss the effects of quantum interference and detuning of external driving field on atomic spontaneous emission properties. For the larger detuning, the interesting phenomena of the spectral line narrowing are found which stem from the contribution of external driving field.

Keywords: spontaneous emission spectrum, four-level atom, driving field
PACC: 3280, 4250, 3250F

1. Introduction

The control of atomic spontaneous emission is an active research topic in quantum optics because of its potential application in high precision spectroscopy,[1−3] quantum information[4−6] etc. One way to control the atomic spontaneous emission is to place an atom in different circumstances, such as in free space, in photonic crystals[7,8] and in optical cavity,[9,10] which have different densities of electromagnetic modes interacting with atoms. Another way to control atomic spontaneous emission is to drive atoms by external driving fields. Narducci *et al.*[11] and Manka *et al.*[12] investigated the steady-state spontaneous emission spectrum of a three-level atom driven by two coherent fields and the absorption spectrum of a weak tunable probe. In this work was made the first suggestion of control of atomic spontaneous emission by external fields that led to line narrowing.

In recent years, substantial attention has been paid to the study of the control of atomic spontaneous emission in multi-level atomic systems by using external driving fields, which lead to many novel and unexpected effects.[13−17] Zhu and Scully[13] and Lee *et al.*[14] have investigated the steady state emission behaviour in a driven four-level atom, in which quantum interference effects such as spectral-line elimination and quenching of spontaneous emission were observed. Paspalakis and Knight[18] proposed a phase control scheme in the atom driven by two lasers with the same frequencies, where the relative phase of two lasers was used to obtain partial cancellation, extreme line-width narrowing and total cancellation in the spontaneous emission spectrum. Moreover, the influence of phase and amplitude of the driving field on spontaneous emission spectrum in a four-level atomic system driven by three fields have been discussed.[19] Recently, Wu *et al.*[20] and Li *et al.*[21] studied the spontaneous emission properties of a double-coherent field driven four-level atom in tripod configuration and showed a few interesting phenomena such as fluorescence quenching, spectral-line narrowing, spectral line enhancement and spectral line elimination due to quantum interference. In the present paper, we present another scheme for the four-level atom in tripod configuration driven by one laser field, and discuss the influence of quantum interference and detuning of external driving field on the spectrum line of each transition. By changing the initial condition of system and the detuning of external driving field we can control the atomic spontaneous emission.

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2. Theoretical model and equations

We consider a four-level atom that consists of one upper level $|2\rangle$, and three lower levels $|0\rangle$, $|1\rangle$ and $|3\rangle$ as shown in Fig. 1, where we set the level $|2\rangle$ to be the upper state and the $|3\rangle$ the lowest ground state. The ground state $|3\rangle$ is coupled by a laser field with frequency ν to the excited state $|2\rangle$.

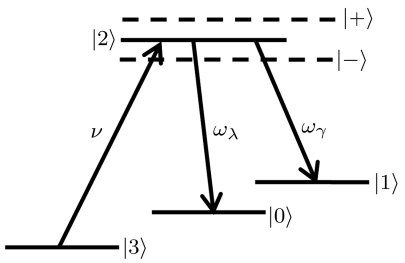


Fig. 1. Level scheme of the model atom, where the dash lines represent the dressed levels.

The interaction picture Hamiltonian in the dipole and rotating-wave approximation is given by ($\hbar = 1$)

$$\begin{aligned} & H \\ = {} & \Omega \left|3\right\rangle \left\langle 2\right| \mathrm{e}^{\mathrm{i}\Delta t} + \sum_{\lambda} g_{\lambda} \mathrm{e}^{-\mathrm{i}(\omega_{\lambda}-\omega_{20})t} \left|2\right\rangle \left\langle 0\right| a_{\lambda} \\ & + \sum_{\gamma} g_{\gamma} \mathrm{e}^{-\mathrm{i}(\omega_{\gamma}-\omega_{21})t} \left|2\right\rangle \left\langle 1\right| a_{\gamma} + \Omega^{*} \left|2\right\rangle \left\langle 3\right| \mathrm{e}^{-\mathrm{i}\Delta t} \\ & + \sum_{\lambda} g_{\lambda}^{*} \mathrm{e}^{\mathrm{i}(\omega_{\lambda}-\omega_{20})t} \left|0\right\rangle \left\langle 2\right| a_{\lambda}^{+} \\ & + \sum_{\gamma} g_{\gamma}^{*} \mathrm{e}^{\mathrm{i}(\omega_{\gamma}-\omega_{21})t} \left|1\right\rangle \left\langle 2\right| a_{\gamma}^{+}, \end{aligned} \tag{1}$$

where Ω is the Rabi frequency of the driving field. The transition frequencies from level $|2\rangle$ to levels $|0\rangle$, $|1\rangle$, and $|3\rangle$ are denoted by ω_{20}, ω_{21} and ω_{23}, respectively; the $\Delta = \nu - \omega_{23}$ is the detuning of external driving field from resonance of the $|2\rangle \rightarrow |3\rangle$; the $g_{\lambda}(g_{\gamma})$ is the coupling constant between the $\lambda(\gamma)$-th mode and the atomic dipoles between levels $|0\rangle$, $|1\rangle$ and $|2\rangle$. We assume the coupling constants to be real for simplicity. $a_{\lambda}(a_{\gamma})$ and $a_{\lambda}^{+}(a_{\gamma}^{+})$ are the annihilation and the creation operators for the $\lambda(\gamma)$-th reservoir modes with frequency $\omega_{\lambda}(\omega_{\gamma})$. At any time t, the atom-field state vector can be written as

$$\begin{aligned} \left|\Psi(t)\right\rangle = {} & b_{3}(t) \left|3,0\right\rangle + b_{2}(t) \left|2,0\right\rangle + \sum_{\lambda} b_{\lambda}(t) \left|0,1_{\lambda}\right\rangle \\ & + \sum_{\gamma} b_{\gamma}(t) \left|1,1_{\gamma}\right\rangle, \end{aligned} \tag{2}$$

where $|0\rangle$ denotes the vacuum state of the radiation field, and $|1_{\lambda}\rangle$ ($|1_{\gamma}\rangle$) means that there is one photon in the $\lambda(\gamma)$-th reservoir mode. Substituting the Hamiltonian and wavefunction of system into the time-dependent Schrodinger equation $\mathrm{i}\partial\Psi/\partial t = H\Psi$, the equations of motion for the probability amplitudes are given by

$$\mathrm{i}\dot{b}_{3}(t) = \Omega b_{2}(t) \mathrm{e}^{\mathrm{i}\Delta t}, \tag{3}$$

$$\begin{aligned} \mathrm{i}\dot{b}_{2}(t) = {} & \Omega^{*} b_{3}(t) \mathrm{e}^{-\mathrm{i}\Delta t} + \sum_{\lambda} g_{\lambda} b_{\lambda}(t) \mathrm{e}^{-\mathrm{i}(\omega_{\lambda}-\omega_{20})t} \\ & + \sum_{\gamma} g_{\gamma} b_{\gamma}(t) \mathrm{e}^{-\mathrm{i}(\omega_{\gamma}-\omega_{21})t}, \end{aligned} \tag{4}$$

$$\mathrm{i}\dot{b}_{\lambda}(t) = g_{\lambda}^{*} b_{2}(t) \mathrm{e}^{\mathrm{i}(\omega_{\lambda}-\omega_{20})t}, \tag{5}$$

$$\mathrm{i}\dot{b}_{\gamma}(t) = g_{\gamma}^{*} b_{2}(t) \mathrm{e}^{\mathrm{i}(\omega_{\gamma}-\omega_{21})t}. \tag{6}$$

We proceed by performing a formal time integration of Eqs. (5) and (6), and substitute the result into Eq. (4) to obtain the integrodifferential equation

$$\begin{aligned} & \mathrm{i}\dot{b}_{2}(t) \\ = {} & \Omega^{*} b_{3}(t) \mathrm{e}^{-\mathrm{i}\Delta t} \\ & - \mathrm{i} \int_{0}^{t} \mathrm{d}t' b_{2}(t') \sum_{\lambda} g_{\lambda}^{2} \mathrm{e}^{-\mathrm{i}(\omega_{\lambda}-\omega_{20})(t-t')} \\ & - \mathrm{i} \int_{0}^{t} \mathrm{d}t' b_{2}(t') \sum_{\gamma} g_{\gamma}^{2} \mathrm{e}^{-\mathrm{i}(\omega_{\gamma}-\omega_{21})(t-t')}, \end{aligned} \tag{7}$$

where we have used $b_{\lambda}(0) = 0$ and $b_{\gamma}(0) = 0$. Because two transitions were coupled to free space, we can use Weisskopf–Wigner approximation $\sum_{\lambda(\gamma)} g_{\lambda(\gamma)}^{2} \mathrm{e}^{-\mathrm{i}(\omega_{\lambda(\gamma)}-\omega_{20(21)})(t-t')} = \Gamma_{1(2)}\delta(t-t')/2$, where $\Gamma_{1(2)} = 2\pi \left|g_{\lambda(\gamma)}\right|^{2} D(\omega_{\lambda(\gamma)})$ is the spontaneous-decay rate from the upper level to the two lower levels, and $D(\omega_{\lambda(\gamma)})$ is the mode density at frequency $\omega_{\lambda(\gamma)}$ in the free space. Thus, Eq. (7) can be written as

$$\mathrm{i}\dot{b}_{2}(t) = \Omega^{*} b_{3}(t) \mathrm{e}^{-\mathrm{i}\Delta t} - \mathrm{i}\frac{\Gamma_{1}+\Gamma_{2}}{2} b_{2}(t). \tag{8}$$

To eliminate the exponential terms of Eqs. (3)–(6), we perform the following change of variable $b_{3}(t) = a_{3}(t) \mathrm{e}^{\mathrm{i}\Delta t}$, $b_{2}(t) = a_{2}(t)$, $b_{\lambda}(t) = a_{\lambda}(t) \mathrm{e}^{\mathrm{i}(\omega_{\lambda}-\omega_{20})t}$ and $b_{\gamma}(t) = a_{\gamma}(t) \mathrm{e}^{\mathrm{i}(\omega_{\gamma}-\omega_{21})t}$, and then we have

$$\mathrm{i}\dot{a}_{3}(t) = \Delta a_{3}(t) + \Omega a_{2}(t), \tag{9}$$

$$\mathrm{i}\dot{a}_{2}(t) = \Omega^{*} a_{3}(t) - \mathrm{i}\left(\frac{\Gamma_{1}}{2} + \frac{\Gamma_{2}}{2}\right) a_{2}(t), \tag{10}$$

$$\mathrm{i}\dot{a}_{\lambda}(t) = (\omega_{\lambda} - \omega_{20}) a_{\lambda}(t) + g_{\lambda}^{*} a_{2}(t), \tag{11}$$

$$\mathrm{i}\dot{a}_{\gamma}(t) = (\omega_{\gamma} - \omega_{21}) a_{\gamma}(t) + g_{\gamma}^{*} a_{2}(t). \tag{12}$$

Taking the Laplace transforms of Eqs. (9) and (10) we can obtain

$$a_{2}(s) = \frac{a_{2}(0) - \mathrm{i}\Omega^{*} a_{3}(0)/(s+\mathrm{i}\Delta)}{s + \Omega^{2}/(s+\mathrm{i}\Delta) + (\Gamma_{1}+\Gamma_{2})/2}. \tag{13}$$

Taking the Laplace transform of Eqs. (11) and (12) we have

$$\mathrm{i}sa_\lambda(s) = (\omega_\lambda - \omega_{20})a_\lambda(s) + g_\lambda^* a_2(s), \quad (14)$$

$$\mathrm{i}sa_\gamma(s) = (\omega_\gamma - \omega_{21})a_\gamma(s) + g_\gamma^* a_2(s). \quad (15)$$

In the deriving of the above two equations we have used the properties of Laplace transforms $L\{\dot{f}(t)\} = sf(s) - f(0)$. From final-value theorem $\lim_{t\to\infty} f(t) = \lim_{s\to 0} sf(s)$ we can obtain (setting $g_\lambda = g_1,\ g_\gamma = g_2$)

$$a_\lambda(t\to\infty) = g_1 \frac{b_2(0)(\delta_\lambda - \omega_{10}/2 - \Delta) + \Omega^* b_3(0)}{(\delta_\lambda - \omega_{10}/2)(\delta_\lambda - \omega_{10}/2 - \Delta) - \Omega^2 + \mathrm{i}(\Gamma_1/2 + \Gamma_2/2)(\delta_\lambda - \omega_{10}/2 - \Delta)}, \quad (16)$$

$$a_\gamma(t\to\infty) = g_2 \frac{b_2(0)(\delta_\gamma + \omega_{10}/2 - \Delta) + \Omega^* b_3(0)}{(\delta_\gamma + \omega_{10}/2)(\delta_\gamma + \omega_{10}/2 - \Delta) - \Omega^2 + \mathrm{i}(\Gamma_1/2 + \Gamma_2/2)(\delta_\gamma + \omega_{10}/2 - \Delta)}, \quad (17)$$

where $\delta_\lambda = \omega_\lambda - (\omega_{20} + \omega_{21})/2$ and $\delta_\gamma = \omega_\gamma - (\omega_{20} + \omega_{21})/2$. For convenience of the calculation of spectrum we set $g_1 = g_2 = \omega_{10} = c$, where c is an arbitrary constant. The individual spectral lines associated with transition $|2\rangle \to |0\rangle$ and $|2\rangle \to |1\rangle$ are denoted by $S(\delta_\lambda)$ and $S(\delta_\gamma)$, respectively. By using formulas $S(\delta_\lambda) = |a_\lambda(t\to\infty)|^2$ and $S(\delta_\gamma) = |a_\gamma(t\to\infty)|^2$ we can have the individual spectral lines and then discuss the influence of different initial states and detuning of external driving field on spontaneous emission spectrum, respectively.

In order to explicitly illustrate the origin of spontaneous properties of the atomic system under consideration, here we also derive the probability amplitude equations in the dressed state picture of driving field ν. In the dressed state picture, the levels $|2\rangle$ and $|3\rangle$ together with the driving field ν can be treated as an 'atom+field' system consisting of two close-lying dressed state levels $|+\rangle$ and $|-\rangle$ decaying to lower levels $|0\rangle$ and $|1\rangle$. The probability amplitudes of the two dressed states are given by

$$a_+ = a_3\cos\theta + a_2\sin\theta, \quad (18)$$

$$a_- = a_3\sin\theta - a_2\cos\theta, \quad (19)$$

where $\mathrm{tg}\theta = (\Delta/2 + \sqrt{\Delta^2/4 + \Omega^2})/\Omega$. With Eqs. (18) and (19) we can replace Eqs. (9) and (10) by

$$\dot{a}_+(t) = -\mathrm{i}(\Delta_+ - \mathrm{i}\Gamma_+)a_+ - \mathrm{i}\Gamma_{+-}a_-, \quad (20)$$

$$\dot{a}_-(t) = -\mathrm{i}(\Delta_- - \mathrm{i}\Gamma_-)a_- - \mathrm{i}\Gamma_{+-}a_+, \quad (21)$$

where $\Delta_+ = \Delta\cos^2\theta + 2\Omega\sin\theta\cos\theta$, $\Delta_- = \Delta\sin^2\theta - 2\Omega\sin\theta\cos\theta$, $\Gamma_+ = (\Gamma_1/2 + \Gamma_2/2)\sin^2\theta$, $\Gamma_- = (\Gamma_1/2 + \Gamma_2/2)\cos^2\theta$ and $\Gamma_{+-} = \Delta\sin\theta\cos\theta - \Omega(\cos^2\theta - \sin^2\theta) + \mathrm{i}(\Gamma_1/2 + \Gamma_2/2)\sin\theta\cos\theta$.

3. Results and discussion

Because the atom is embedded in free vacuum, the atomic system is entirely determined by the present state rather than the past state, that is, the atom loses all memories of its past. However, it should be noted that the past and the initial conditions are not equivalent to each other. In Markovian reservoir, the future of system is not related to its past, but to the initial condition of atom. Thus, we will discuss the properties of atomic spontaneous emission in different initial conditions.

When the atom initially is at level $|2\rangle$ i.e., $b_2(0) = 1$, the influence of detuning of external driving field on the individual spectral-lines is shown in Fig. 2. As shown in Fig. 2(a), it is found that in the resonance case, each individual spectral line shows a symmetrical double-peak structure. Furthermore, we find a dark line (zeros in the spectrum at a certain value of the emitted photon frequency) in each individual spectrum. The reason for appearing of the dark lines in the individual spectra can be obtained via analysing in the dressed state picture. In the dressed state picture, due to the existence of external driving field, the levels $|2\rangle$ and $|3\rangle$ can be replaced by two new states $|+\rangle$ and $|-\rangle$. Then, the transition $|2\rangle \to |0\rangle$ ($|2\rangle \to |1\rangle$) equals to two close-lying levels $|+\rangle$ and $|-\rangle$ decaying to $|0\rangle$ ($|1\rangle$), namely, each transition in a bare state corresponds to that of a V-type three-level structure. So, the dark lines in the individual spectra can be regarded as a consequence of destructive interference between the two decay channels in the V-type structure. In the nonresonant case, the peaks show the same heights, but the spectral line narrowing effect appears.

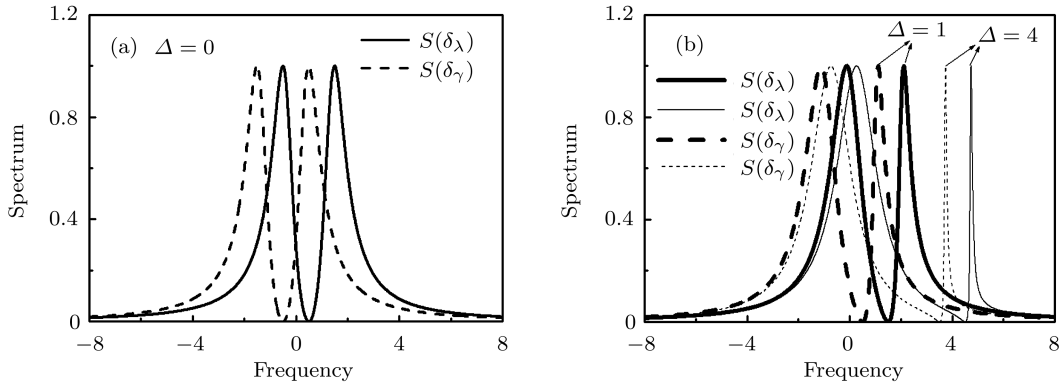


Fig. 2. Individual spectral lines for the atom initially being in the state $|2\rangle$, where the solid line (dash line) represents the spectrum line associated with the transition $|2\rangle \to |0\rangle$ ($|2\rangle \to |1\rangle$) for the resonant case (a) and the nonresonant case (b).

The influence of detuning of external driving field on individual spactral line is given as shown in Fig. 2(b). From Fig. 2(b) it is noted that when the external driving field is detuned away from the transition frequency ω_{23}, the individual spectral-lines are no longer fully symmetric. As the detuning increases from zero, the right peaks of individual spectral lines narrow while the others broaden, but all the peaks still have the same heights. These phenomena are quite similar to those in a driven two-level atomic system,[22] however, they did not give explanation about them. In our system, we try to give a qualitative explanation. In terms of dressed states, the dressed states $|+\rangle$ and $|-\rangle$ are the linear superposition of bare states $|2\rangle$ and $|3\rangle$. For the resonant case, the decay rates $\Gamma_+ = \Gamma_- = (\Gamma_1 + \Gamma_2)/4$. Thus, both the widths and the heights of two peaks in the individual spectral lines are equal. However, as the detuning increases, $\Gamma_+ = (\Gamma_1/2 + \Gamma_2/2)\sin^2\theta$ grows large but the $\Gamma_- = (\Gamma_1/2 + \Gamma_2/2)\cos^2\theta$ reduces, therefore the left peak broadens while the right peak narrows.

When the atom is initially in $|3\rangle$ i.e. $b_3(0) = 1$, we give the individual spectral lines in Fig. 3. In the resonance case as shown in Fig. 3(a), the individual spectra for both transitions each still have a two-peak structure. However, there is no dark line in the individual spectra due to constructive quantum interference between two decay channels in each V-type structure. In Fig. 3(b), in the small detuning case, the individual spectral-lines associated with transitions $|2\rangle \to |1\rangle$ and $|2\rangle \to |0\rangle$ still show two peaks, but the intensity of left peak decreases. These phenomena can also be explained in dressed state analysis. In the resonant case, the populations in dressed levels $|+\rangle$ and $|-\rangle$ are equal. As the detuning increases, the population depopulates in level $|+\rangle$, and so the corresponding emission spectral lines weaken. With the detuning further increasing to a larger value, the left peak almost vanishes.

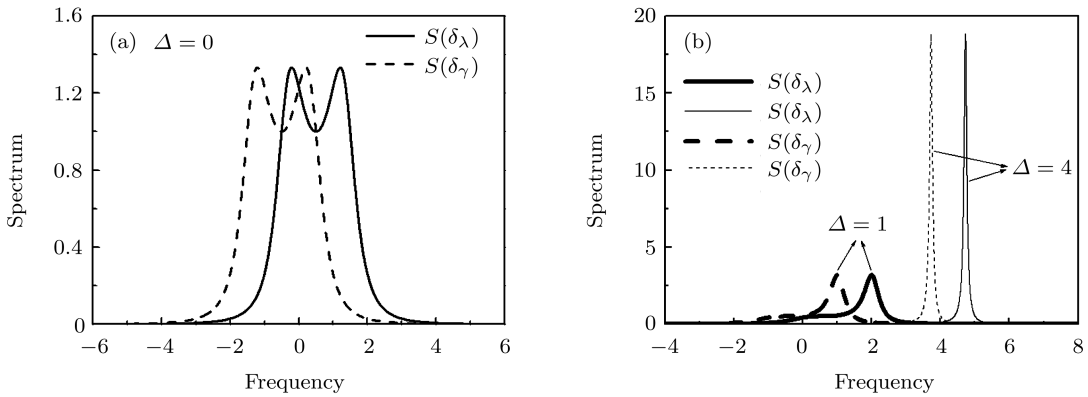


Fig. 3. Individual spectral lines for the atom initially being in the state $|3\rangle$, where the solid line (dashline) represents the spectral line associated with the transition $|2\rangle \to |0\rangle$ ($|2\rangle \to |1\rangle$) for the resonant (a) and the nonresonant cases (b). All the parameters are in units of c and c's in all the figures each are an arbitrary constant.

Figure 4 shows time evolutions of atomic population of dressed levels $|+\rangle$ and $|-\rangle$. It is found that as the detuning increases, the population transfer time from dressed level $|+\rangle$ to lower levels decreases, while for the transition from dressed level $|-\rangle$ to lower levels the time for the atom to reach the steady state becomes longer. These phenomena can also be proved by Γ_+ and Γ_-, as the detuning increases the Γ_+ increases but the Γ_- decreases, the corresponding relaxation time from $|+\rangle$ ($|-\rangle$) to lower levels decreases (increases).

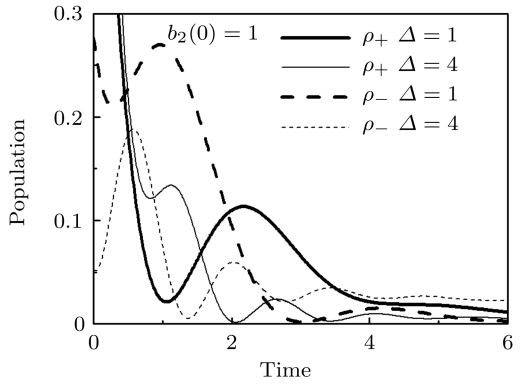


Fig. 4. Time evolutions of atomic population in the dressed levels $|+\rangle$ and $|-\rangle$.

4. Conclusion

The steady state behaviour of spontaneous emission and the time evolution of population from a driven four-level atom are studied, and the effects of quantum interference and detuning of external driving field on individual spectral line are discussed. In the individual spectral lines, the dark line originates from the destructive quantum interference between two spontaneous decay channels in each transition. For the larger detuning, the spectral line narrowing can be attributed to the fact that the corresponding spontaneous emission rate decreases. It is possible to find a real tripod four-level atom in an ^{87}Rb atomic system. The atomic levels $|5\,^2S_{1/2},\ F = 2,\ M_F = -2,-1,0\rangle$, $|5\,^2S_{1/2},\ F = 1,\ M_F = -1,0,+1\rangle$, and $|5\,^2S_{1/2},\ F = 2,\ M_F = 0,+1,+2\rangle$ are used as the three ground states in the tripod-type system, which are denoted as $|3\rangle$, $|0\rangle$, and $|1\rangle$. The atomic level $\left|5\,^2P_{1/2},\ F' = 1,\ M_F = -1,0,+1\right\rangle$ is used as the upper state in the system. The wavelength of driving field is 794 nm, which can be obtained from external cavity diode lasers.

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