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SPECIAL LINE-SHAPES OF TWO-PHOTON TRANSITIONS IN A Y-TYPE FOUR-LEVEL SYSTEM BASED ON QUANTUM INTERFERENCE*

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The experimentally observed special line-shape induced by the interaction between two counter-propagating equal-frequency fields and an Y-type four-level system with two mixed upper levels has been analyzed theoretically. Serious line-shape variations of the two-photon transitions due to the quantum interference are discussed and predicted.

Keywords: quantum interference, two-photon transition, Y-type four-level system
PACC: 3280K, 3270

I. INTRODUCTION

In recent years, quantum interference in the multi-level system has become an active subject. Many novel physical phenomena,[1−9] such as spontaneous emission cancellation, coherent population trapping, electromagnetically induced transparency and lasing without inversion, based on quantum interference have been discovered. All energy-level configurations in relation to these phenomena and effects are atomic three-level schemes, such as V-type, Λ-type, and Ξ-type.

Using Doppler-free two-photon spectroscopy, the important experiment about spontaneous emission cancellation based on quantum interference in the Y-type four-level molecular system was performed in our laboratory.[10] The recorded two-photon transition line-shape without Doppler-free peak and with a large line-width showed obvious differences from the typical line-shape. The aim of this paper is to analyze the quantum interference and the subsequent line-shape variations of the two-photon transitions in an Y-type four-level system. The results are helpful to understand the observed special line-shape and the further application of such an energy-level scheme.

It's well know that the spin-orbit perturbation between the rotational levels with same J value in the single and triplet states, respectively, in a diatomic molecule (such as Na_2) gives rise to a pair of mixed levels not only close to each other but also with their wave function sharing. Thus we are able to investigate the quantum interference by using the special energy-level system with such a pair of mixed levels. The so-called Y-type four-level system includes a pair of upper mixed levels 3 and 4, an initial level 1, and an intermediate enhancing level 2 for the two-photon transition, as shown in Fig.1. By solving the density-matrix motion equation, we find that the two-photon transition line-shape in such a system has many differences from that in typical three-level system with only one upper level. Especially, the Doppler-free peak of the two-photon transition is broadened so seriously that we cannot distinguish it from the Doppler-broadened pedestal.

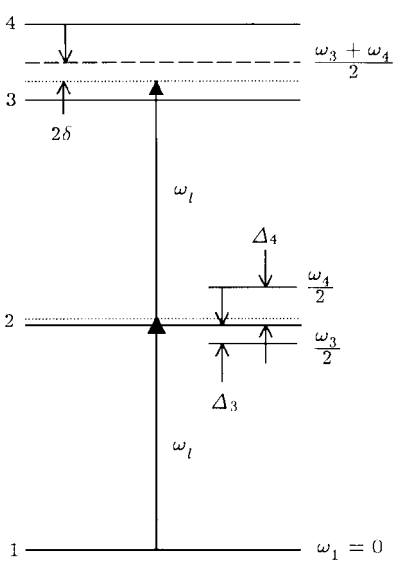


Fig.1. The Y-type four-level diagram for the analysis.

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II. CALCULATION AND RESULTS

In the four-level system shown in Fig.1, $\Delta_3 = \frac{\omega_3}{2} - \omega_2$ and $\Delta_4 = \frac{\omega_4}{2} - \omega_2$ denote the relative positions of the levels 2, 3 and 4, respectively. In the rotating wave and slowly varying envelope approximations, the density-matrix motion equations which describe the interaction between two equal-frequency fields (E_+ and E_- travelling in the opposite directions from a tunable single-mode laser) and the four-level system can be written as

$$\dot{\rho}_{11} = \Lambda_{11} - \gamma_1\rho_{11} - \mathrm{i}\Omega\tilde{\rho}_{12} + \mathrm{i}\Omega\tilde{\rho}_{21}, \tag{1a}$$

$$\begin{aligned}\dot{\rho}_{22} =&\Lambda_{22} - \gamma_2\rho_{22} - \mathrm{i}\Omega\tilde{\rho}_{23} + \mathrm{i}\Omega\tilde{\rho}_{32}\\ &+ \mathrm{i}\Omega\tilde{\rho}_{12} - \mathrm{i}\Omega\tilde{\rho}_{21} - \mathrm{i}\Omega\tilde{\rho}_{24} + \mathrm{i}\Omega\tilde{\rho}_{42},\end{aligned} \tag{1b}$$

$$\dot{\rho}_{33} = \Lambda_{33} - \gamma_3\rho_{33} + \mathrm{i}\Omega\tilde{\rho}_{23} - \mathrm{i}\Omega\tilde{\rho}_{32}, \tag{1c}$$

$$\dot{\rho}_{44} = \Lambda_{44} - \gamma_4\rho_{44} + \mathrm{i}\Omega\tilde{\rho}_{24} - \mathrm{i}\Omega\tilde{\rho}_{42}, \tag{1d}$$

$$\begin{aligned}\dot{\tilde{\rho}}_{23} =&- \mathrm{i}(\Delta_{23}^q - \mathrm{i}\gamma_{23})\tilde{\rho}_{23} - \mathrm{i}\Omega\rho_{22}\\ &+ \mathrm{i}\Omega\rho_{33} + \mathrm{i}\Omega\tilde{\rho}_{13} + \mathrm{i}\Omega\tilde{\rho}_{43},\end{aligned} \tag{1e}$$

$$\begin{aligned}\dot{\tilde{\rho}}_{12} =&- \mathrm{i}(\Delta_{12}^p - \mathrm{i}\gamma_{12})\tilde{\rho}_{12} - \mathrm{i}\Omega\rho_{11}\\ &+ \mathrm{i}\Omega\rho_{22} - \mathrm{i}\Omega\tilde{\rho}_{13} - \mathrm{i}\Omega\tilde{\rho}_{14},\end{aligned} \tag{1f}$$

$$\dot{\tilde{\rho}}_{13} = -\mathrm{i}(\Delta_{13}^{p+q} - \mathrm{i}\gamma_{13})\tilde{\rho}_{13} + \mathrm{i}\Omega\tilde{\rho}_{23} - \mathrm{i}\Omega\tilde{\rho}_{12}, \tag{1g}$$

$$\begin{aligned}\dot{\tilde{\rho}}_{24} =&- \mathrm{i}(\Delta_{24}^r - \mathrm{i}\gamma_{24})\tilde{\rho}_{24} - \mathrm{i}\Omega\rho_{22}\\ &+ \mathrm{i}\Omega\rho_{44} + \mathrm{i}\Omega\rho_{34} + \mathrm{i}\Omega\tilde{\rho}_{14},\end{aligned} \tag{1h}$$

$$\dot{\tilde{\rho}}_{34} = -\mathrm{i}(\omega_3 - \omega_4 - \mathrm{i}\gamma_{34})\rho_{34} - \mathrm{i}\Omega\tilde{\rho}_{32} + \mathrm{i}\Omega\tilde{\rho}_{24}, \tag{1i}$$

$$\dot{\tilde{\rho}}_{14} = -\mathrm{i}(\Delta_{14}^{p+r} - \mathrm{i}\gamma_{14})\tilde{\rho}_{14} - \mathrm{i}\Omega\tilde{\rho}_{12} - \mathrm{i}\Omega\tilde{\rho}_{24}, \tag{1j}$$

where Ω is the Rabi frequency, $\Lambda_i = \Lambda_{ij}(i = j)$ is the thermal excitation rate of level i, and $\gamma_i = \gamma_{ij}(i = j)$ is the relaxation rate of level i. Ignoring collisional dephasing, $\gamma_{ij} = (\gamma_i + \gamma_j)/2$. The detunings in Eq.(1) are defined as $\Delta_{ij}^l = \omega + lkv_z + \omega_i - \omega_j (ij = 12, 23, 24; l = p, q, r)$ and $\Delta_{1m}^{s+t} = 2\omega + (s + t)kv_z + \omega_1 - \omega_m (m = 3, 4; s, t = p, q, r)$, where ω is laser frequency, and p, q and r denote the two directions of the travelling waves and have the value 1 and (or) –1, respectively. The sign '$\sim$' denotes the slowly varying amplitudes of the off-diagonal matrix elements oscillating with optical frequency.

Under the steady-state conditions, we can obtain the following expressions for the populations in the upper levels 3 and 4 from Eq.(1)

$$\rho_{33} = \frac{\Omega^2\left(\dfrac{\tilde{\rho}_{13} + \rho_{43} - \rho_{22}}{\Delta_{23} - \mathrm{i}\gamma_{23}} + \dfrac{\tilde{\rho}_{31} + \rho_{34} + \rho_{22}}{-\Delta_{23} - \mathrm{i}\gamma_{32}}\right)}{\dfrac{\Omega^2}{-\Delta_{23} - \mathrm{i}\gamma_{32}} - \dfrac{\Omega^2}{\Delta_{23} - \mathrm{i}\gamma_{23}} - \mathrm{i}\gamma_3}, \tag{2a}$$

$$\rho_{44} = \frac{\Omega^2\left(\dfrac{\tilde{\rho}_{14} + \rho_{34} - \rho_{22}}{\Delta_{24} - \mathrm{i}\gamma_{24}} + \dfrac{\tilde{\rho}_{41} + \rho_{43} + \rho_{22}}{-\Delta_{24} - \mathrm{i}\gamma_{42}}\right)}{\dfrac{\Omega^2}{-\Delta_{24} - \mathrm{i}\gamma_{42}} - \dfrac{\Omega^2}{\Delta_{24} - \mathrm{i}\gamma_{24}} - \mathrm{i}\gamma_4}. \tag{2b}$$

From Eq.(2) we can obviously see that the nonzero off-diagonal elements ρ_{34} and $\rho_{43}(\rho_{43} = \rho_{34}^*)$ of the density matrix appear in the expressions for the upper level populations. That is to say, the quantum interference due to the coherent superimposition between the two upper levels 3 and 4 has direct contribution to the upper level populations. We can also find from Eq.(1)

$$\rho_{34} = \frac{\Omega^2\left(\dfrac{\tilde{\rho}_{14} + \rho_{44} - \rho_{22}}{\Delta_{24} - \mathrm{i}\gamma_{24}} + \dfrac{\tilde{\rho}_{31} - \rho_{33} + \rho_{22}}{-\Delta_{23} - \mathrm{i}\gamma_{32}}\right)}{(\omega_3 - \omega_4 - \mathrm{i}\gamma_{34}) - \dfrac{\Omega^2}{\Delta_{24} - \mathrm{i}\gamma_{24}} - \dfrac{\Omega^2}{-\Delta_{23} - \mathrm{i}\gamma_{32}}}. \tag{3}$$

It can be seen that the expressions of ρ_{33}, ρ_{44} and ρ_{34} display the feature of a linear combination. Thus the populations in the upper levels 3 and 4 can coherently transfer to each other under the influence of the quantum interference characterized by ρ_{34} and ρ_{43}.

In order to predict the influence of the coherent superimposition on the upper level populations, we take the full numerical calculations to $|\rho_{34}|^2$, $|\rho_{33}|^2$ and $|\rho_{44}|^2$. Doppler broadening is taken into account by integrating over the velocity distribution (i.e., over V_z). The total populations in levels 3 and 4 are obtained by adding up over the different travelling directions of the laser beams. The calculated results are shown in Figs.2 and 3.

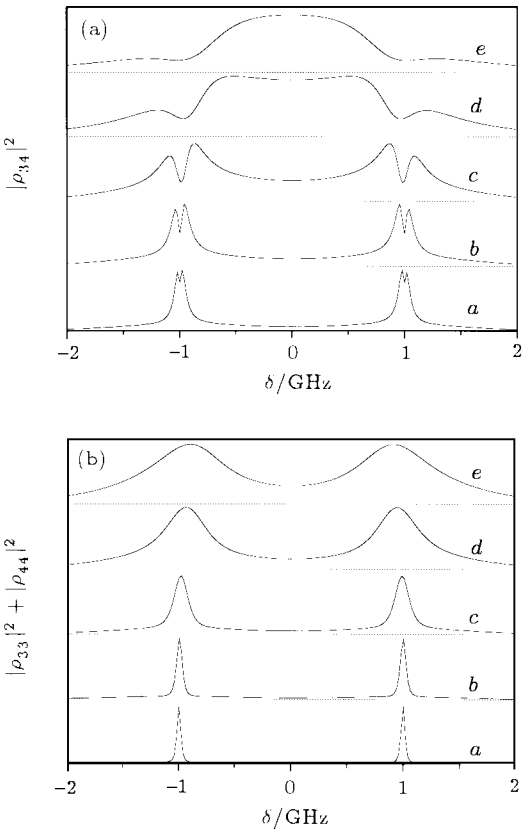


Fig.2. The calculated normalized traces of $|\rho_{34}|^2$ (a) and $(|\rho_{33}|^2 + |\rho_{44}|^2)$ (b) under different Rabi frequencies for $\omega_{43} = 4\text{GHz}, \gamma_1 = \gamma_2 = \gamma_3 = 1/15\text{GHz}$, $-\Delta_3 = \Delta_4 = 1\text{GHz}$ and $\Omega = 0.05, 0.1, 0.2, 0.5, 0.8\text{GHz}$ (traces a to e, respectively).

Figures 2(a) and 2(b) display the predicted traces of $|\rho_{34}|^2$ and $(|\rho_{33}|^2+|\rho_{44}|^2)$ against laser frequency detuning $\delta(2\delta = 2\omega - \frac{\omega_3+\omega_4}{2})$ under different Rabi frequencies. From Fig.2(a), we can see that each trace of $|\rho_{34}|^2$ has a center at $\omega_0 = \frac{\omega_3+\omega_4}{2}$ (center position of the two upper levels) and two dips at the intermediate resonant frequencies $\frac{\omega_3}{2}$ and $\frac{\omega_4}{2}$ of the two-photon transitions. The two dips may superimpose with the Doppler-free peaks which should be produced by the two independent three-level systems composed of levels 1, 2, 3 and levels 1, 2, 4, respectively. As the Rabi frequency Ω gradually increases (curves a to e), the widths of the two dips and the value of $|\rho_{34}|^2$ around the center of the two upper levels increase simultaneously. When the driving field is strong enough (curve e), $|\rho_{34}|^2$ has a maximum at ω_0. As a result, the Doppler-free peaks can be obviously broadened and lowered. Such line-shape variations due to the quantum interference can be clearly seen in the calculated Fig.2(b). When the Rabi frequency Ω has a small value (curve a), the calculated line-shape is similar to that of the typical three-level system. However, as Ω gradually increases, the value of $|\rho_{34}|^2$ increases and the coherent population transfer between levels 3 and 4 becomes obvious (curves b to e). Meanwhile the Doppler-free peaks of the two-photon transitions are broadened so seriously that we cannot distinguish them from the Doppler-broadened pedestals.

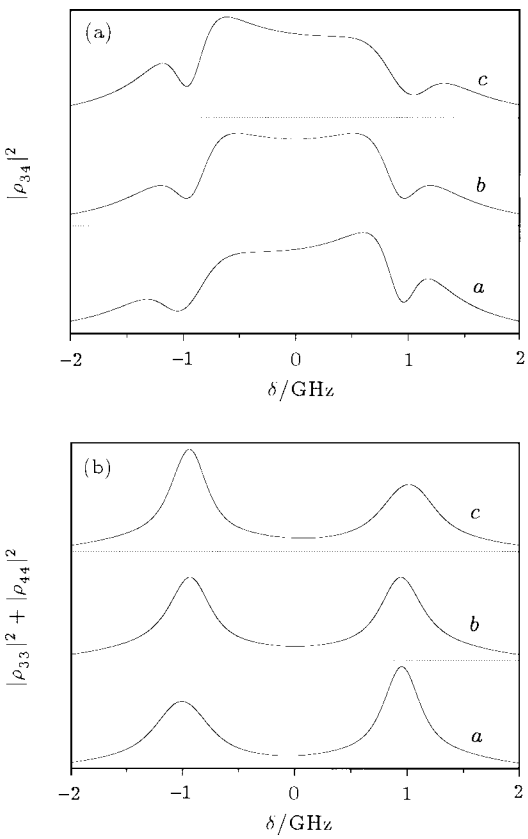


Fig.3. The calculated normalized traces of $|\rho_{34}|^2$ (a) and $(|\rho_{33}|^2 + |\rho_{44}|^2)$ (b) under different alignments of the two upper levels for $\omega_{43} = 4\text{GHz}, \gamma_1 = \gamma_2 = \gamma_3 = 1/15\text{GHz}, \omega_{43} = 4\text{GHz}, \Omega = 0.5\text{GHz}$ and $\Delta_3 = 0, 1, 2\text{GHz}$ (traces a to c, respectively).

On the other hand, the alignment of the two upper levels has also influences on the population distribution between the two upper levels. Figures 3(a) and 3(b) display the predicted traces of $|\rho_{34}|^2$, $|\rho_{33}|^2$ and $|\rho_{44}|^2$ against laser detuning δ under different alignments. We can see that when the alignment satisfies $\Delta_3 = 0$ (curve a), the line-shape of $|\rho_{34}|^2$ is asymmetric. The dip corresponding to level 3 is wider than that corresponding to level 4, thus making the broadening and lowering of the Doppler-free peak on the left side of the profile of $(|\rho_{33}|^2+|\rho_{44}|^2)$ more serious. In contrast, when the alignment satisfies $\Delta_4 = 0$ (curve c), the broadening and lowering of the Doppler-free peak on the right side are more serious. Only when $\Delta_3 = -\Delta_4$ (curve b), the line-shape variations are equivalent.

Ⅲ. CONCLUSION

In summary, the quantum interference due to the coherent superimposition between the two upper levels in the Y-type four-level system can result in the line-shape variations of the two-photon transitions. Different from the typical three-level system, there are certain populations between the two upper levels, and the Doppler-free peaks of the two-photon transitions are seriously broadened. It is helpful for the understanding the phenomena of the line-shapes seriously broadened, as shown in Ref.[10]. It is also shown that the novel spectral effects depend on the laser Rabi frequency and the alignment of the two upper levels.

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