Probing molecular chirality by coherent optical absorption spectra

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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, these absorption spectra can be utilized to identify molecular chirality and determine enantiomer excess. The feasibility of the proposal with chiral molecules confined in hollow-core photonic crystal fiber (HC-PCF) is also discussed.

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Introduction.—The coexistence of left- and right-handed chiral molecules (called “enantimators”) 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 fundamentally important tasks 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 has provided a powerful physical probe of molecular chirality. The most common techniques providing chirality-specific spectroscopic information are, e.g., circular dichroism (CD), and Raman optical activity (ROA), etc. $^4$. Note that all 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 chirality of a small amount of sample. While, recent studies $^5$–$^8$ show that, purely electric-dipolar nonlinear optical process, such as second-harmonic generation (SHG) and sum-frequency generation (SFG), can act as desirable sensitive probes of chirality.

Alternatively, in this letter 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 ($\Delta$-type) quantum systems $^9$–$^{11}$. Under strong driving the coexistence of electromagnetic induced transparency (EIT) effects $^{12}$ and two-photon process in a $\Delta$-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 enantioproduction (rather than chirality probes) have already been suggested $^3$, $^{10}$, $^{13}$, $^{14}$. Since our protocol is based on the electric-dipole interactions between the molecules and the coherent optical fields, it may provide a more sensitive chirality probe compared with the conventional ones by measuring the optical activity $^4$.

Chirality probe based on manipulating quantum coherence.—It is well-known that a pair of chiral molecules (e.g., the $D_2S_2$ enantiomers) can be modeled as a system with mirror-symmetric double-well potential $^{15}$. Due to the broken-parity symmetries, the lowest three localized chiral eigenstates can form a $\Delta$-type cyclic-transition structure $^2$, $^10$. We assume that $|i\rangle$ ($i = 1, 2, 3$) are the selected levels of a chiral molecule with according eigenfrequencies $\omega_i$. 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 $E_{ij}(z,t) = \frac{\hbar}{2} \delta_{ij}(z,t) e^{i(-\nu_{ij}t-k_{ij}z+\phi_{ij}(z,t))} + c.c. (i > j)$ with slowly varying amplitude $\delta_{ij}$, wave vectors $k_{ij}$, frequencies $\nu_{ij}$ and phase factor $\phi_{ij}$, 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

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

Here, the Rabi frequencies are defined as $\Omega_{ij} = (\mu_{ij}\delta_{ij} e^{-i\theta_{ij}}) / \hbar$, with $\mu_{ij}$ being the amplitude of the dipole matrix elements and $\theta_{ij} = \phi_{ij} + \chi_{ij}$. $\phi_{ij}$ and $\chi_{ij}$ are the phases of the electric field components and the dipole matrix

![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 $\Delta$.](image-url)
elements, respectively. The master equation describing the dynamical evolution of the system can be written as

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

(2)

We now assume that the strong control field $\Omega_{32}$ is resonantly applied and the two weak probe fields, $\Omega_{31}$ and $\Omega_{21}$, are applied with the same detunings, namely, $\Delta_{32} = 0$, $\Delta_{31} = \Delta_{21} = \Delta$. Then, the condition $\nu_{31} = \nu_{21} + \nu_{32}$ 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 $\sigma_{ii} = \rho_{ii}$, $\sigma_{ij} = \rho_{ij} e^{i\nu_i t - ik_j z + \theta_{ij}}$, $(i > j)$. Finally, under the rotating wave approximation the redefined density matrix elements obey the following equations of motion:

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

(3a)

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

(3b)

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

(3c)

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

(3d)

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

(3e)

where $\Gamma_{ij}$ $(i > j)$ are the relaxation rates between the levels $|i\rangle$ and $|j\rangle$; $\lambda_{21} = \lambda_{32} = \gamma_{12} - i\Delta$, $\lambda_{31} = \lambda_{13} = \gamma_{13} - i\Delta$; $\gamma_{ij} = \gamma_{ji}$ 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 $\sigma_{ij}^{(+)}$ should be phase dependent [16, 17]. Additionally, all of the Rabi frequencies for the two enantiomers could differ by a sign, resulting in a $\pi$ difference of total phase factor [9]. Consequently, the steady-state density matrix elements of the left- and right-handed molecules can be 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 be served as the chirality probe, e.g., we can choose the one with Rabi frequency $\Omega_{21}$ for specific analysis. For this purpose, we firstly give the analytical expressions of $\sigma_{21}^{(\pm)}$ (which represent the induced coherence between levels $|1\rangle$ and $|2\rangle$) in the first order of $\Omega_{21}$ and $\Omega_{31}$:

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

(4)

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

Typically, if the amplitudes and phases of Rabi frequencies 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). Namely, we get chirality-dependent single-peak structures of the absorption-related quantities $\text{Im} \sigma_{21}^{(+)}$. Therefore, by the locations of the absorption peaks one can mark the left- and right-handed molecules.

Physically, the above results can be further explained using the dressed-state picture, see the inset in Fig. 2. For left-handed molecules, under the 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 differences of each pair of dressed sublevels is $|\Omega_{32}|$. When the probe detuning 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 $\text{Im} \sigma_{21}^{(+)} \simeq 0$. For the right-handed molecules case, the explain can be given in the same way.

Based on above qualitative analysis, we now specifically consider a medium consisting of a mixture of left- and right-handed ($\Delta$-type) molecules. The optical waves propagating in the medium is described by the Maxwell equation [18]:

$$-\partial_{zz} E_{ij} + c^{-2} \partial_t E_{ij} = -\mu_0 \partial_t P_{ij}.$$
The coherent driving fields enter the mediums with different enantiomeric excess. The coherent driving fields enter the mediums with \( \Omega_{32} (0) = 10 \gamma \), \( \Omega_{21} (0) = \gamma / 10 \), satisfying the condition (5). Optical depth is set as \( \zeta = 0.2 \). The other parameters are the same as those in Fig. 3.

\[
\frac{1}{2} P_{ij} e^{-i(\nu_{ij} t - k_{ij} z + \phi_{ij})} + \text{c.c.}, \quad \text{with slowly varying polarization} \quad P_{ij} = 2N\mu_{ij}(p^{(+)} \sigma_{ij}^{(+)} + p^{(-)} \sigma_{ij}^{(-)}), \quad \text{where} N \text{ is the total molecular density,} \quad p^{(\pm)}, \quad \text{are the percentages of the two enantiomers. In the slowly-varying amplitude and phase approximation, the Maxwell equation are reduced to}
\]

\[
\frac{\partial \Omega_{ij}}{\partial z} = i \frac{N\mu_{ij}^2 \nu_{ij}}{\hbar \epsilon_{0} c} \left( p^{(+)} \sigma_{ij}^{(+)} + p^{(-)} \sigma_{ij}^{(-)} \right) e^{-i\theta_{ij}}. \tag{6}
\]

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

\[
\frac{\partial \Omega_{11}}{\partial z} = i \frac{N\mu_{11}^2 \nu_{11}}{\hbar \epsilon_{0} c} \left( -\frac{1}{4} \Omega_{21} \Omega_{32} \delta P + \frac{1}{2} i \lambda_{21} \Omega_{31} \right) \tag{7a}
\]

\[
\frac{\partial \Omega_{21}}{\partial z} = i \frac{N\mu_{21}^2 \nu_{21}}{\hbar \epsilon_{0} c} \left( -\frac{1}{4} \Omega_{31} \Omega_{32} \delta P + \frac{1}{2} i \lambda_{31} \Omega_{21} \right) \tag{7b}
\]

with \( \delta P = p^{(+)} - p^{(-)} \) being the enantiomeric difference. To achieve desirable chirality identification, the three coherent fields should satisfy the condition (5) at \( z = 0 \). Furthermore, we define the transmission coefficient of the probe \( \Omega_{21} \) as \( T = |\Omega_{21}(z)|^2 / |\Omega_{21}(0)|^2 \) and the corresponding absorption intensity \( I = 1 - T \). Additionally, if assuming \( |\Omega_{32}| \gg \gamma_{12}, \gamma_{23}, \) 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 gotten by solving Eqs. (7a)-(7b):

\[
h^{(\pm)} = 1 - \frac{1}{4B} e^{-C \zeta} \times \left[ (1 - A \pm 2\delta P) \left( 1 - e^{D \zeta} \right) + \sqrt{B} \left( 1 + e^{D \zeta} \right) \right]^2 \tag{8}
\]

with \( \zeta = N\mu_{11}^2 \nu_{11} z / (\hbar \epsilon_{0} c \Gamma_{21}), \quad A = \mu_{21}^2 \nu_{21} / (\mu_{11}^2 \nu_{11}), \quad B = (1 - A)^2 / 4A(\delta P)^2, \quad C = \Gamma_{21}(1 + A + \sqrt{B}) / [2(\gamma_{12} + \gamma_{13})], \quad D = \Gamma_{21} \sqrt{B} / [2(\gamma_{12} + \gamma_{13})] \). 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

\[
h^{(\pm)} \simeq \frac{2\Gamma_{21} \zeta}{\gamma_{12} + \gamma_{13}} p^{(\pm)}, \tag{9}
\]

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

The above analysis can be verified by numerical solving the master equations (5a)-(5e) and the relevant Maxwell equation (6). Fig. 3 presents the probe transmission \( T \) and the normalized absorption intensity \( I = T / (h^{(+)} + h^{(-)}) \) at optical depth \( \zeta = 0.2 \) versus the detuning \( \Delta \). 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; (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 range the enantiomeric difference \( \delta P \) can be read directly from the difference of the heights of two characteristic peaks \( \delta P = h^{(+)} - h^{(-)} \), namely, \( \delta P \simeq \delta p' \). Table I also tells us that for stronger \( \Omega_{32} \) and smaller \( \zeta, \delta p' \) 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 \( \delta P \). When \( \zeta \) increases, \( \delta p' \) is never linearly.

| \( \delta P \) (%) | \( \zeta = 0.05 \) | \( \zeta = 0.1 \) | \( \zeta = 0.2 \) |
|-----------------|----------------|----------------|----------------|
| 0               | 0              | 0              | 0              |
| 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         |
| ±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         |
FIG. 4: (Color online) The difference of the heights of two characteristic peaks $\delta p' = h^{(+)} - h^{(-)}$ as a function of enantiomeric difference $\delta p = p^{(+)} - p^{(-)}$ at different optical depth: (a) $\zeta = 0.2$; (b) $\zeta = 2$. The other parameters are the same as those in Fig. 3.

proportional to $\delta p$, see specifically Fig. 4(b). For this case we can not read directly the percentages of the chiral molecules from the heights of characteristic peaks. However, note that whether in linear regime or not, $\delta p'$ is always a monotonic function of $\delta p$. Thus, for a given $\delta p'$ (attained by measuring the absorption peaks) the percentage of two enantiomers can always be accurately identified from the $\delta p' \cdot \delta p$ curve.

Discussion and conclusions—Coherence effects, such as EIT, coherent population trapping, lasing without inversion, and so on, was firstly 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, such as 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 have been demonstrated for significantly enhancing various coherence effects, such as EIT [22, 23] and four-wave mixing [25]. On the other hand, 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 cold molecular ensemble in HC-PCF, an effective method to probe chirality of a small amount of molecular sample is possible.

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, $\lambda_{ij}$ 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 is then obtained by integrating the density matrix elements over the molecular distribution on velocities, i.e., $\sigma_{ij} = \langle \pi u^2 \rangle^{-1/2} \int_{-\infty}^{+\infty} \sigma_{ij}(u_z) \exp \left(-u_z^2/2\langle u^2 \rangle\right) du_z$ with $\langle u^2 \rangle$ 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 $\Omega_{32}$ 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 an 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 sensitive chirality-probe and lead to applications in organic chemistry, biochemistry, and pharmacology.

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