Optical bistability and multistability via quantum coherence in chiral molecules

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Abstract: The optical bistability (OB) and multistability (OM) in chiral molecules are investigated by placing the sample into a unidirectional ring cavity. Because of broken mirror symmetry of the effective potential, the chiral molecules have a cyclic three-level \( \Delta \)-configuration structure, in which one- and two-photon transitions can coexist. We find that the OB is achievable in this system on exact one-, two- and three-photon resonance conditions but absent in the three-level \( \Lambda \)-type system under the two-photon resonance. Moreover, the OM and the switching between OB and OM are also realized by choosing parameters properly. Interestingly, the left- and right-handed chiral molecules exhibit different bistable and multistable behaviors. It is shown that the threshold intensity of OB is strongly dependent on the percentage of the two enantiomers in the mixture. This provides an effective approach to probe molecular chirality and to determine enantiomer excess, which may find potential application in organic chemistry, pharmacology, biochemistry, etc..

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1. Introduction

Quantum interference and atomic coherence effects play a crucial role in the most important quantum optics phenomena such as coherent population trapping (CPT) [1], electromagnetically induced transparency (EIT) [2–7], optical bistability (OB) and multistability (OM), and so on. In past years, there has been continuous interest in OB and OM because of their important applications in optical transistors, memory elements and all optical switches. A large number of schemes including natural atomic systems, quantum well structures and quantum dot molecules were proposed to realize OB and OM both theoretically and experimentally [8–30].

The earlier experimental observations of OB were reported in a two-level alkali atomic system [8–10]. In comparison with two-level system, the OB and OM showed different behaviors in three level systems since the atomic coherence effects can greatly modify the absorption, dispersion and nonlinearity of the systems [11, 12]. Harshawardhan and Agarwal proposed a scheme to demonstrate OB using field-induced atomic coherence and quantum interference effects [13], wherein the threshold intensity was rather small. Gong et al. showed that the OB was strongly influenced by the initial atomic coherence in a ground-state doublet three-level system [14]. In addition, by applying trichromatic fields to drive atomic system, the OM as well as the switching between OB and OM can be effectively controlled through modifying the relative intensities and phases of the fields [15]. The experimental realization of OB and OM in the three-level system has been studied extensively as well [16–18]. On the other hand, several groups have taken into account the spontaneously generated coherence or vacuum induced coherence effects on OB and OM behaviors in the three-level V-type, A-type and ladder-type system [19–22]. It is found that the linear and nonlinear response of the probe field are heavily dependent on the quantum interference, resulting in the occurrence of bistable and multistable behaviors. However, we note that the quantum interference arises from the coupling of two transitions with parallel moments to the same vacuum mode [23]. The parallel dipole moments are difficult to find in realistic system because it requires the same quantum numbers J and mJ.

To overcome this difficult, one usually employs a DC field or a microwave field to couple to the near-degenerate states. In this way, the quantum interference effects are simulated equivalently in the dressed-state representation. Henceforth, the closed three-level atomic structure could be formed by using a microwave field to couple with the near-degenerate states in A-type
or V-type system. The OB can be effectively controlled by modifying the amplitude and the relative phase of the microwave fields in the three-level closed systems [24, 25]. In addition to the above mentioned schemes, the multilevel atomic systems, the quantum well structures and the quantum dot molecules have also been investigated widely to obtain OB and OM based on coherence effects [26–30]. Not only that, the Kerr nonlinearity, which is closely related to the OB/OM behaviors, has also been paid special attention in past and recent years [31–34]. Physically, the OB and OM are very sensitive to the coherence-controlled absorption-dispersion relation and the nonlinear interactions of lasers with different types of systems. Taking the three-level Λ-type system as an example, if the system is tuned to be away from two-photon resonance slightly, the OB or OM are obtainable because of the absorption and large nonlinearity experienced by the probe [17, 18]. Under the exact two-photon resonance, however, the system would evolve into dark-state and the linear absorption and the higher order nonlinear effects would vanish, leading to the absence of OB and OM.

Recently, an interesting cyclic structure in chiral molecules [35, 36] has been paid special attention, in which the coherence-related effects such as CPT and EIT were reported [37–39]. Because of broken mirror symmetry of the effective potential, the chiral molecules have a three-level Δ-configuration structure with three simultaneous transitions between the lowest three eigenstates states, thereby rendering the one- and two-photon processes to coexist [35, 36]. This feature makes the optical properties of the cyclic structure being different from other three-level systems. In general, the chiral molecules are divided into left- and right-handed chiral molecules (called “enantiomers”), which are coexistent because of the fundamental broken symmetries in nature [40, 41]. It is worthwhile to note that only one enantiomeric form is useful in biology, while the other one could be harmful or fatal. Consequently, how to distinguish the left- and right-hand chiral molecules is of great interest in organic chemistry, pharmacology, biochemistry, etc.. Král et al. [35] proposed a method to discriminate the left- and right-handed chiral molecules through a “cyclic population transfer” process. Li et al. [42] and Jia et al. [43] showed respectively that left- and right-handed chiral molecules can evolve into different final states by a purely dynamic transfer process or using two-step coherent pulses regardless of decay. Furthermore, several optical methods were suggested to probe effectively molecule chirality based on optical nonlinear processes [44–46]. Subsequently, a different approach to probe the chirality is provided by measuring their linear absorption spectra [47]. These results display that the absorption, dispersion and nonlinear effects in Δ-type molecule system have novel features. This motivates us to study the OB and OM behaviors in this special system. Our numerical results show that the OB and OM, as expected, are attained via quantum coherence effect in the cyclic structure under proper conditions. More interestingly, the OB and OM behaviors of the left- and right-handed molecules are different from each other. For a sample consisting of the two enantiomers, the threshold intensity varies monotonically as the ratio of the mixture changes. In this regard, via studying the bistable behaviors of the mixture, we provide a novel approach to identify molecular chirality and to determine enantiomer excess. The remainder of this paper is organized as follows. In section 2, the model is described and the master equations of the chiral molecules are derived. In section 3, the numerical results of OB and OM are given and the corresponding mechanisms are analyzed briefly. Section 4 ends with conclusion.

2. Model and equations

It has been demonstrated that a pair of chiral molecules can be treated as a system with mirror-symmetric double potential [35, 36]. When the mirror symmetry of the effective potential is broken, the three transitions between the lowest three localized chiral eigenstates are permitted simultaneously, leading to a cyclic transition structure [35]. As depicted in Fig.
Fig. 1. The energy level structure of the Δ-type chiral molecules: (a) left-handed chiral molecules; (b) right-handed chiral molecules. The enantiomers are driven by three optical fields, wherein the transition $|2\rangle_L\langle 2|_R \rightarrow |1\rangle_L\langle 1|_R$ is driven by a strong control field while the other two transitions are coupled by two probe fields.

1, the Δ-configuration chiral molecules have three levels $|j\rangle$ with transition frequencies $\omega_{jk}$ and decay rates $\gamma_{jk}$ between levels $|j\rangle$ and $|k\rangle$, where $\omega_j$ are eigenfrequencies of the levels and $\omega_{jk} = \omega_j - \omega_k$ for $j, k = 1, 2, 3, (j > k)$. Here $|j\rangle_L$ and $|j\rangle_R$ represent the left- and right-handed chiral molecules, respectively. Three coherent driven fields are applied to three transitions $|3\rangle \rightarrow |1\rangle$, $|3\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |1\rangle$. In electronic-dipole and rotating wave approximation, the Hamiltonian of the system is given as $\hbar=1$ \cite{47}

$$
H = \sum_{j=1}^{3} \omega_j \sigma_{jj} - \frac{1}{2} \sum_{j,k=1,j>k}^{3} \Omega_{jk} e^{-i(\nu_{jk} t + \phi_{jk})} \sigma_{jk} + H.c.,
$$

(1)

where $\Omega_{jk} = \mu_{jk} E_{jk}$ are assumed to be real Rabi frequencies of the applied coherent fields with the frequencies $\nu_{jk}$ and initial phases $\phi_{jk}$. $\mu_{jk}$ represent the electric-dipole moments. $\sigma_{jj} = |j\rangle \langle j|$ and $\sigma_{jk} = |j\rangle \langle k|$, $j \neq k$ are the projection operators and spin-flip operators. In the appropriate rotating frame, we rewrite the system Hamiltonian as

$$
H_I = -(\Delta_{21} \sigma_{22} + \Delta_{31} \sigma_{33}) - \frac{1}{2} \sum_{j,k=1,j>k}^{3} (\Omega_{32} \sigma_{32} + \Omega_{31} \sigma_{31} + \Omega_{21} \sigma_{21}) e^{-i(\delta t + \Phi)} + H.c.,
$$

(2)

where $\Delta_{jk} = \nu_{jk} - \omega_{jk}$, $\delta = \Delta_{32} + \Delta_{21} - \Delta_{31}$, $\Phi = \phi_{32} + \phi_{21} - \phi_{31}$. We consider the three-photon resonance case, i.e., $\delta = 0$ and assume that the strong field $\Omega_{21}$ is a control field and the two weak fields $\Omega_{31}, \Omega_{32}$ coupling with the two transitions $|3\rangle \rightarrow |1, 2\rangle$ are probe fields. The master equation describing the dynamical evolution of the system can be written as

$$
\dot{\rho} = -i[H_I, \rho] + \mathcal{L}[\rho + \sum_{j,k=1,j>k}^{3} \sqrt{\gamma_{jk}} \sigma_{jk} |\rho|, \rho],
$$

(3)

while the mirrors M3 and M4 are perfect reflectors (cavity. The symbol approximation, the dynamic response of the probe fields is described by Maxwell’s equation, resulting in a \( \Omega - \Omega \) while that of the right-handed molecules are \( \Gamma \) right-handed molecules are replaced by \( L \) where \( \Delta \) difference of total phase factor [35, 42, 47]. As a result, the matrix elements \( |3\rangle \rightarrow |1\rangle \) and \( |3\rangle \rightarrow |2\rangle \), respectively, and given \( E_{c} \) is the amplitude of the control field.

\[
\rho_{33} = - (\gamma_{31} + \gamma_{32}) \rho_{33} + i \frac{1}{2} \Omega_{31} (\rho_{13} - \rho_{31}) + i \frac{1}{2} \Omega_{32} (\rho_{23} - \rho_{32}) ,
\]
\[
\rho_{22} = - \gamma_{21} \rho_{22} + \gamma_{23} \rho_{33} + i \frac{1}{2} \Omega_{21} (\rho_{12} e^{i\phi} - \rho_{21} e^{i\phi}) + i \frac{1}{2} \Omega_{32} (\rho_{32} - \rho_{23}) ,
\]
\[
\rho_{12} = - \Gamma_{12} \rho_{12} + i \frac{1}{2} \Omega_{21} e^{i\phi} (\rho_{22} - \rho_{11}) + i \frac{1}{2} \Omega_{31} \rho_{32} - i \frac{1}{2} \Omega_{32} \rho_{13} ,
\]
\[
\rho_{13} = - \Gamma_{13} \rho_{13} + i \frac{1}{2} \Omega_{31} (\rho_{33} - \rho_{11}) + i \frac{1}{2} \Omega_{21} e^{i\phi} \rho_{23} - i \frac{1}{2} \Omega_{32} \rho_{12} ,
\]
\[
\rho_{23} = - \Gamma_{23} \rho_{23} + i \frac{1}{2} \Omega_{21} e^{i\phi} \rho_{13} + i \frac{1}{2} \Omega_{32} (\rho_{33} - \rho_{22}) - i \frac{1}{2} \Omega_{31} \rho_{21} ,
\]

Fig. 2. Unidirectional ring cavity with a chiral molecule sample of length \( L \). \( E_{p}^{i} \) and \( E_{p}^{T} \) are the incident and transmitted fields, respectively. \( E_{c} \) is the amplitude of the control field.

where \( \mathcal{L} |e\rangle \bullet = e \bullet e^{\dagger} - \{ e^{\dagger} \bullet, \} / 2 \). According to the density-operator master equation and the total Hamiltonian \( H \), we have the equations of motion of the density matrix elements

\[
\rho_{jk} = \rho_{kj}^{*}, (j, k = 1, 2, 3, j \neq k). \]

The closure of the system is \( \rho_{11} + \rho_{22} + \rho_{33} = 1 \). The damping rates of the off-diagonal elements are \( \Gamma_{12} = 1/2 \gamma_{21} + i \Delta_{21}, \Gamma_{13} = 1/2 (\gamma_{21} + \gamma_{31}) + i \Delta_{31}, \Gamma_{23} = 1/2 (\gamma_{21} + \gamma_{31} + \gamma_{32}) + i \Delta_{32} \). Particularly, the Rabi frequencies of the left-handed molecules are labeled as “\( \Omega \)” while that of the right-handed molecules are ““\( \Omega \)””, resulting in a \( \pi \)-difference of total phase factor [35, 42, 47]. As a result, the matrix elements \( \rho_{jk} \) of the left- and right-handed molecules are replaced by \( \rho_{jk}^{(+)} \) and \( \rho_{jk}^{(-)} \) with the relation of \( \rho_{jk}^{(-)} = \rho_{jk}^{(+)} (\Phi + \pi) \).

In order to study the bistable behavior of the chiral molecules, we place the sample consisting of a mixture of left- and right-handed molecules into a unidirectional ring cavity as shown in Fig. 2, in which the reflectivity(transmissivity) of mirrors \( M_{1} \) and \( M_{2} \) are \( R \) and \( T \) \((R + T = 1)\), while the mirrors \( M_{3} \) and \( M_{4} \) are perfect reflectors \((R = 1)\). Adopting a standard method, the total electromagnetic field can be written as

\[
E = E_{31} e^{-i\nu_{31} t} + E_{32} e^{-i\nu_{32} t} + E_{21} e^{-i\nu_{21} t} + c.c.,
\]

wherein \( E_{31} = E_{32} = E_{p} \) are the amplitude of the probe fields, which circulate in the ring cavity and \( E_{21} \) is the amplitude of the control field denoted by \( E_{c} \) that does not circulate in the ring cavity. The symbol c.c. means the complex conjugation. Applying a slowly varying envelope approximation, the dynamic response of the probe fields is described by Maxwell’s equation

\[
\frac{\partial E_{p}}{\partial t} + c \frac{\partial E_{p}^{*}}{\partial z} = \frac{i}{2 \epsilon_{0}} \left[ |v_{31} P(v_{31}) + v_{32} P(v_{32})| \right],
\]

where \( c \) and \( \epsilon_{0} \) are the speed of light and permittivity of free space, respectively, \( P(v_{jk}) \) are the slowly varying polarization in the transitions \(|3\rangle \rightarrow |1\rangle \) and \(|3\rangle \rightarrow |2\rangle \), respectively, and given
by $P(v_{jk}) = N\mu_{jk}(\eta^{(+)}\rho^{(+)}_{jk} + \eta^{(-)}\rho^{(-)}_{jk})$, where $N$ is the total molecular density, $\eta^{(\pm)}$ are the percentage of the left- and right-handed molecules in the mixture and $\eta^{(+)} + \eta^{(-)} = 1$.

We consider the field in equation (6) in the steady case, i.e., $\partial E_p/\partial t = 0$, the Maxwell equation is reduced to

$$\frac{\partial E_p}{\partial z} = iN \sqrt{\varepsilon_0} [\nu_31\mu_31\dot{\rho}_{31} + \nu_32\mu_32\dot{\rho}_{32}], \quad (7)$$

with $\dot{\rho}_{31} = \eta^{(+)}\rho^{(+)}_{31} + \eta^{(-)}\rho^{(-)}_{31}$, $\dot{\rho}_{32} = \eta^{(+)}\rho^{(+)}_{32} + \eta^{(-)}\rho^{(-)}_{32}$. For a perfectly tuned cavity, in the steady state limit, the boundary conditions between the incident field $E_p^i$ and the transmitted field $E_p^T$ are given as [48]

$$E_p(L) = \frac{E_p^T}{\sqrt{T}},$$
$$E_p(0) = \sqrt{T}E_p^i + RE_p(L), \quad (8)$$

where $L$ is the length of the molecule sample. The $R$ term, which describes the feedback mechanism, is essential to the occurrence of bistability. Once $R = 0$, the bistability will not exist anymore. Using the boundary conditions (8) – (9), normalizing the fields by letting $Y = \frac{(\mu_{31} + \mu_{32})E_p^T}{\sqrt{T}}$ and $X = \frac{(\mu_{31} + \mu_{32})E_p^i}{2\sqrt{T}}$, we can get an input-output relationship

$$Y = 2X - i\gamma(\dot{\rho}_{31} + \dot{\rho}_{32}). \quad (10)$$

The cooperative parameters are defined as $C_1 = \frac{NL\nu_31\mu_{31}}{2\varepsilon_0\gamma T}$ and $C_2 = \frac{NL\nu_32\mu_{32}}{2\varepsilon_0\gamma T}$ with $\gamma_31 = \gamma_32 = \gamma$. It should be pointed out that the coherence terms $\dot{\rho}_{31}$ and $\dot{\rho}_{32}$ in equation (7) are vital for OB and OM to occur. By setting all time derivatives of Eq. (4) to zero, we can obtain the steady values $\phi_{31}$ and $\phi_{32}$. Arising from Eq. (4) and Eq. (10), we can study the bistable and multistable behaviors in this system.

3. Numerical results and discussions

In the following numerical calculation, for simplicity, we assume that $\gamma_31 = \gamma_32 = \gamma$ and $C_1 = C_2 = C$, $\Omega_{21} = \Omega$, $\Omega_{31} = \Omega_{32} = \Omega_p$ and the relative phase $\Phi = 0$. The Rabi frequencies, the detunings and the decaying rates are always scaled in units of $\gamma$ throughout the paper. Our numerical results demonstrate that the OB and OM are possible to realize in the chiral molecules under different conditions. The range of the hysteresis cycle, the threshold value and the switching between OB and OM curves are controllable by adjusting effectively the detunings, the amplitude of the control field, and the percentages of the two enantiomers appropriately.

We first consider the bistable behaviors of left- and right-handed molecules separately. In Fig. 3, the input-output curves for (a) left-handed molecules ($\eta^{(+)} = 1$, $\eta^{(-)} = 0$) and (b) right-handed molecules ($\eta^{(+)} = 0$, $\eta^{(-)} = 1$) are plotted under the two-photon resonance conditions, i.e., $\Delta_{31} = \Delta_{32} = \Delta$. The probe absorption $\text{Im}(\dot{\rho}_{31} + \dot{\rho}_{32})$ are numerically plotted in Fig. 3 (c). The parameters are chosen as $\gamma_0 = \gamma$, $\Omega_c = 4\gamma$, $C = 100$, $\Delta_{21} = 0$. From these figures, it can be seen that the OB is obtained in the two enantiomers when the two-photon resonance conditions are satisfied. In particular, under the exact one- , two- and three-photon resonance conditions, i.e., $\Delta = 0$, the bistable curves of the left- and right-handed molecules are the same since the absorption is equal to each other at this time. As the detunings change from $0$ to $2\gamma$, the threshold of the left-handed molecules (Fig. 3 (a)) increases while that of the right-handed molecules (Fig. 3 (b)) decreases. This can be understood from Fig. 3 (c), wherein the absorption of the left-handed molecules first rises to a maximal value and then falls in the region of $\Delta > 0$. For the right-handed chiral molecules, the absorption curve is symmetrical about $\Delta = 0$ with
Δ by taking the Ω value for the two enantiomers would be reversed. As a matter of fact, when the control field Φ molecules while Γ interaction Imρ are obtained from Eq. (11) as follows.

\( \rho_{31}^{(1)} = \frac{2i\Omega_p \Gamma_3 \rho_{11}^s}{4\gamma_3 \Gamma_3 + \Omega_c^2}, \)
\( \rho_{32}^{(1)} = \frac{2i\Omega_p \Gamma_3 \rho_{22}^s + \Omega_p \Omega_c \rho_{12}^s}{4\gamma_3 \Gamma_3 + \Omega_c^2}, \)

where Γ₁ = Γ₁⁺, Γ₂ = Γ₂⁺. In terms of the above expression, we can find that the probe absorption Im(ρ₁ + ρ₂) is obviously phase dependent. At Φ = 0, it corresponds to the left-handed molecules while Φ = π corresponds to the right-handed molecules owing to the internal π-difference of phase for the two types of molecules [35, 42, 47]. Notably, the analytical result is approximately consistent with the numerical results plotted in Fig. 3 (c) and Fig. 5 (c, d). However, it is not applicable to the results shown in Fig. 4 because the perturbation theory is not valid at this time. Specially, the probe absorption under the exact resonance and non-exact resonance can be obtained from Eq. (11) as follows.

(i) For the exact resonant cases, i.e., \( \Delta_21 = \Delta_32 = \Delta_31 = 0 \), we have

\[
\text{Im}(\rho_{31}^{(1)} + \rho_{32}^{(1)}) = \frac{\Omega_p (\gamma_31 + \gamma_32 + \gamma_21) \rho_{12}^s + (\gamma_31 + \gamma_32) \rho_{22}^s - \Omega_c (\rho_{32}^s - \rho_{11}^s) \sin \Phi)}{(\gamma_31 + \gamma_32 + \gamma_21)(\gamma_31 + \gamma_21) + \Omega_c^2}. 
\]

(ii) For the non-exact resonant cases, i.e., \( \Delta_21 = 0, \Delta_32 = \Delta_31 = \Delta \), we have

\[
\text{Im}(\rho_{31}^{(1)} + \rho_{32}^{(1)}) = \frac{\Omega_p A D + B C}{A^2 + B^2}. 
\]
with

\begin{align}
A &= (\gamma_{31} + \gamma_{32} + \gamma_{21})(\gamma_{31} + \gamma_{21}) + \Omega_c^2 - 4\Delta^2, \\
B &= 2\Delta(2\gamma_{31} + \gamma_{32} + 2\gamma_{21}), \\
C &= 2\Delta + \Omega_c \cos \Phi, \\
D &= (\gamma_{31} + \gamma_{32} + \gamma_{21})^2 + (\gamma_{31} + \gamma_{21})\rho_{22}^s - \Omega_c(\rho_{22}^s - \rho_{11}^s) \sin \Phi.
\end{align}

From Eq. (12), it can be seen clearly that the probe absorption is identical for \( \Phi = 0 \) and \( \Phi = \pi \), resulting in the same OB behaviors for left- and right-handed molecules. However, for the cases of \( \Delta \neq 0 \), the probe absorption for \( \Phi = 0 \) and \( \Phi = \pi \) is distinct from each other, which can be straightforwardly deduced from Eq. (13) and Eq. (14).

The physical interpretation of the above results can be attributed to dark-state resonance effect. As is well known, in the three-level \( \Lambda \)-type atomic system, the light fields, if they satisfy two-photon resonance conditions, would drive the system into a dark state with vanishing absorption, leading to the absence of OB. In the present system, nevertheless, since the transition \( |2\rangle \rightarrow |1\rangle \) is permitted, the system would deviate away from dark state slightly although the two-photon resonance conditions are satisfied. The probe absorption is no longer equal to zero and then the bistable behaviors emerge. To verify that, the damping effects \( \gamma_{21} \) on the probe absorption under one- and two-photon resonance conditions are plotted in Fig. 4 by taking \( \Omega_c = 0 \).

At \( \gamma_{21} = 0 \), corresponding to the three-level \( \Lambda \)-type system, the absorption is zero and the OB would be absence at this time. Once the transition channel between the two ground states exists, i.e., \( \gamma_{21} \neq 0 \), the OB occurs and is enhanced as \( \gamma_{21} \) increases. In fact, the cyclic structure is equivalent approximately to a combination of three-level \( \Lambda \)-type and Ladder-type system under certain conditions [50], which can be attributed to the appearance of OB under exact resonance conditions.

In Fig. 5, the input-output curves for (a) left- and (b) right-handed chiral molecules are plotted by changing the intensity of the control field. The other parameters are the same as those in Fig. 3 except for \( \Delta = 2\gamma \). The probe absorption is also plotted in Fig. 5 (c, d). With increasing \( \Omega_c \) from \( \gamma \) (solid line) to \( 5\gamma \) (dashed line), \( 10\gamma \) (dotted line), the threshold intensity and the range of the hysteresis cycle in Fig. 5 (a) first increase and then decrease but in Fig. 5 (b) they decrease always. From Fig. 5 (c, d), it is easy to see that the absorption of the left-handed chiral molecules at \( \Delta = 2\gamma \) is first enhanced and then reduced. Differently, shown in Fig. 5 (d), the absorption of the right-handed molecules at \( \Delta = 2\gamma \) decreases remarkably, resulting in the reduction of threshold intensity and the scope of hysteresis cycle.

Next, we show the OM curves and the switching between OB and OM under different conditions. In Fig. 6, we plot the input-output curves for (a) left- and (b) right-handed chiral molecules by choosing the probe detunings as \( \Delta_{31} = -\Delta_{31} = 0 \) (solid line), \( 0.2\gamma \) (dashed line),...
Fig. 5. (a,b) Input-output field curves by choosing different intensities of control field for left- and right-handed chiral molecules, respectively. (c, d) Probe absorption $\text{Im}(\rho_{31} + \rho_{32})$ of the two enantiomers as a function of the detuning $\Delta$ by changing the strength of the control field. The other parameters are chosen as $C = 100$, $\gamma_2 = \gamma$, $\Delta_2 = 0$.

Fig. 6. Input-output field curves by choosing different detunings $\Delta_3 = -\Delta_3 = 0$ (solid line), $0.2 \gamma$ (dashed line), $0.4 \gamma$ (dotted line) for (a) left-handed chiral molecules; (b) right-handed chiral molecules. The other parameters are chosen as $\gamma_2 = 0.1 \gamma$, $\Omega_c = 4 \gamma$, $C = 100$, $\Delta_2 = \Delta_3 - \Delta_3$. It is found that the OM behaviors arise in the two enantiomers but the variation trends are different from each other. In Fig. 6 (a), when the asymmetrical detunings are changed from 0 to $0.2 \gamma$, the OB switches to OM. By further increasing the detunings to $0.4 \gamma$, the threshold of the first hysteresis circle falls while the second one rises as the detunings increase. In comparison with left-handed chiral molecules, both the threshold intensities of the right-handed molecules shown in Fig. 6 (b) are enhanced during this process. In addition, as shown in Fig. 7, the input-output curves are plotted by changing the strength of the control field. We choose the different Rabi frequencies as $\Omega_c = \gamma$ (solid line), $5 \gamma$ (dashed line), $10 \gamma$ (dotted line). The other parameters are the same as those in Fig. 6 except for $\Delta_3 = -\Delta_3 = 0.2 \gamma$. Evidently, in spite of the variation trends of the curves for the two enantiomers are similar to each other in this case, the threshold values of OM in the left-handed chiral molecules (Fig. 7 (a)) are smaller than that in the right-handed ones (Fig. 7 (b)).

In brief, we can draw a conclusion from the above discussion that the bistable and multi-stable behaviors of the left- and right-handed chiral molecules are different from each other. Nevertheless, the two enantiomers are usually in coexistence because of the fundamental broken symmetries in nature [40, 41]. Henceforth, it is of realistic interest to study the sample
Fig. 7. Input-output field curves by choosing different intensities of control field: \( \Omega_c = \gamma \) (solid line), \( 5\gamma \) (dashed line), \( 10\gamma \) (dotted line) for (a) left- and (b) right-handed chiral molecules, respectively. The other parameters are the same as those in Fig. 6 except for \( \Delta_{32} = -\Delta_{31} = 0.2\gamma \).

Fig. 8. Input-output field curves by varying the ratios of the two enantiomers. The parameters are chosen as \( C = 100, \Omega_c = 4\gamma, \Delta_{31} = \Delta_{32} = 2\gamma, \Delta_{21} = 0, \gamma_{21} = \gamma \).

consisting of a mixture of left- and right-handed (\( \Delta \)-type) molecules. In Fig. (8), the bistable curves are plotted by taking different ratio of the two enantiomers. The parameters are the same as those in Fig. 5 except for \( \Omega_c = 4\gamma \). It can be seen that the threshold intensity of the OB becomes smaller as the ratio of left-handed chiral molecules \( \eta(+) \) falls. To make it more clearly, we plot the evolution of threshold intensity as a function of \( \eta^{(+)} \) for two cases (a) \( \Delta > 0 \) and (b) \( \Delta < 0 \) in Fig. 9. Remarkably, the threshold is monotonically enhanced in the region of \( \Delta > 0 \) but decreases in the region of \( \Delta < 0 \). The threshold intensity and the percentage satisfy one-to-one relation, which can be utilized to identify molecular chirality and to determine enantiomer excess.

Before ending this section, we would like to emphasize the main differences between the present scheme and the previous works. Firstly, the three dipole transitions between the lowest three eigenstates of the chiral molecules are permitted simultaneously \[35, 36, 42\]. This leads to the fact that the one- and two-photon processes are coexistent. The bistable behavior can be obtained under exact one-, two- and three-photon resonance conditions but it would be absent in the three-level \( \Lambda \)-type system under two-photon resonance. The transition channel between the two ground states plays an important role in the generation of OB. Secondly, it is explored that the bistable and multistable behaviors of the left- and right-handed molecules are different from each other. The threshold intensity and scope of the hysteresis loop are strongly dependent on the percentage of the two “enantiomers” in the mixture, which provides a novel way to probe left- and right-handed chiral molecules. Ultimately, it is well known that a similar cyclic transition structure can be found in the superconducting flux quantum circuits \[49, 50\]. However, one of the differences between the two systems is that the transition frequencies for the chiral molecules are in the optical frequency region while those for the superconducting system...
are in the microwave frequency domain. Besides, in general, merely a single superconducting atom mediates into the interaction in the 1D open space. When a large number of fluxonium qubits are put together, there is the inductively coupling between those fluxonium qubits. This would be harmful for the generation of OB in experiment. What is noteworthy is that the optical response in molecule system is relatively weak. In order to solve this problem in experiment, one has proposed a method by confining the molecules into a hollow-core photonic crystal fiber (HC-PCF) to enhance the light-matter interactions [51]. To do so, the typically quantum coherence effects, such as EIT [37, 38] and four-wave mixing [52], are realized. In addition, it has been reported that the EIT related coherence effects could be achievable in laser cooled molecule ensemble [53]. These pioneering works demonstrate that the experimental investigation of OB and OM in chiral molecules are implementable.

4. Conclusion

In summary, it is shown that the optical bistable and multistable behaviors are obtained based on the quantum coherence in three-level $\Delta$-type chiral molecules. Since three transitions between the lowest three eigenstates of the chiral molecules are permitted simultaneously, the one- and two-photon processes can coexist. This gives rise to the distinctive quantum coherence effect being different from natural three-level atomic systems. By applying two probe fields and a control field to the cyclic structure, the OB is generated even under the exact one-, two-, and three-photon resonance conditions for both left- and right-handed chiral molecules. Moreover, the OM patterns and the switching between OB and OM are also realized by choosing the parameters properly. More importantly, it has been found that the two enantiomers display different OB and OM behaviors under the same conditions. For a sample consisting of the two enantiomers, the threshold intensity and scope of hysteresis loop increase or decrease as the percentages change in the mixture. This provides a different method to probe molecular chirality and to determine enantiomer excess.

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