Probing molecular chirality by coherent optical absorption spectra

W. Z. Jia[1,*] and L. F. Wei[1,2,†]

[1]*Quantum Optoelectronics Laboratory, School of Physics and Technology, Southwest Jiaotong University, Chengdu 610031, China*
[2]*State Key Laboratory of Optoelectronic Materials and Technologies, School of Physics and Engineering, Sun Yat-Sen University, Guangzhou 510275, China*

We propose an approach to sensitively probe the chirality of molecules by measuring their coherent optical-absorption spectra. It is shown that quantum dynamics of the cyclic three-level chiral molecules driven by appropriately designed external fields is total-phase dependent. This will result in chirality-dependent absorption spectra for the probe field. As a consequence, the charality-dependent information in the spectra (such as the locations and relative heights of the characteristic absorption peaks) can be utilized to identify molecular chirality and determinate enantiomer excess (i.e., the percentages of different enantiomers). The feasibility of the proposal with chiral molecules confined in hollow-core photonic crystal fiber is also discussed.

PACS number(s): 42.50.Gy, 33.15.Bh, 33.55.+b, 42.50.Hz

I. INTRODUCTION

The coexistence of left- and right-handed chiral molecules (called "enantiomers") originates from the fundamental broken symmetries in nature [1]. The physiological effects of enantiomers of biologically active compounds may differ significantly [2]. In general, only one enantiomeric form has the potential to be biologically beneficial, while the other one could be harmful or fatal. Thus, probing molecular chirality is a fundamentally important task in organic chemistry, pharmacology, biochemistry, etc.

Since Pasteur's pioneering experiments on optical activity [3], asymmetries in the interaction of polarized light with chiral molecules have provided a powerful physical probe of molecular chirality. The most common techniques providing chirality-specific spectroscopic information are, e.g., circular dichroism and Raman optical activity. [4]. Note that these techniques are based on certain higher-order interactions (such as magnetic-dipole and electric-quadrupole couplings) between the molecular systems and the probing lights. Consequently, the sensitivity of these conventional techniques is usually not sufficient to detect the chirality of a small amount of sample. Recent studies [5–8] show that purely electric-dipolar nonlinear optical processes, such as second-harmonic generation and sum-frequency generation, can act as desirable sensitive probes of chirality.

Alternatively, in this paper we propose an optical method to achieve the desirable chirality probe. Our proposal is based on manipulating quantum coherence in chiral molecules, which can be modeled as cyclic-transition (Δ-type) quantum systems [9–11]. Under strong driving, the coexistence of electromagnetic-induced transparency (EIT) effects [12] and the two-photon process in a Δ-type chiral molecule will result in chirality-dependent absorption spectra for the applied probe field. In fact, based on this kind of transition structure, some optical methods for enantioseparation (rather than chirality probes) have already been suggested [9,10,13,14]. Our protocol is based on the electric-dipole interactions between the molecules and the coherent optical fields. It may provide an alternative method to probe chirality, other than the conventional ones, which measure the optical activity [4].

II. CHIRALITY PROBE BASED ON MANIPULATING QUANTUM COHERENCE

It is well known that a pair of chiral molecules can be modeled as a system with mirror-symmetric double-well potential [10,15]; the left-handed molecules are localized in one of the potentials, and the right-handed ones are in another potential. Therefore, the parities of either single left- or right-handed molecules are broken. Due to these broken-parity symmetries, the lowest three localized chiral eigenstates of either the left- or right-handed molecules can form a Δ-type cyclic-transition structure [9,10]. A typical example is that of the $D_2 S_2$ enantiomers [10], whose three pairs of rovibrational eigenstates form a pair of loop-transition structures. All the absorption frequencies of these molecules are located in the infrared range.

We assume that $|i\rangle$ $(i = 1,2,3)$ are the selected levels of a chiral molecule with according eigenfrequenies ω_i. Specifically, $|i\rangle$ may be left-handed states $|i\rangle_L$ or right-handed states $|i\rangle_R$, as shown in Fig. 1. Three coherent driving fields,

$$E_{ij}(z,t) = \tfrac{1}{2}\mathscr{E}_{ij}(z,t)e^{-i[v_{ij}t - k_{ij}z + \phi_{ij}(z,t)]} + \text{c.c.},$$

with slowly varying amplitude $\mathscr{E}_{ij}$, wave vectors k_{ij}, frequencies v_{ij}, and phase factor ϕ_{ij} are applied to couple all the permissible transition channels $|i\rangle \leftrightarrow |j\rangle$.

The Hamiltonian describing the present three-level cyclic-transition system can be written as

$$\hat{H} = \hbar \sum_{i=1}^{3} \omega_i |i\rangle\langle i| - \frac{\hbar}{2} \sum_{i>j=1}^{3} (\Omega_{ij} e^{-iv_{ij}t + ik_{ij}z} |i\rangle\langle j| + \text{H.c.}).$$

$$(1)$$

Here, the Rabi frequencies are defined as $\Omega_{ij} = (\mu_{ij}\mathscr{E}_{ij}e^{-i\theta_{ij}})/\hbar$, with μ_{ij} being the amplitude of the dipole matrix elements and $\theta_{ij} = \phi_{ij} + \chi_{ij}$. ϕ_{ij} and χ_{ij} are the phases of the electric field components and the dipole matrix elements,
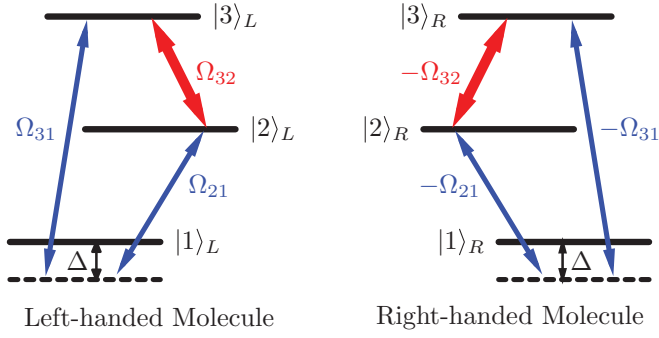
FIG. 1. (Color online) Three-level chiral molecules with cyclic transition structures. The enantiomers are driven by three optical fields. Levels $|2\rangle_L$ ($|2\rangle_R$) and $|3\rangle_L$ ($|3\rangle_R$) are resonantly coupled by a strong control field. The other two transition channels are coupled by two probes with the same detuning Δ.

respectively. The master equation describing the dynamical evolution of the system can be written as

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}[\hat{H},\hat{\rho}] + \mathcal{L}[\hat{\rho}]. \tag{2}$$

We now assume that the strong control field Ω_{32} is resonantly applied and the two weak probe fields, Ω_{31} and Ω_{21}, are applied with the same detunings, namely, $\Delta_{32} = 0$, $\Delta_{31} = \Delta_{21} = \Delta$. Then, the conditions $\nu_{31} = \nu_{21} + \nu_{32}$ and consequently $k_{31} = k_{21} + k_{32}$ are satisfied (here, three fields are assumed to be propagating in the same direction). Furthermore, we redefine the density-matrix elements as $\sigma_{ii} = \rho_{ii}$, $\sigma_{ij} = \rho_{ij}e^{i\nu_{ij}t - ik_{ij}z + i\theta_{ij}}$ ($i > j$). Finally, under the rotating wave approximation the redefined density-matrix elements obey the following equations of motion:

$$\dot{\sigma}_{11} = \Gamma_{31}\sigma_{33} + \Gamma_{21}\sigma_{22} + \frac{1}{2}(-i|\Omega_{31}|\sigma_{13} \\ - i|\Omega_{21}|\sigma_{12} + \text{H.c.}), \tag{3a}$$

$$\dot{\sigma}_{22} = -\Gamma_{21}\sigma_{22} + \Gamma_{32}\sigma_{33} + \frac{1}{2}(i|\Omega_{21}|\sigma_{12} \\ - i|\Omega_{32}|\sigma_{23} + \text{H.c.}), \tag{3b}$$

$$\dot{\sigma}_{21} = -\lambda_{21}\sigma_{21} + \frac{1}{2}[i|\Omega_{32}|\sigma_{31}e^{i\Theta} - i|\Omega_{31}|\sigma_{23}e^{i\Theta} \\ - i|\Omega_{21}|(\sigma_{22} - \sigma_{11})], \tag{3c}$$

$$\dot{\sigma}_{31} = -\lambda_{31}\sigma_{31} + \frac{1}{2}[i|\Omega_{32}|\sigma_{21}e^{-i\Theta} - i|\Omega_{21}|\sigma_{32}e^{-i\Theta} \\ - i|\Omega_{31}|(\sigma_{33} - \sigma_{11})], \tag{3d}$$

$$\dot{\sigma}_{32} = -\gamma_{23}\sigma_{32} + \frac{1}{2}[i|\Omega_{31}|\sigma_{12}e^{i\Theta} - i|\Omega_{21}|\sigma_{31}e^{i\Theta} \\ - i|\Omega_{32}|(\sigma_{33} - \sigma_{22})], \tag{3e}$$

where Γ_{ij} ($i > j$) are the relaxation rates between the levels $|i\rangle$ and $|j\rangle$; $\lambda_{21} = \lambda_{12}^* = \gamma_{12} - i\Delta$, $\lambda_{31} = \lambda_{13}^* = \gamma_{13} - i\Delta$; $\gamma_{ij} = \gamma_{ji}$ are the damping rates of the off-diagonal terms, and $\Theta = \theta_{32} + \theta_{21} - \theta_{31}$ are the relative phases. The above equations imply that the steady-state density-matrix elements $\sigma_{ij}^{(s)}$ should be phase dependent [16,17]. Additionally, a pair of Rabi frequencies for the two enantiomers could differ by a sign $\Omega_{ij}^L = -\Omega_{ij}^R$ [9]. Namely, the according phase factors satisfy $\theta_{ij}^L - \theta_{ij}^R = \pi$, and thus the total phase factors (defined by $\Theta^{L,R} = \theta_{32}^{L,R} + \theta_{21}^{L,R} - \theta_{31}^{L,R}$) satisfy the relation $\Theta^L - \Theta^R = \pi$. Clearly, if we define $\Theta^L = \Theta$, then, inevitably, $\Theta^R = \Theta + \pi$. Consequently, the steady-state density-matrix elements of the left- and right-handed molecules can be
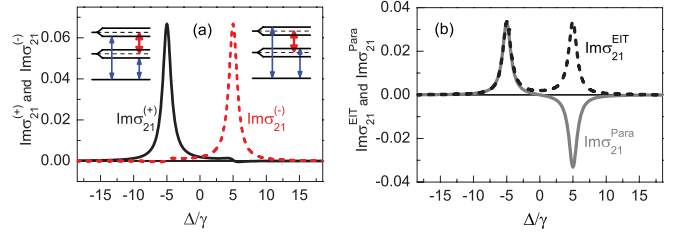

FIG. 2. (Color online) (a) Parameters $\text{Im}\sigma_{21}^{(\pm)}$ vs the dutuning Δ. Insets: schematics of the quantum interference between the single- and two-photon transition channels at probe detunings $\Delta = \pm|\Omega_{32}|/2$. (b) Parameters $\text{Im}\sigma_{21}^{\text{EIT}}$ and $\text{Im}\sigma_{21}^{\text{Para}}$ vs the dutuning Δ. (a) and (b) show that under condition (5) the chirality-dependent quantity $\text{Im}\sigma_{21}^{(+)}$ [$\text{Im}\sigma_{21}^{(-)}$] is the symmetric (antisymmetric) superposition of $\text{Im}\sigma_{21}^{\text{EIT}}$ and $\text{Im}\sigma_{21}^{\text{Para}}$. Here, we set $\Theta = 0$, $|\Omega_{21}| = |\Omega_{31}| = \gamma/10$, $|\Omega_{32}| = 10\gamma$, $\Gamma_{ij} = \gamma$, $\gamma_{12} = \Gamma_{21}/2$, $\gamma_{13} = (\Gamma_{31} + \Gamma_{32})/2$, and $\gamma_{23} = (\Gamma_{21} + \Gamma_{31} + \Gamma_{32})/2$.

presented as $\sigma_{ij}^{(+)} = \sigma_{ij}^{(s)}(\Theta)$ and $\sigma_{ij}^{(-)} = \sigma_{ij}^{(s)}(\Theta + \pi)$, respectively. This phase-sensitive optical response can be utilized to probe molecular chirality. In our protocol, either of the two weak probes can serve as the chirality probe. In the following, we choose the one with Rabi frequency Ω_{21} for specific analysis.

The analytical expressions of $\sigma_{21}^{(\pm)}$ (which represent the induced coherence between levels $|1\rangle$ and $|2\rangle$), up to the first order of Ω_{21} and Ω_{31}, can be written as

$$\sigma_{21}^{(\pm)} = \frac{i\lambda_{31}|\Omega_{21}|}{2Z} \pm \frac{-|\Omega_{31}||\Omega_{32}|e^{i\Theta}}{4Z}, \tag{4}$$

with $Z = \frac{1}{4}|\Omega_{32}|^2 + \lambda_{21}\lambda_{31}$. Above, the first term (denoted by σ_{21}^{EIT}) and the second one (denoted by $\sigma_{21}^{\text{Para}}$) are related to the (ladder-type) EIT effect and the two-photon parametric process, respectively. Obviously, due to the intrinsic π difference between the two enantiomers, the resultant phase-dependent interference terms $\sigma_{21}^{(+)}$ and $\sigma_{21}^{(-)}$ are the symmetric and antisymmetric superpositions of σ_{21}^{EIT} and $\sigma_{21}^{\text{Para}}$, respectively.

Typically, if the amplitudes and phases of Rabi frequencies are chosen as

$$|\Omega_{21}| = |\Omega_{31}|, \quad \Theta = 0, \tag{5}$$

one can check from Eq. (4) that $\text{Im}\sigma_{21}^{(-)} \simeq 0$ and $\text{Im}\sigma_{21}^{(+)}$ has a peak at the detuning $\Delta = -|\Omega_{32}|/2$, while $\text{Im}\sigma_{21}^{(+)} \simeq 0$ and $\text{Im}\sigma_{21}^{(-)}$ has a peak at the detuning $\Delta = |\Omega_{32}|/2$ [see also Fig. 2(a)]. Namely, we get chirality-dependent single-peak structures of the quantities $\text{Im}\sigma_{21}^{(\pm)}$ [which is related to the absorption of the probe Ω_{21} by left-handed (right-handed) molecules]. Therefore, by the locations of the absorption peaks one can mark the different chiral molecules.

Physically, the above results can be further explained using the dressed-state picture; see the inset in Fig. 2(a). For left-handed molecules, under strong driving the levels $|2\rangle_L$ and $|3\rangle_L$ are split into $|2,\pm\rangle_L$ and $|3,\pm\rangle_L$, respectively. The energy difference of each pair of dressed sublevels is $|\Omega_{32}|$. When the detuning of the probes is set as $\Delta = -|\Omega_{32}|/2$, constructive interference between transition channels $|1\rangle_L \leftrightarrow |2,-\rangle_L$ and $|1\rangle_L \leftrightarrow |3,-\rangle_L \leftrightarrow |2,-\rangle_L$ can generate a characteristic peak for left-handed molecules, while, when $\Delta = |\Omega_{32}|/2$, the transitions $|1\rangle_L \leftrightarrow |2,+\rangle_L$ and $|1\rangle_L \leftrightarrow |3,+\rangle_L \leftrightarrow |2,+\rangle_L$

interfere destructively and thus $\mathrm{Im}\sigma_{21}^{(+)} \simeq 0$. For the right-handed molecules case, the explanation can be given in the same way.

We now specifically discuss how the applied driving fields propagate through a medium consisting of a mixture of left- and right-handed (Δ-type) molecules. The optical waves propagating in the medium are described by the Maxwell equation [18]:

$$-\frac{\partial^2 E_{ij}}{\partial z^2} + \frac{1}{c^2}\frac{\partial^2 E_{ij}}{\partial t^2} = -\mu_0 \frac{\partial^2 P_{ij}}{\partial t^2}. \tag{6}$$

The induced oscillating polarizations P_{ij} are given by

$$P_{ij}(z,t) = \tfrac{1}{2}\mathscr{P}_{ij}e^{-i(v_{ij}t - k_{ij}z + \phi_{ij})} + \text{c.c.},$$

with slowly varying polarization:

$$\mathscr{P}_{ij} = 2N\mu_{ji}[p^{(+)}\sigma_{ij}^{(+)} + p^{(-)}\sigma_{ij}^{(-)}],$$

where N is the total molecular density and $p^{(\pm)}$ are the percentages of the two enantiomers. In the slowly varying amplitude and phase approximation, the Maxwell equation is reduced to

$$\frac{\partial\Omega_{ij}}{\partial z} = i\frac{N\mu_{ji}^2 v_{ij}}{\hbar\epsilon_0 c}[p^{(+)}\sigma_{ij}^{(+)} + p^{(-)}\sigma_{ij}^{(-)}]e^{-i\theta_{ij}}. \tag{7}$$

In addition, we can neglect the change of the strong control field Ω_{32} and approximately express $\sigma_{31}^{(\pm)}$ and $\sigma_{21}^{(\pm)}$ in the first order of Ω_{31} and Ω_{21}. Thus the probes Ω_{31} and Ω_{21} obey the following propagation equations:

$$\frac{\partial\Omega_{31}}{\partial z} = i\frac{N\mu_{13}^2 v_{31}}{Z\hbar\epsilon_0 c}\left(-\frac{1}{4}\Omega_{21}\Omega_{32}\delta p + \frac{1}{2}i\lambda_{21}\Omega_{31}\right), \tag{8a}$$

$$\frac{\partial\Omega_{21}}{\partial z} = i\frac{N\mu_{12}^2 v_{21}}{Z\hbar\epsilon_0 c}\left(-\frac{1}{4}\Omega_{31}\Omega_{32}^*\delta p + \frac{1}{2}i\lambda_{31}\Omega_{21}\right), \tag{8b}$$

with $\delta p = p^{(+)} - p^{(-)}$ being the enantiomeric difference. To achieve desirable chirality identification, the three coherent fields should satisfy condition (5) at $z = 0$. Furthermore, we define the transmission coefficient of the probe Ω_{21} as $T = |\Omega_{21}(z)|^2/|\Omega_{21}(0)|^2$ and the corresponding absorption intensity $I = 1 - T$. Additionally, if assuming $|\Omega_{32}| \gg \gamma_{12}, \gamma_{13}$, then at $\Delta = \mp|\Omega_{32}|/2$ we have $Z \simeq \pm i|\Omega_{32}|(\gamma_{12} + \gamma_{13})/2$, $\lambda_{31} \simeq \lambda_{21} \simeq \pm i|\Omega_{32}|/2$. Consequently, the heights of characteristic peaks $h^{(\pm)}$ (i.e., the absorption intensity at $\Delta = \mp|\Omega_{32}|/2$) in the absorption spectrum can be found by solving Eqs. (8):

$$h^{(\pm)} = 1 - \frac{1}{4B}e^{-C\zeta}[(1 - A \pm 2\delta p)(1 - e^{D\zeta})$$
$$+ \sqrt{B}(1 + e^{D\zeta})]^2, \tag{9}$$

with

$$\zeta = \frac{N\mu_{21}^2 v_{21}z}{\hbar\epsilon_0 c\Gamma_{21}}, \quad A = \frac{\mu_{31}^2 v_{31}}{\mu_{21}^2 v_{21}},$$

$$B = (1 - A)^2 + 4A(\delta p)^2,$$

$$C = \frac{\Gamma_{21}(1 + A + \sqrt{B})}{2(\gamma_{12} + \gamma_{13})},$$

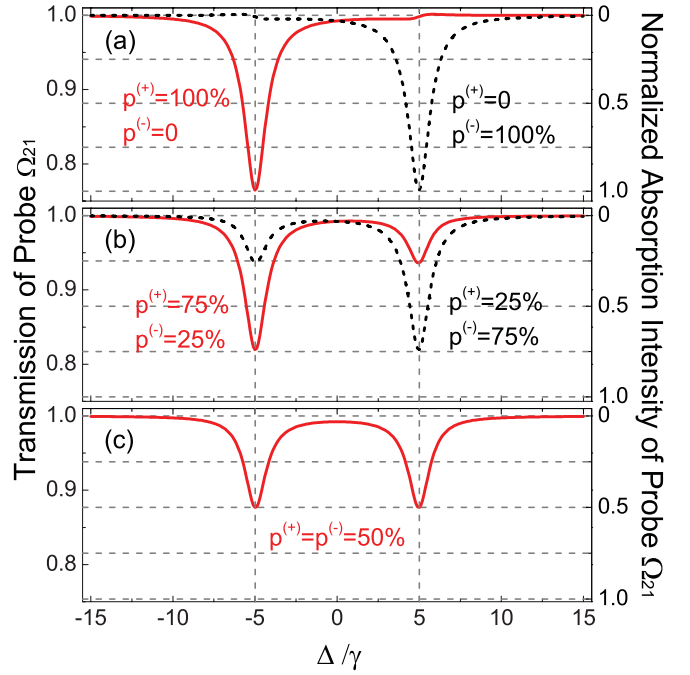$$D = \frac{\Gamma_{21}\sqrt{B}}{2(\gamma_{12} + \gamma_{13})}.$$

FIG. 3. (Color online) Transmission coefficient T and normalized absorption intensity $\tilde{I} = (1 - T)/[h^{(+)} + h^{(-)}]$ of the probe field Ω_{21} as functions of detuning, with different enantiomeric excess. The coherent driving fields enter the mediums with $\Omega_{32}(0) = 10\gamma$ and $\Omega_{21}(0) = \Omega_{31}(0) = \gamma/10$, satisfying condition (5). Optical depth is set as $\zeta = 0.2$. The other parameters are the same as those in Fig. 2.

Typically, in the optical thin region (i.e., the dimensionless optical depth $\zeta \ll 1$), the height of each characteristic peak can be approximately expressed as

$$h^{(\pm)} \simeq \frac{2\Gamma_{21}\zeta}{\gamma_{12} + \gamma_{13}}p^{(\pm)}, \tag{10}$$

which is linearly proportional to the percentage of the according chiral molecules. Therefore, if we define the normalized heights of the characteristic peaks as $\tilde{h}^{(\pm)} = h^{(\pm)}/(h^{(+)} + h^{(-)})$, then $\tilde{h}^{(\pm)} \simeq p^{(\pm)}$ in the linear range $\zeta \ll 1$.

The above analysis can be verified by numerically solving the master equations (3) and the relevant Maxwell equation (7). Figure 3 presents the probe transmission T and the normalized absorption intensity $\tilde{I} = I/[h^{(+)} + h^{(-)}]$ at optical depth $\zeta = 0.2$ versus the detuning Δ. It is seen that (1) at detuning $\Delta = \mp|\Omega_{32}|/2$, there exist two characteristic peaks corresponding to left- and right-handed molecules, respectively, and (2) the percentages of the enantiomers are directly related to the heights of the characteristic peaks. Furthermore, Table I shows also that in the linear region the enantiomeric difference δp can be read directly from the difference of the heights of two characteristic peaks $\delta p' = \tilde{h}^{(+)} - \tilde{h}^{(-)}$, namely, $\delta p \simeq \delta p'$. Table I also tells us that for stronger Ω_{32} and smaller ζ, $\delta p'$ can more accurately reflect the values of enantiomeric difference. In Fig. 4(a), we show that, for relatively weak optical depths, e.g., $\zeta = 0.2$, an approximately linear relation exists between the characteristic peak difference $\delta p'$ and the enantiomeric difference δp. When ζ increases, $\delta p'$ is never linearly proportional to δp; see Fig. 4(b). For this case we cannot read directly the percentages of the chiral molecules

TABLE I. Relation between $\delta p'$ and δp in the linear range of optical depth ζ, for certain typical parameters of ζ and $|\Omega_{32}|$. The other parameters are the same as those in Fig. 3.

δp (%)	$\zeta = 0.05$ $\delta p'$ (%)	$\zeta = 0.1$ $\delta p'$ (%)	$\zeta = 0.2$ $\delta p'$ (%)	$|\Omega_{32}|/\gamma$
0	0	0	0	
±25	±24.44	±24.21	±23.75	
±50	±48.96	±48.59	±47.85	10
±75	±73.66	±73.33	±72.66	
±100	±98.64	±98.62	±98.59	
0	0	0	0	
±25	±24.77	±24.53	±24.07	
±50	±49.62	±49.25	±48.50	100
±75	±74.66	±74.34	±73.67	
±100	±99.99	±99.99	±99.99	

from the heights of the characteristic peaks. However, note that, whether in the linear regime or not, $\delta p'$ is always a monotonic function of δp. Thus, for a given $\delta p'$ (attained by measuring the absorption peaks), the percentage of two enantiomers can always be accurately identified from the $\delta p'$-δp curve.

III. DISCUSSIONS AND CONCLUSIONS

Coherence effects, such as EIT, coherent population trapping, lasing without inversion, and so on, were first observed in atomic systems [12,19,20] and recently have been demonstrated in certain molecular systems [21–23]. Typically, to realize these effects in a molecular system should overcome the relatively weak optical response of the molecules. This can be achieved in, e.g., the hollow-core photonic crystal fiber (HC-PCF). In this system atoms and molecules can be confined in the core and thus the light-matter interactions can be enhanced significantly [24]. Indeed, HC-PCF filled with molecular gas has been demonstrated to significantly enhance various coherence effects, such as EIT [22,23] and four-wave mixing [25]. On the other hand, a laser cooled ensemble containing few atoms trapped inside the HC-PCF has been achieved to demonstrate the EIT related effects [26]. Hopefully, similar experiments to confine few cold molecules could also be implemented to realize the desirable coherence effects. Thus, if our coherence effect relevant protocol is implemented based on the cold molecular ensemble in HC-PCF, an effective method to probe chirality of a small amount of molecular sample is possible.
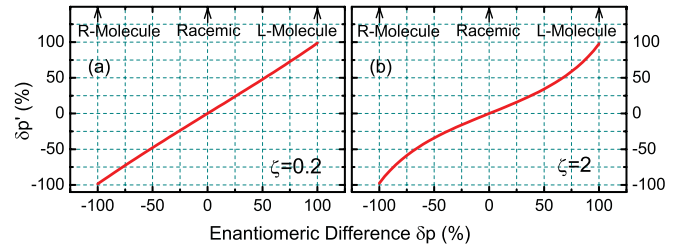

FIG. 4. (Color online) The difference of the heights of two characteristic peaks $\delta p' = \tilde{h}^{(+)} - \tilde{h}^{(-)}$ as a function of enantiomeric difference $\delta p = p^{(+)} - p^{(-)}$ at different optical depths: (a) $\zeta = 0.2$. (b) $\zeta = 2$. The other parameters are the same as those in Fig. 3.

Although our calculations and discussions are focused on the cold molecules case, our protocol can still be applied to probe the chirality of hot molecules. Certainly, including the Doppler shifts due to the thermal motion of molecules, λ_{ij} should be redefined as $\lambda_{ij} = \gamma_{ij} + i(k_{ij}v_z + \Delta_{ij})$. Here, v_z is the z component of the translational velocity of the molecules. The according medium polarization components are then obtained by integrating the density-matrix elements over the molecular distribution on velocities, i.e.,

$$\tilde{\sigma}_{ij} = \frac{1}{\sqrt{\pi u_D^2}} = \int_{-\infty}^{+\infty} \sigma_{ij}(v_z) e^{-\frac{v_z^2}{u_D^2}} dv_z, \tag{11}$$

with u_D being the most probable velocity of molecules. This implies that the Doppler shifts will lead to the broadening of spectral lines and may obscure the characteristic peaks. However, a stronger control field Ω_{32} could be applied to assure the Rabi split is larger than the Doppler broadening, and consequently similar chirality-related spectra could be observed.

In summary, we have proposed a quantum optical approach to probe molecular chirality based on phase-dependent quantum coherence and interference effects. The molecular chirality could be identified by the chirality-dependent information in the absorption spectrum. Our method may provide a new chirality probe and lead to applications in organic chemistry, biochemistry, and pharmacology.

ACKNOWLEDGMENTS

[1] R. G. Wooley, Adv. Phys. **25**, 27 (1975); *Origins of Optical Activity in Nature*, edited by D. C. Walker (Elsevier, Amsterdam, 1979).

[2] N. P. Franks and W. R. Lieb, Science **254**, 427 (1991); K. Bodenhofer, A. Hierlemann, J. Seemann, G. Gauglitz, B. Koppenhoefer, and W. Gopel, Nature (London) **387**, 577 (1997).

[3] L. Pasteur, Ann. Chim. Phys. **24**, 442 (1848).

[4] L. D. Barron, *Molecular Light Scattering and Optical Activity* (Cambridge University Press, Cambridge, 1982).

[5] T. Petralli-Mallow, T. M. Wong, J. D. Byers, H. I. Yee, and J. M. Hicks, J. Phys. Chem. **97**, 1383 (1993); J. D. Byers, H. I. Yee, T. Petralli-Mallow, and J. M. Hicks, Phys. Rev. B **49**, 14643 (1994).

[6] J. J. Maki, M. Kauranen, and A. Persoons, Phys. Rev. B **51**, 1425 (1995); M. Kauranen, T. Verbiest, and A. Persoons, J. Mod. Opt. **45**, 403 (1998).

[7] P. Fischer, A. D. Buckingham, K. Beckwitt, D. S. Wiersma, and F. W. Wise, Phys. Rev. Lett. **91**, 173901 (2003); P. Fischer and F. Hache, Chirality **17**, 421 (2005).

[8] M. A. Belkin, T. A. Kulakov, K. H. Ernst, L. Yan, and Y. R. Shen, Phys. Rev. Lett. **85**, 4474 (2000); M. A. Belkin, S. H. Han, X. Wei, and Y. R. Shen, *ibid.* **87**, 113001 (2001); M. A. Belkin and Y. R. Shen, *ibid.* **91**, 213907 (2003); N. Ji and Y. R. Shen, Chirality **18**, 146 (2006).

[9] P. Král and M. Shapiro, Phys. Rev. Lett. **87**, 183002 (2001).

[10] P. Král, I. Thanopulos, M. Shapiro, and D. Cohen, Phys. Rev. Lett. **90**, 033001 (2003); I. Thanopulos, P. Král, and M. Shapiro, J. Chem. Phys. **119**, 5105 (2003).

[11] Yu-xi Liu, J. Q. You, L. F. Wei, C. P. Sun, and F. Nori, Phys. Rev. Lett. **95**, 087001 (2005); L. F. Wei, J. R. Johansson, L. X. Cen, S. Ashhab, and Franco Nori, *ibid.* **100**, 113601 (2008).

[12] K. J. Boller, A. Imamoglu, and S. E. Harris, Phys. Rev. Lett. **66**, 2593 (1991); S. E. Harris, Phys. Today **50**, 36 (1997); M. Fleischhauer, A. Imamoglu, and J. P. Marangos, Rev. Mod. Phys. **77**, 633 (2005).

[13] Y. Li, C. Bruder, and C. P. Sun, Phys. Rev. Lett. **99**, 130403 (2007).

[14] Y. Li and C. Bruder, Phys. Rev. A **77**, 015403 (2008); W. Z. Jia and L. F. Wei, J. Phys. B **43**, 185402 (2010).

[15] F. Hund, Z. Phys. **43**, 805 (1927).

[16] H. Li, V. A. Sautenkov, Y. V. Rostovtsev, G. R. Welch, P. R. Hemmer, and M. O. Scully, Phys. Rev. A **80**, 023820 (2009).

[17] W. Z. Jia, L. F. Wei, and Z. D. Wang, Phys. Rev. A **83**, 023811 (2011).

[18] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, 1997).

[19] E. Arimondo and G. Orriols, Lett. Nuovo Cimento Soc. Ital. Fis. **17**, 333 (1976); E. Arimondo, in *Progress in Optics* XXXV, edited by E. Wolf (North-Holland, Amsterdam, 1996).

[20] S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989); A. S. Zibrov, M. D. Lukin, D. E. Nikonov, L. Hollberg, M. O. Scully, V. L. Velichansky, and H. G. Robinson, *ibid.* **75**, 1499 (1995).

[21] J. Qi, G. Lazarov, X. Wang, L. Li, L. M. Narducci, A. M. Lyyra, and F. C. Spano, Phys. Rev. Lett. **83**, 288 (1999); J. Qi, F. C. Spano, T. Kirova, A. Lazoudis, J. Magnes, L. Li, L. M. Narducci, R. W. Field, and A. M. Lyyra, *ibid.* **88**, 173003 (2002).

[22] S. Ghosh, J. E. Sharping, D. G. Ouzounov, and A. L. Gaeta, Phys. Rev. Lett. **94**, 093902 (2005).

[23] P. S. Light, F. Benabid, G. J. Pearce, F. Couny, and D. M. Bird, Appl. Phys. Lett. **94**, 141103 (2009).

[24] F. Benabid, J. C. Knight, G. Antonopoulos, and P. Russell, Science **298**, 399 (2002).

[25] S. O. Konorov, A. B. Fedotov, and A. M. Zheltikov, Opt. Lett. **28**, 1448 (2003).

[26] M. Bajcsy, S. Hofferberth, V. Balic, T. Peyronel, M. Hafezi, A. S. Zibrov, V. Vuletic, and M. D. Lukin, Phys. Rev. Lett. **102**, 203902 (2009).