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POPULATION TRAPPING IN MULTI-LEVEL SYSTEM VIA QUANTUM INTERFERENCE*

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Population trapping via quantum interference in a multi-level system driven by a coherent field is investigated. The influences of the alignment of the transition dipole moments and the energy-level separation of the upper levels are taken into account. Detailed trapping population profiles under the different conditions are given. The research result is helpful for simplifying the theoretical model and selecting rational energy level system.

PACC: 4250C; 3280B; 4250G; 4250L

I. INTRODUCTION

Population trapping and spontaneous emission cancellation via quantum interference is an active respearch topic in quantum optics.[1-10] The existence of a dark line is predicted in the spontaneous emission spectrum of a driven atom, when a pair of close-lying levels is coupled to a third level by a strong driving field.[2,3] The conditions exhibiting spontaneous emission cancellation and population trapping were theoretically studied by Zhu and Scully *et al*.[4-8] And the dressed-state picture is successfully introduced into theoretical analysis for the quantum interference.[4,7,9]

It is very difficult to find the suitable atomic energy-level system satisfying the conditions of quantum interference. So, molecular samples have been considered for the experiment in our laboratory.[10] However, the amount of the close-lying upper levels in a molecular sample is usually more than 2 because of the complexity of the molecular energy-level structure. Thus some questions arise: Is the contribution of each level in a multi-upper-level system to the quantum interference equivalent? Do the interference conditions need to be strictly satisfied? And how does the existence of a third upper level influence the interference effects between two upper levels? The answers of these questions would be of great value in identifying suitable molecular systems and experimental parameters.

In this paper, we study the trapping population in a five-level system with a simplified model, and give some important results for determining the suitable energy-level system and experimental parameters.

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II. THEORETICAL MODEL

A five-level system with three close-lying upper levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ is considered, as issustrated in Fig.1. The three upper levels are coupled by the same vacuum modes to the lower level $|4\rangle$ and driven by a coherent field with frequency ω to an auxiliary level $|g\rangle$. The five levels' characteristic frequencies are ω_1, ω_2, ω_3, ω_4 and ω_g, respectively.

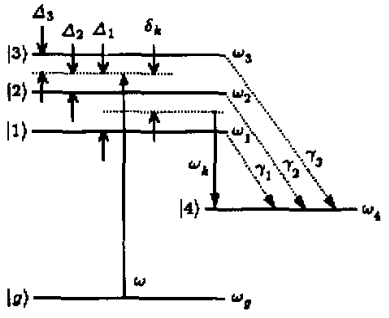

Fig.1. Energy-level diagram for the analysis

Using the similar theoretical steps given in Ref.[5], the equation of motion in the interaction picture for the amplitudes of the levels $|1\rangle$, $|2\rangle$, $|3\rangle$ and $|g\rangle$ can be derived as:

$$\frac{\mathrm{d}}{\mathrm{d}t}A(t) = TA(t)\,, \tag{1}$$

where

$$A(t) = \begin{pmatrix} a_g(t) \\ a_1(t)\mathrm{e}^{-i\Delta_1 t} \\ a_2(t)\mathrm{e}^{-i\Delta_2 t} \\ a_3(t)\mathrm{e}^{-i\Delta_3 t} \end{pmatrix}\,, \tag{2}$$

$$T = \begin{pmatrix} 0 & -\Omega_1^* & -\Omega_2^* & -\Omega_3^* \\ \Omega_1 & -\gamma_1/2 - i\Delta_1 & -p_{12}\sqrt{\gamma_1\gamma_2}/2 & -p_{13}\sqrt{\gamma_1\gamma_3}/2 \\ \Omega_2 & -p_{21}\sqrt{\gamma_2\gamma_1}/2 & -\gamma_2/2 - i\Delta_2 & -p_{23}\sqrt{\gamma_2\gamma_3}/2 \\ \Omega_3 & -p_{31}\sqrt{\gamma_3\gamma_1}/2 & -p_{32}\sqrt{\gamma_3\gamma_2}/2 & -\gamma_3/2 - i\Delta_3 \end{pmatrix}\,, \tag{3}$$

$a_1(t)$, $a_2(t)$, $a_3(t)$ and $a_g(t)$ are the amplitudes of the states at time t, respectively, Ω_i and γ_i $(i = 1, 2, 3)$ are the Rabi frequencies of the driving field and the radiative decay rates from the upper levels, respectively. Detunings of the driving field are $\Delta_i = \omega_i - \omega$, and $p_{ij} = \boldsymbol{\mu}_i \times \boldsymbol{\mu}_j / |\boldsymbol{\mu}_i| \times |\boldsymbol{\mu}_j|$ denotes the alignment of the matrix elements of two transition dipole moments $\boldsymbol{\mu}_i$ and $\boldsymbol{\mu}_j$.

Under the conditions of quantum interference shown below:

$$|p_{ij}| = 1, \quad p_{ij}p_{jk} = p_{ki}(i \neq j \neq k; i, j, k = 1, 2, 3)\,, \tag{4}$$

$$\frac{\Omega_i}{\Omega_j} = p_{ij}\sqrt{\frac{\gamma_i}{\gamma_j}}\,, \tag{5}$$

$$\frac{\Omega_1^2}{\Delta_1} + \frac{\Omega_2^2}{\Delta_2} + \frac{\Omega_3^2}{\Delta_3} = 0\,, \tag{6}$$

one can prove, that the characteristic equation $\det(T - \lambda I = 0)$ of matrix T has a zero solution. It results in the effects of population trapping and spontaneous emission cancellation in the discussed system.

Equation (6) shows the limitation for the driving frequencies for generating the quantum interference. On substituting Eqs.(4) and (5) into Eq.(6), we find

$$\sum_{i=1}^{3}\gamma_i\omega^2 - \sum_{i,j=1,j\neq i}^{3}\gamma_i\omega_j\omega + \sum_{i,j,k=1,k\neq j\neq i}^{3}\gamma_i\omega_j\omega_k = 0\,. \tag{7}$$

We have proved that Eq.(7) always has two real roots denoted as ω_{small} and ω_{big}, and they always satisfy $\omega_1 < \omega_{\rm small} \leq \omega_2 \leq \omega_{\rm big} < \omega_3$. This means there are two possible driving frequencies that result in quantum interference in the discussed system, rather than one frequency in a system with only two upper levels.

If the level spacing ω_{32} between two upper levels $|2\rangle$ and $|3\rangle$ is unlimitedly large (that is, Δ_3 is unlimitedly large), we can obtain from Eqs.(6) and (7):

$$\frac{\Omega_1^2}{\Delta_1} + \frac{\Omega_2^2}{\Delta_2} = 0\,, \tag{8}$$

$$\omega_{\rm small} = \frac{\gamma_1\omega_2 + \gamma_2\omega_1}{\gamma_1 + \gamma_2}\,. \tag{9}$$

They exactly coincide with the conditions of quantum interference in a four-level system studied in Ref.[5]. That is to say, the effects of quantum interference in our discussed system have the features in a four-level system when the level spacing ω_{32} is unlimitedly large.

The solution of Eq.(1) has the form

$$A_i = \sum_{j=1}^{4}\alpha_{i,j}\mathrm{e}^{-\lambda_j t}\,, \tag{10}$$

where λ_j's are the four roots of the secular equation corresponding to matrix T, the coefficients $\alpha_{i,j}$ are determined by the initial population $A(0)$ of the system. From Eq.(10) we can find, when the quantum interference is present in the system, the trapping population

$$\rho = \sum_i |A_i|^2_{t\to\infty} = \sum_{i,j=1}^{4}\left|\alpha_{i,j}\mathrm{e}{-\lambda_j t}\right|^2_{t\to\infty}\,. \tag{11}$$

III. RESULTS AND DISCUSSION

In the preceding section we have derived the expression of trapping population in the discussed system. To explore the unusual features produced by quantum interference, we

carry out a series of numerical calculation on Eq.(11). The calculated results are illustrated and discussed in this section.

Equations (10) and (11) show that the trapping population ρ is directly determined by the initial population $A(0)$ and the eigenvalue λ_j. Furthermore, the Rabi frequency Ω_i and the detuning Δ_i also affect the value of ρ by affecting the coefficients $\alpha_{i,j}$. We will concentrate on the effects of the alignment of the transition dipole moments p_{ij} (which affects λ_j) and the level spacing ω_{32} (which affects Δ_i) on the trapping population.

A. Effect of the alignment of transition dipole moments p_{ij}

Equation (10) shows that if all λ_j's are nonzero (namely, the system does not satisfy the interference conditions), then the population $|A_i|^2 \to 0$ in the limit $t \to \infty$, and there is no trapping population in the levels. On the other hand, if the interference conditions are well satisfied, one of λ_j's will be zero and it is possible to find nonzero population in every level when $t \to \infty$. The alignment of the transition dipole moments p_{ij} is the only condition of trapping population when the sytem satisfies Eqs.(4) and (5).

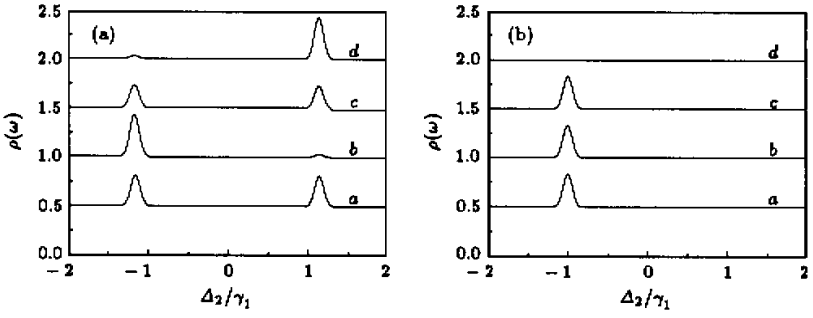

Fig.2. (a) Trapping population versus frequency ω of the driving field with $\gamma_3 = \gamma_2 = \gamma_1$, $\omega_{32} = \omega_{21} = 2\gamma_1$, $\Omega_3 = \Omega_2 = \Omega_1 = \gamma_1$ and $p_{ij} = 1$. The four curves corresponding to the initial populations in (a) $|g\rangle$, (b) $|1\rangle$, (c) $|2\rangle$ and (d) $|3\rangle$ are moved up by 0.5, 1, 1.5 and 2 units for clarity. (b) Same as (a), but with $p_{21} = 1$ and $p_{31} = p_{32} = 0$.

In Fig.2, we plot the trapping population ρ versus the driving frequency ω with several types of initial population and with $\gamma_3 = \gamma_2 = \gamma_1$, $\omega_{32} = \omega_{21} = 2\gamma_1$ and $\Omega_1 = \Omega_2 = \Omega_1 = \gamma_1$ when p_{ij} has two different values. Under such conditions, we obtain

$$\begin{aligned} \omega_{\text{small}} &< \frac{\omega_2 + \omega_1}{2}, \quad (\Delta_2/\gamma_1 < -1)\,; \\ \omega_{\text{big}} &> \frac{\omega_2 + \omega_3}{2}, \quad (\Delta_2/\gamma_1 > 1) \end{aligned} \tag{12}$$

from Eqs.(7) and (9).

For the case of $p_{ij} = 1$, the dipole moments from the three upper levles to $|4\rangle$ are parallel. Thus the quantum interference occurs so long as Eq.(7) is satisfied. It is easy to see from Fig.2(a) that in all cases of initial population the system has trapping population at frequencies ω_{small} and ω_{big}, and the initial population has strong influence on the values of trapping populations at the two frequencies. In principle, the quantum interference effect

of the whole system is the sum of the interference effects in two subsystems that consist of $|g\rangle$, $|1\rangle$, $|2\rangle$, $|4\rangle$ and $|g\rangle$, $|2\rangle$, $|3\rangle$, $|4\rangle$, respectively. The initial populations in $|g\rangle$ and $|2\rangle$ have equal contribution to the two subsystems, so the trapping population ρ at $\omega_{\rm small}$ is equal to ρ at $\omega_{\rm big}$, as shown in curves *a* and *c*. Whereas in curves *b* and *d*, the initial populations in $|1\rangle$ and $|3\rangle$ have different contributions to the two subsystems. Therefore ρ in $\omega_{\rm small}$ is not equal to ρ in $\omega_{\rm big}$, and the subsystem with more initial population shows stronger capability in population trapping.

In the case of $\boldsymbol{\mu}_1 /\!/ \boldsymbol{\mu}_2$ and $\boldsymbol{\mu}_1 \perp \boldsymbol{\mu}_3$, Eq.(4) is not satisfied, hence there is no population trapped at either $\omega_{\rm small}$ or $\omega_{\rm big}$, as shown in Fig.2(b). However, the condition of quantum interference Eq.(8) is satisfied in the subsystem with $|g\rangle$, $|1\rangle$, $|2\rangle$ and $|4\rangle$. As a result, there is population trapped at the interference frequency $\frac{\omega_1+\omega_2}{2}$ (namely $\Delta_2/\gamma_1 = -1$), as shown in curves *a*, *b* and *c*, under the condition that the upper level $|1\rangle$ or $|2\rangle$ share the initial population. In this case, the upper level $|3\rangle$ has no impact on the interference between levels $|1\rangle$ and $|2\rangle$. Only in curve *d*, because the initial population inhabits level $|3\rangle$ and $p_{31} = p_{32} = 0$, level $|3\rangle$ loses the couple with $|1\rangle$ and $|2\rangle$, and there is no population trapped at $\omega_{\rm small}$, $\omega_{\rm big}$ and $\frac{\omega_1+\omega_2}{2}$.

On the whole, when the level spacings between arbitrary two neighboring upper levels are nearly commensurate, the main influence of p_{ij} on quantum interference will be brought into play by changing the interference frequencies. In the case of $p_{ij} = 1$, the system exactly satisfies the interference conditions, and there are populations trapped at the frequencies determined by Eq.(7). In the case of $p_{21} = 1$ and $p_{31} = p_{32} = 0$, although the whole five-level system does not satsty its interference conditions, but the four-level subsystem does, and there are also populations trapped at the frequency determined by Eq.(9). In the second situation, level $|3\rangle$ has little influence on the interference between $|1\rangle$ and $|2\rangle$ unless it monopolizes all the initial population.

B. Effect of the level spacing ω_{32}

From Eq.(1), we can see that the detuning Δ_i of the driving field can affect the values of $\alpha_{i,j}$ and ρ, so $\omega_{32} = \Delta_3 - \Delta_2$ is certainly a key factor of quantum interference in the subsystem with levels $|g\rangle$, $|1\rangle$, $|2\rangle$ and $|4\rangle$.

In Fig.3, we plot the trapping population ρ versus ω_{32} with initial populations in the various levels when the system shown in Fig.1 always satisfies the interference conditions (4) and (5). For comparison, we hold the frequency of the driving field at the small solution of Eq.(7) in Fig.3(a) and at the solution of Eq.(9) in Fig.3(b).

In Fig.3(a), the system exactly satisfies the interference conditions (4), (5) and (6), hence there is always population trapped at $\omega_{\rm small}$ and the effect of ω_{32} is limited on the condition that level $|g\rangle$, $|1\rangle$ or $|2\rangle$ have shared some initial population (curves *a*, *b*, and *c*). The trapping population ρ tends to a constant while ω_{32} gradually increases. Only when level $|3\rangle$ monopolizes the initial population (curve *d*), ρ tends to zero while ω_{32} grows to a certain value.

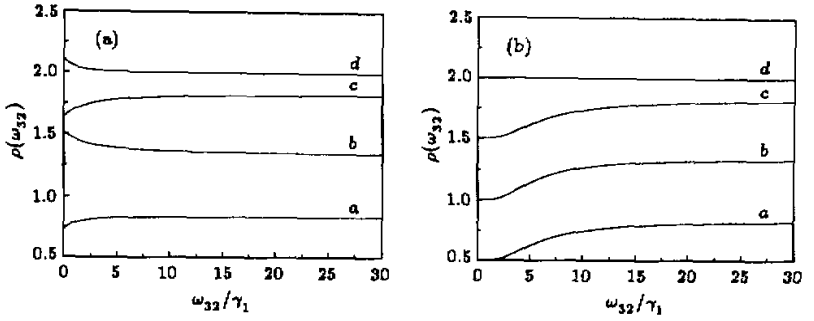

Fig.3. (a) Trapping population versus the spcaing ω_{32} with $\gamma_3 = \gamma_2 = \gamma_1$, $\omega_{21} = 2\gamma_1$, $\Omega_3 = \Omega_2 = \Omega_1 = \gamma_1$ and $p_{ij} = 1$. $\omega \equiv \omega_{small}$ is the smaller solution of Eq.(7). The four curves corresponding to the initial populations in (a) $|g\rangle$, (b) $|1\rangle$, (c) $|2\rangle$ and (d) $|3\rangle$ are moved up by 0.5, 1, 1.5 and 2 units for clarity. (b) Same as (a), but $\omega \equiv \frac{\omega_1 + \omega_2}{2}$ is the solution of Eq.(9).

In Fig.3(b), the interference condition (6) is not satisfied because the driving frequency is kept at $\frac{\omega_1 + \omega_2}{2}$. In this case, ω_{small} is not equal to $\frac{\omega_1 + \omega_2}{2}$ and the trapping population is zero when ω_{32} is a small value. However, while the spacing ω_{32} gradually increases, $\frac{\omega_1 + \omega_2}{2}$ tends to ω_{small} and we find some populations trapped in the system. When the value of ω_{32} is big enough, ρ tends to a constant and the curves in Fig.3(b) tend to the curves in Fig.3(a).

From the above discussion, it is easy to see that the influence of the spacing ω_{32} on quantum interference is determined by its value if the system satisfies Eqs.(4) and (5). When ω_{32} is fairly small, these are populations only trapped at the frequencies determined by Eq.(7). When ω_{32} gradually increases (e.g., from one to several times of ω_{21}), the influence of level $|3\rangle$ on quantum interference goes from strong to weak and there is also population trapped at the frequency determined by Eq.(9). On the whole, when ω_{32} is big enough (e.g., several tens of ω_{21}), the influence of level $|3\rangle$ vanishes and the five-level system degenerates to a four-level system with levels $|g\rangle$, $|1\rangle$, $|2\rangle$ and $|4\rangle$.

IV. CONCLUSION

We have studied the population trapping via quantum interference in a five-level system with three upper levels. The influences on population trapping of the initial population, the alignment of the transition dipole moments and the level spacing have been taken into account.

Our analysis suggests that in a multi-level system the influence of some energy levels on population trapping can be neglected under certain conditions. On the one hand, when one upper level is far away from the other upper levels, the influence of this level is quite small. On the other hand, the influence of some upper levels is ignorable under a certain alignment of the dipole moments of the transitions from the upper levels. These properties can be used to simplify the theoretical model and to rationally select the energy level system in experiment.

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