QUANTUM INTERFERENCE EFFECTS IN MOLECULAR Y- AND RHOMB-TYPE SYSTEMS

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Abstract

In this paper we report the first observation of molecular population trapping in four-level systems. Constructive and destructive quantum interferences between two sum-frequncy two-photon transitions in Y- and rhomb-type four-level systems, respectively, im sodium molecules have been experimentally achieved by using only one laser source. Their energy-level schemes are featured by the extremely near-rsonant enhancement of the equal-frequency two-photon transitions, sharing both the initial and the intermediate levels for the Y-type, and sharing both the initial and the final levels for the rhomb-type systems. Their novel spectral effects are to show seriously restrained Doppler-free UV peak at the nominal location of the induced two-photon transition with visible fluorescence in rhomb-type schems, and to show a strong extra UV peak but null visible fluorescence in the middle between the two dipole allowed two-photon transitions.

1 Introduction

In last three decades many physical phenomena have been discovered for a multi-level quantum system drived by coherent light field. Among them, coherent population trapping (CPT) in a Λ -type and V- type three-level system has been an interesting topic in the field of quantum optics for many years [1]-[8]. The significance of the topic, in addition to be interested by basic research for laser-matter interactions, deals with the recently interested topics such as laser without inversion [9]-[12], quantum interference and new phenomena [13]-[14]. However, so far all of the experimental demonstrations for CPT are for a V-type three-level system in atomic samples [3]-[8] by using two sets of lasers.

In this paper we report the first observation of molecular population trapping in four-level systems. We use molecules as the sample for taking the advantages of their abundant selectable gradual changing energy-level schems. Constructive and destructive quantum interferences between two sum-frequncy two-photon transitions in Y- and rhomb-type four-level systems, respectively, in sodium molecules have been experimentally achieved by using only one laser source. Their energy-level schemes are featured by the extremely near-rsonant enhancement of the equal-frequency two-photon transitions, sharing both the initial and the intermediate levels for the Y-type, and sharing both the initial and the final levels for the rhomb-type systems. Their novel spectral effects are to show seriously restrained Doppler-free UV peak at the nominal location of the induced two-photon transition with visible fluorescence in rhomb-type schems, and to show a strong extra UV peak but null visible fluorescence in the middle between the two dipole allowed two-photon transitions.

2 Theory

The schems available in sodium dimers for our study is attributing to the existence of the spin-orbital perturbation between the rotational levels with same J in the singlet and triplet states, located in available dye laser regions. Such a mutual perturbation can form a pair of levels, not only close each other but also with their wavefunctions sharing. So that, once they have proper frequency location as the final level or as the intermediate levels for near-resonantly enhanced two-photon transition for the so called Y-type or rhomb-type four-level systems, respectively, they will show their characteristic quantum interference effects in their observable lineshape patterns. Indeed, we have found a series schemes with gradually changing parameters in Na_2 for study each of the models.

In the calculation with density matrix equations, we use the form of the interacting Hamiltonian

$$H_Y^I = \begin{bmatrix} 0 & -\mu_{ab}E & 0 & 0 \\ -\mu_{ba}E & 0 & \mu_{bc}E & \mu_{bd}E \\ 0 & -\mu_{cb}E & 0 & 0 \\ 0 & -\mu_{db}E & 0 & 0 \end{bmatrix}$$

for the Y-type four-level system, where c and d are the perturbation coupled levels in the molecular high-lying states. Similarly, we use the interacting Hamiltonian of

$$H_R^I = \begin{bmatrix} 0 & -\mu_{ab}E & -\mu_{ac}E & 0 \\ -\mu_{ba}E & 0 & 0 & -\mu_{bd}E \\ -\mu_{ca}E & 0 & 0 & -\mu_{cd}E \\ 0 & -\mu_{db}E & -\mu_{dc}E & 0 \end{bmatrix}$$

for the rhomb-type four-level system, where b and c are the perturbation coupled levels in the molecular intermediate states. Substituting these forms, instead of the known form as

$$H_1^I = \begin{bmatrix} 0 & -\mu_{ab}E & 0 \\ -\mu_{ba}E & 0 & \mu_{bd}E \\ 0 & -\mu_{db}E & 0 \end{bmatrix} + \begin{bmatrix} 0 & -\mu_{ac}E & 0 \\ -\mu_{ca}E & 0 & \mu_{cd}E \\ 0 & -\mu_{dc}E & 0 \end{bmatrix}$$

for two independent two-photon transitions individually enhanced by the middle level b and c in two three-level systems, or the form as

$$H_2^I = \begin{bmatrix} 0 & -\mu_{ab}E & 0 \\ -\mu_{ba}E & 0 & \mu_{bc}E \\ 0 & -\mu_{cb}E & 0 \end{bmatrix} + \begin{bmatrix} 0 & -\mu_{ab}E & 0 \\ -\mu_{ba}E & 0 & \mu_{bd}E \\ 0 & -\mu_{db}E & 0 \end{bmatrix}$$

for two independent two-phton transitions reaching separated upper levels in two three-level systems, we get different results.

The calculation with H_Y^I for the steady state solution of the density matrix equations reveals the existing constructive quantum interference as showing an extra UV peak, origining from the non zero and non diagonal element ($\rho_{cd} \neq 0$), and predicts its maximum location right in the middle between the two usual lines, according to H_1^I. The dependence of the term on the perturbation coupling coefficients is also obtained.

The calculations with H_R^I for the steady state solution of the density matrix equations reveals that the destructive quantum interference can completely cancel each other for their enhancement for the two-photon transition from a to d, origining from the non zero and non diagonal element ($\rho_{bc} \neq 0$). The dependence of the phenomenon on the relative detunings and the signs between the two enhancements are obtained.

3 Experimental Demonstrations

The experiments are performed by using an Argon ion laser pumped single mode scannable dye laser at R6G and DCM dye regions and with a four-arm stainless steel oven containing sodium. For study both constructive and destructive quantum interferences mentioned above we search to find two serierses of the coupled levels consisting of paired spin-orbital perturbation levels with small separations from tens MHz to few GHz [15]-[18].

We observed the constructive quantum interference characterized by showing a strong extra fluorescence peak with null visible emission in the middle between the dipole allowed signals of the sum-frequency two-phton transitions, as predicted by the calculations. The relative intensity of the extra signal to the dipole allowed signals is determined by the degree of the wavefunction coupling as shown by the upper traces in Fig.1 for two distinct cases : The left trace is for 20% wavefunction sharing, whereas the right for 40%. The observed serious pressure influnce on the signal intensity revealed the disparity pressure-shift among these levels.

The destructive quantum interference was characterized by showing varying location of the Doppler-free peak on its Doppler-broadened peddestal, accompanied by the varying reduction (until complete null !) from the sharp UV peak, in a series of the observed near-resonantly enhanced two-photon absorption lines. The spectral patterns reveal that destructive quantum interference is dominated by the magnitudes as well as the relative signs of the detunings of the intermediate levels from two-phton resonance. The lower traces in Fig.1 present the degree different of the destructive interference for two distinct cases : The left trace is resulted by disparity detunings, whereas the right is with comparable magnitudes but opposit signs of the intermediate detunings.

In conclusion, molecular population trapping in intermidiate and in high-lying Rydberg states can be sufficiently achieved, especially via the mechanism of quantum interference between extremely near-esonantly enhanced two-photon absorptions, in comparison with that in atomic two-photon transitions.

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