Permanent dipole moments induced stability and group index switching in a three-level molecule

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In this paper, the effect of permanent dipole moments on optical bistability of weak probe light in a unidirectional ring cavity with three-level molecule is investigated. It is found that the intensity threshold of optical bistability (OB) and optical multistability (OM) can be controlled by quantum coherence induced by permanent dipole moments. Also, it is shown that switching from OB to OM and lasing without population inversion can be occurred by this coherence under different conditions.

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Introduction. Electromagnetically induced transparency (EIT) [1] is an important quantum interference effect which renders a medium transparent over a narrow spectral region within an absorption line. Great efforts were made in this area and its wide applications attracted considerable attention, such as laser without inversion (LWI) [2], four-wave mixing (FWM) [3–5], electron localization [6–9], optical bistability (OB) and optical multistability (OM) [10-18] and so on [19-30]. Optical bistability has been extensively developed due to its potential application in many optical switches and optical transistors, which are very useful devices in quantum computing and quantum communications. Up until now, there have been a large number of theoretical contributions and experimental demonstration of OB in atomic gases [31–33], ion-doped crystals [34–36] and semiconductors [37–40]. Li et al. [10] showed that quantum interference can be used to modifying intensity threshold of optical bistability and multistability. Joshi and Xiao showed that OB in multi-level systems has advantages over the two-level systems [14]. In ion-doped crystals, Wang [34] investigated the optical bistability in an Er³⁺-doped yttrium-aluminum-garnet (YAG) crystal inside a unidirectional ring cavity. He found that the intensity and the frequency detuning of the coherent field as well as the rate of incoherent field can affect the optical bistability dramatically, which can be used to manipulate efficiently the threshold intensity and the hysteresis loop. In our recent study [35], we study the effect of Er³⁺ ion concentration on optical bistability and multistability in Er³⁺ doped YAG crystal. In semiconductors, Li et al. studied the behavior of OB in a triple semiconductor quantum well structure with tunneling induced interference [37], Wang and Yu, reported OB behavior in an asymmetric three-coupled quantum well inside a unidirectional ring cavity via coherent driven field [40].

A dipolar molecule refers to a system with states that are not parity eigenstates and have nonzero difference d between the diagonal dipole matrix elements (permanent dipole moments). Permanent dipole moments (PDMs) are responsible for some new optical properties which are different from a regular atomic system. It is shown that μ_{jj} , the PDMs can affect the molecular-laser coupling and allow transitions to occur in such media which would otherwise be forbidden [41– 43]. Thus not only significant differences can occur between systems with and without μ_{ij} but also new mechanisms for some phenomena can be introduced, for instance, two-photon absorption [41], two-photon phase conjugation [44], EIT [45]. To investigation and importance of permanent moments in multiphoton absorption the perturbation theory is used by Meath and power [46]. A rotating-wave approximation (RWA) was derived for the single-photon and multiphoton resonance profiles arising from the interaction between a two-level dipolar molecule and an applied continuous-wave laser [47, 48]. The transition rates and the linear and nonlinear optical properties of a homogeneously broadened two-level system with permanent dipole moments in the presence of two-pump fields are discussed [49]. Based on a perturbative solution of the density-matrix equations, Lavoine et al., studied a two-photon degenerate four-wave mixing process that occurs in an isotropic media modeled by homogeneously broadened two-level system having unequal permanents [50]. Zhou et al. investigated EIT in a three-level Λ -type molecular system with nonzero permanent dipole moments. They found in the (2+2)-

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Fig. 1. (a) – Schematic diagram for the three-level dipole molecule. (b) – Unidirectional ring cavity with a dipole molecule sample of length L

transition processes, when the sign of d_{21} , the difference in permanent dipole moments of the probe transition, is positive, perfect EIT with steep normal dispersion could be obtained under specific conditions [45]. Ma et al. studied the effect of quantum coherence induced by permanent dipole moments in a three-level dipolar molecule. They discussed about absorption and dispersion properties under different conditions [51]. To the best of our knowledge, in all the previous works rarely attention has been paid to OB and OM in polar molecules. In this paper, we investigate the OB properties of a three-level dipole molecule. It is found that switching from OB to OM can be obtained by PDMs. Moreover, the transmission, dispersion and group index of weak probe light in the presence of permanent dipole moments is discussed. We found that under the quantum coherence induced by permanent dipole moments leads to probe gain instead of usual absorption.

Model and equations. A three-level ladder dipole molecule with ground state $|1\rangle$ and excited states $|2\rangle$ and $|3\rangle$ are illustrated in Fig. 1a. A strong coupling field of frequency ν_c is applied on transition $|2\rangle \rightarrow |3\rangle$ with frequency ω_{32} . The transition $|1\rangle \rightarrow 2\rangle$ of frequency ω_{21} is driven by a weak probe laser of frequency ν_p . The detunings between the field and the system frequencies are given by: $\delta_c = \nu_c - \omega_{32}$, and $\delta_p = \nu_p - \omega_{21}$. Γ_2 and Γ_3 are the spontaneous decay rates of the states $|2\rangle$ and $|3\rangle$, respectively. Under the rotating-wave approximation, the density-matrix equations of motion can be written as:

$$\begin{split} \dot{\rho}_{11} &= \frac{i}{2} [\Omega_p \rho_{12} - \Omega_p \rho_{21}] + \Gamma_2 \rho_{22} + \Gamma_3 \rho_{33}, \\ \dot{\rho}_{21} &= \frac{i}{2} [\Omega_o (\rho_{22} - \rho_{11}) - \Omega_c \rho_{31}] - \left\{ \gamma_{21} + \right. \\ &+ i \left[-\delta_p + \frac{\mu_{22} - \mu_{11}}{\mu_{21}} \frac{\Omega_p}{2} \exp(-i\nu_p t) + \right] \end{split}$$

 $+ \frac{\mu_{22}}{\mu_{32}} \frac{\Omega_c}{2} \exp(-i\nu_c t) \bigg] \bigg\} \rho_{21},$ $\dot{\rho}_{22} = \frac{i}{2} [\Omega_c \rho_{23} - \Omega_c \rho_{32} + \Omega_p \rho_{21} - \Omega_p \rho_{12}] - \Gamma_2 \rho_{22},$ $\dot{\rho}_{31} = \frac{i}{2} [\Omega_p \rho_{32} - \Omega_c \rho_{21}] - \bigg\{ \gamma_{32} - i \bigg[\delta_p + \delta_c + \frac{\mu_{11}}{\mu_{21}} \frac{\Omega_p}{2} \times \\ \times \exp(-i\nu_p t) - \frac{\mu_{33}}{\mu_{32}} \frac{\Omega_c}{2} \exp(-i\nu_c t) \bigg] \bigg\} \rho_{31},$ $\dot{\rho}_{32} = \frac{i}{2} [\Omega_c (\rho_{33} - \rho_{22}) + \Omega_p \rho_{31}] - \bigg\{ \gamma_{32} - \\ - i \bigg[\delta_c - \frac{\mu_{33} - \mu_{22}}{\mu_{32}} \frac{\Omega_c}{2} \exp(-i\nu_c t) + \\ + \frac{\mu_{22}}{\mu_{21}} \frac{\Omega_p}{2} \exp(-i\nu_p t) \bigg] \bigg\} \rho_{32},$ $\dot{\rho}_{33} = \frac{i}{2} [\Omega_c \rho_{32} - \Omega_c \rho_{23}] - \Gamma_3 \rho_{33},$ (1)

where the Rabi-frequency $\Omega_c = 2\mu_{32}E_c/\hbar$, $\Omega_p = 2\mu_{21}E_p/\hbar$, and $\gamma_{jj} = (\Gamma_i + \Gamma_j)/2$. For the convenience of the calculation, we have assumed that the Rabi frequency Ω_p and Ω_c are real. There are complex exponential terms in the equations which are time-dependent, so it is difficult to get analytical solution. In order to obtain the density matrix elements in the steady state, the above equations can be solved numerically at asymptotic time, i.e. the time which is much greater than the largest characteristic time of the molecule. These set of equation can be solved numerically to obtain the steady state response of the medium. In fact, the response of the medium to the applied fields is determined by the susceptibility χ , which defines as:

$$\chi = \frac{2N\mathfrak{P}_{12}}{\varepsilon_0 E_p}\rho_{21},\tag{2}$$

where N is the atomic density number in the medium, and $\chi = \chi' + i\chi''$. The Rabi frequency Ω_p of the probe

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laser field along the propagating direction of "z" obeys the following MaxwellTs wave equation

$$\frac{\partial \Omega_p}{\partial z} + \frac{1}{c} \frac{\partial \Omega_p}{\partial t} = i\alpha\gamma\rho_{21},\tag{3a}$$

where c is the permittivity of free space the light speed and $\alpha = N|\mathfrak{p}_{21}|^2 \nu_p/2\hbar\varepsilon_0 c\gamma$ is the propagation constant of the probe field, respectively. In the linear regime, for given $\Omega_p^{\rm in}$ at the atomic medium input z = 0. We can easily arrive at the steady state probe field $\Omega_p^{\rm out}$ at the output z = L

$$\Omega_p^{\text{out}} = \Omega_p^{\text{in}} \exp\left[-\alpha L\gamma \operatorname{Im}\left(\frac{\rho_{21}}{\Omega_p}\right)\right],\qquad(3b)$$

where L denotes the length of the atomic sample. Then the normalized transmission coefficient of the probe laser field for the transition $|2\rangle \leftrightarrow |1\rangle$ can be derived from expression (3b) as

$$T_p = \frac{\Omega_p^{\text{out}}}{\Omega_p^{\text{in}}}.$$
 (3c)

Now, we consider a medium of length L composed of the above described molecular system immersed in unidirectional ring cavity as shown in Fig. 1b. For simplicity, we assume that the both mirrors 3 and 4 are perfect reflectors, and the reflection and transmission coefficients of mirrors 1 and 2 are R and T (with R + T = 1), respectively. The total electromagnetic field for the probe beam can be written as:

$$E_{p}e^{-i(\nu_{p}t-k_{p}z)} + E_{c}e^{-i(\nu_{c}t-k_{c}z)} + \text{c.c.}, \qquad (4)$$

where E_p is the amplitude of probe field, which circulate in the ring cavity and E_c is a control field and is not circulate in the cavity. Under slowly varying envelop approximation, the dynamics response of the probe beam is governed by Maxwell's equations:

$$\frac{\partial E_p}{\partial t} + c \frac{\partial E_p}{\partial z} = \frac{i\omega_p}{2\varepsilon_0} P(\nu_p), \tag{5}$$

where $P(\nu_p)$ is the induced polarization, $P(\nu_p) = N\mathfrak{P}_{21}\rho_{21}$. For a perfectly tuned cavity, the boundary conditions in the steady-state limit between the incident field E_P^I and transmitted field E_P^T are [12–15]:

$$E_p(L) = \frac{E_p^T}{\sqrt{T}},\tag{6a}$$

$$E_p(0) = \sqrt{T}E_P^I + RE_p(L), \tag{6b}$$

where L is the length of the atomic sample. Note that the second term on the right-hand side of Eq. (6b) is the feedback mechanism due to the reflection from mirrors. It is responsible for the bistable behavior, so we do not expect any bistability when R = 0 in Eq. (6b). According to the mean-field limit and by using the boundary condition, the steady state behavior of elliptically polarized transmitted field is given by:

$$y = 2x - iC\rho_{21},\tag{7}$$

where $y = \mathfrak{P}_{21}E_p^I/\hbar\sqrt{T}$ and $x = \mathfrak{P}_{21}E_p^T/\hbar\sqrt{T}$ are the normalized input and output field, respectively. The parameter $C = N\nu_p L\mathfrak{P}_{21}^2/2\hbar\varepsilon_0 cT$ is the cooperatively parameter for atoms in a ring cavity. Transmitted field depends on the incident probe field and the coherence terms ρ_{21} via Eq. (7).

Results and discussion. A molecule system which is similar to the three-level dipole molecule in [52] is considered. It is assumed that the population at the initial time is in the ground state $|1\rangle$, i.e. $\rho_{11}(t =$ = 0) = 1. We are interested in the weak probe field scheme, so $\Omega_p = 1.5 \cdot 10^{-11}$ arb. units and $\Omega_p =$ $= 3.0 \cdot 10^{-8}$ arb. units in the following numerical calculations. The selected parameters are $\omega_{21} = 0.1$ arb. units, $\omega_{32} = 9.1 \cdot 10^{-10}$ arb. units, $\Gamma_2 = 5.7 \cdot 10^{-9}$ arb. units, $\Gamma_3 = 9.6 \cdot 10^{-10}$ arb. units, $\mu_{21} = 3.0$ arb. units, and $\mu_{32} = 1$ arb. units. We consider the situation in which the strong coupling field is resonant with the transition frequency ω_{32} , i.e. $\delta_c = 0$. In order to studying the effects of permanent dipole moments on the OB, it is as-



Fig. 2. Output field versus input field in the absence $(d_{21} = \mu_{22} - \mu_{11} = 0 \text{ arb. units}, d_{31} = \mu_{33} - \mu_{11} = 0 \text{ arb. units}, and <math>d_{32} = \mu_{33} - \mu_{22} = 0 \text{ arb. units})$, and presence of PDM $(d_{21} = \mu_{22} - \mu_{11} = 6.5 \text{ arb. units}, d_{31} = \mu_{33} - \mu_{11} = 6.5 \text{ arb. units}, and <math>d_{32} = \mu_{33} - \mu_{22} = 6.5 \text{ arb. units}$ effects. The selected parameters are $\delta_p = \delta_c = 0$, $\Gamma_2 = 5.7 \cdot 10^{-9}$ arb. units, $\Gamma_3 = 9.6 \cdot 10^{-10}$ arb. units and $\Omega_c = 3.0 \cdot 10^{-8}$ arb. units



Fig. 3. The steady-state results for transmission (a), dispersion (b), group velocity (c) as a function of the probe detuning in the absence (solid line) and presence (dashed line) of permanent dipole moments

sumed that there exists a pseudomolecule with the same parameters except for $d_{21} = \mu_{22} - \mu_{11} = 0$ arb. units, $d_{31} = \mu_{33} - \mu_{11} = 0$ arb. units, and $d_{32} = \mu_{33} - \mu_{22} =$ = 0 arb. units. As shown in Fig. 2, it can be seen that in the absence of PDM effects, the OB can be realized in the present molecule system (solid line). However in the presence of PDM effects, i.e. for $d_{21} = \mu_{22} - \mu_{11} =$ = 6.5 arb. units, $d_{31} = \mu_{33} - \mu_{11} = 6.5$ arb. units, and $d_{32} = \mu_{33} - \mu_{22} = 6.5 \text{ arb. units}$, the OB converts to OM. The behavior of transmission (a), dispersion (b) and group index (c) of weak probe light versus probe detuning in the absence and presence of PDM effects is shown in Fig.3. In the absence of PDM effects (solid line), the transmission, dispersion and group index properties of this dipole molecule are analogous to atomic system. At zero detuning of probe light, the transmission reaches near to zero and the slope of dispersion is negative. In this case, the group velocity of probe light has a negative value and corresponds to superluminal light propagation which has a good agreement with dispersion curve. However, in the presence of PDM effect (dashed line), the dips of transmission profile at detunings $\Omega_c/2$ and $-\Omega_c/2$, alters. In this case, the slope of dispersion changes to positive. Therefore,



Fig. 4. Output field versus input field in the presence of PDM ($d_{21} = \mu_{22} - \mu_{11} = 6.5$ arb. units, $d_{31} = \mu_{33} - \mu_{11} = 6.5$ arb. units, and $d_{32} = \mu_{33} - \mu_{22} = 6.5$ arb. units) effects. Solid line corresponds to $\mu_{32} = 1$ arb. units, dashed line corresponds to $\mu_{32} = 0.5$ arb. units, and dotted line corresponds to $\mu_{32} = 0.1$ arb. units. The other parameters are same as Fig. 3



Fig. 5. The steady-state results for transmission (a), dispersion (b), group velocity (c) as a function of the probe detuning in the presence of permanent dipole moments. The $\mu_{32} = 0.1$ arb. units, and others parameters are same as Fig. 4

group velocity of weak probe light has a positive value which corresponds to subluminal light propagation. Due to presence of permanent dipole moments, new coherent effects can be induced in the dipole molecule. It should be pointed out that the physical basis of this phenomenon is that the presence of permanent dipole moments modifies the light-molecule interaction where light is the change (renormalization) of the applied Rabi frequency [45]. When the coherent effects which are induced by PDMs are enhanced, the absorption and dispersion and consequently group velocity properties of this molecule can be completely different from those of an atom. The behavior of new OB and OM spectra are shown in Fig. 4. We use the same parameters as Fig. 3 (solid line), except for $\mu_{32} = 0.5$ arb. units (dashed line) and $\mu_{32} = 0.1$ arb. units (dotted line) [52]. It is shown that the OM converts to OB by the new coherent effects. That is to say, the multistable to bistable can be controlled very effectively by modifying this new

coherent effect. The main reason for the above phenomena is that the new coherent effect will modify the absorption and the Kerr nonlinearity of the molecule medium which makes the multistable behavior disappears. The behaviors of transmission (a), dispersion (n) and group index (c) of weak probe light versus probe detuning in the presence of PDM and new coherence effect is also displayed in Fig. 5. It can be seen that the value of transmission coefficient increases and its maximum reaches to 1.002 which shows the probe absorption converts to probe gain. It can be seen that two small gain profiles are present in the transmission spectrum and simultaneously enhancement of the index of refraction occurs. Therefore, the group velocity can be decreased at the gain point. These results have a good agreement with together. Here, gain without population inversion can be achieved without any incoherent pump field. Physically, permanent dipole moments induced an extra coherence which can be substitute the incoherent pump field, thus GWI can occur in this scheme.

Conclusion. In summary, we have studied the effects of quantum coherence induced by permanent dipole moments on optical bistability and multistability in a three-level dipolar molecule. We found that OB and OM behavior can be controlled by new coherent effects. We also found that enhancement of refractive index with probe amplification can be established. Moreover, we displayed the group index switching from superluminal to subluminal or vice versa can be achieved by new quantum coherence effects.

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