Laser induced persistent orientation of chiral molecules

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We show, both classically and quantum mechanically, enantioselective orientation of gas phase chiral molecules excited by laser fields with twisted polarization. Counterintuitively, the induced orientation, whose direction is laser controllable, does not disappear after the excitation, but stays approximately constant long after the end of the laser pulses, behavior unique to chiral systems. We computationally demonstrate this long-lasting orientation using propylene oxide molecules (CH\(_3\)CH\(_2\)CH\(_2\)O, or PPO) as an example, and consider two kinds of fields with twisted polarization: a pair of delayed cross-polarized pulses, and an optical centrifuge. This novel chiral effect opens new avenues for detecting molecular chirality, measuring enantiomeric excess and separating enantiomers with the help of inhomogeneous external fields.

I. INTRODUCTION

Chiral molecules exist in two enantiomeric forms. The two enantiomers are mirror images of each other, and they are nonsuperimposable by translation and rotation [1]. Molecular chirality is an omnipresent natural phenomenon of extreme importance in physics, chemistry and biology [2]. The ability to discriminate and separate mixtures of enantiomers is important, for example, in drug synthesis as different enantiomers of chiral drugs may exhibit strikingly different biological activity.

The related studies of gas phase chiral molecules focus on the measurements of enantiomeric excess, handedness of a given compound, and on devising techniques for manipulating mixtures containing both enantiomers [3–21]. In addition, over the years more ambitious directions were considered theoretically - laser assisted asymmetric synthesis, enantiomeric interconversion and purification (e.g. [3, 22] and references therein).

Recently, a pair of non-resonant delayed cross-polarized laser pulses was proposed as a new tool for discrimination of chiral molecules [23–25] and the underlying classical enantioselective molecular orientation mechanism was exposed [24–25]. The approach was extended to general fields with time-dependent polarization twisting in a plane. Optical fields with fixed linear polarization are unable to induce molecular orientation because of the symmetry of light interaction with the induced dipole. Polarization twisting in a certain plane breaks that symmetry and defines a preferred spatial direction perpendicular to that plane that depends on the sense of polarization rotation. When interacting with molecules, the twisted field has a two-fold effect. First, it induces unidirectional rotation (UDR) of the most polarizable molecular axis in the plane of polarization twisting [26–31], thereby orienting the averaged angular momentum vector (\(\mathbf{\ell}\)) perpendicular to the plane. In addition, in the case of chiral molecules, the twisted field induces an orienting torque along the most polarizable axis, thus orienting the molecule itself perpendicular to the above plane [24–25]. The direction of orientation depends on both the sense of polarization twisting and the handedness of the molecule. A pair of delayed cross-polarized laser pulses [26–27] provides the simplest example of the field with twisted polarization, but there are also more complex fields, such as chiral pulse trains [22–24], polarization-shaped pulses [35–37] and optical centrifuge [38–42]. Most recently, the orientation of chiral propylene oxide (PPO) molecules by means of an optical centrifuge was experimentally achieved in [43], thus providing the first demonstration of enantioselective laser control over molecular rotation.

In this paper, we substantiate the enantioselective molecular orientation by twisted fields in two significant ways. First, we provide a fully quantum treatment of this behavior and, second, we show that the orientation in chiral molecules is long-lived. Fully quantum studies on laser-driven PPO are provided and compared to classical results. Two implementations of the laser fields with twisted polarization are considered - a pair of delayed cross-polarized pulses, and an optical centrifuge. The laser-induced orientation is shown to be robust against normally detrimental temperature effects.

The paper is organized as follows. In Section II, the conditions needed for long-lasting field-free orientation in a pulse-excited molecular ensemble are discussed, and we show that chiral molecules excited by a laser field with twisted polarization satisfy these conditions. In Section III, the results of classical and fully quantum simulations of the enantio-selective orientation at thermal conditions are provided and compared with one another. Section IV concludes the paper.

These results substantiate, quantum mechanically, the classical expectation [24–25] that the orientation does not disappear after the excitation, but stays at an approximately constant level long after the end of the laser field.

II. CLASSICAL ANALYSIS

In this section we use classical mechanics to analyze conditions leading to the existence of permanent orientation in a gas of field-free rotating asymmetric-top molecules. We show that excitation of an isotropic ensemble of chiral molecules by twisted fields satisfies these conditions.

Background. The Binet construction [44, 45] allows...
one to classify the trajectories traversed by the angular momentum vector in the molecule-fixed frame defined by the three principal axes of moment of inertia tensor. Figure 1(a) shows one of the enantiomers (right handed, R) of our example molecule, propylene oxide (CH$_3$CH=CH$_2$O, or PPO) with two sets of axes: principal axes of moment of inertia and polarizability tensors. Hereafter, the letters $a$, $b$, $c$ refer to the inertia principal frame, whereas the numbers 1, 2, 3 refer to the polarizability principal frame. We briefly describe the construction. Free motion of an asymmetric-top rotor has four constants of motion, energy and the three components of angular momentum expressed in an inertial frame (laboratory fixed frame). In the frame of principal axes of the inertia tensor [see Fig. 1(a)], the allowed trajectories of the angular momentum vector satisfy the following equations

$$\frac{L_a^2}{2EI_a} + \frac{L_b^2}{2EI_b} + \frac{L_c^2}{2EI_c} = 1; \quad \frac{L_a^2 + L_b^2 + L_c^2}{L^2} = 1,$$

where $I_j$ are the moments of inertia, $L_j$ are the components of the angular momentum vector ($j = a, b, c$), $E$ is the rotational energy and $L$ is the magnitude of the angular momentum vector. The moments of inertia are ordered according to $I_a < I_b < I_c$. The first expression defines an ellipsoid with semi-axes $\sqrt{2EI_a}$, $\sqrt{2EI_b}$ and $\sqrt{2EI_c}$, these coincide with the principal axes of inertia tensor. The second expression defines a spherical shell with radius $L$. The angular momentum vector tip moves on the ellipsoid-spherical shell intersections. In addition to the six stationary rotations about each of the three inertia tensor principal axes, the allowed trajectories can be divided into sets of closed curves, as shown in Fig. 1(b).

For $\sqrt{2EI_0} < L < \sqrt{2EI_b}$ and $\sqrt{2EI_b} < L < \sqrt{2EI_c}$, the trajectories are divided into two sets of curves $T_k$ ($k = a, c$) enclosing the poles on $a$ and $c$ axes, respectively [see Fig. 1(b)]. The sign of $L_k$ is conserved on these trajectories, and depending on the enclosed pole (either on positive or negative side of the axis), we denote the corresponding sets by $T^+_k$. Although the Binet construction provides a qualitative picture of trajectories as seen from the molecule-fixed frame, it allows to deduce some valuable information about the motion in the laboratory frame, as well. In case of a single top, we can assume that the conserved vector of angular momentum $\ell$ points along the laboratory $Z$ axis. For definiteness, we assume $T_a$ trajectory, meaning that $\ell$ moves on a “taco-shaped” curve around one of the poles on the $c$ axis [see Fig. 1(b)]. This implies that in the laboratory frame the $c$ axis “precesses” around $\ell$ while the sign of the projection $\hat{c} \cdot \hat{\ell} = \hat{c} \cdot \hat{Z}$ remains unchanged, which means preferred orientation of the $c$ axis in the course of time. Notably, even in case of an ensemble of asymmetric tops, i.e. when initially angular momenta vectors point in various directions, the permanent orientation is still possible. In that case permanent orientation corresponds to the ensemble-averaged quantities $\langle \hat{a} \cdot \hat{Z} \rangle$ or $\langle \hat{c} \cdot \hat{Z} \rangle$, which constitutes a constant sign. This may be achieved by, first, orienting the averaged angular momentum vector $\langle \ell \rangle$ (e.g. along $Z$ axis) and, second, breaking the symmetry between $T^+_a$ vs $T^+_c$ trajectories.

**Particular case of twisted polarization.** Here we consider a specific example of an ensemble of chiral molecules excited by pair of delayed cross-polarized laser pulses [26][28], which constitutes the simplest implementation of a field with twisted polarization. We show that such an excitation leads to the conditions discussed above. Here, the first pulse is polarized along the laboratory $X$ axis, while the polarization of the second one is in the $XY$ plane at $+\pi/4$ to $X$ axis. The first pulse induces alignment of the most polarizable molecular axis. For the sake of simplicity of the qualitative analysis, we assume that after the first pulse all the molecules are perfectly aligned along the $X$ axis, and are stationary [24]. Initially, we consider only half of all molecules, in which the most polarizable axis points along $+X$ [see Fig. 2(a)]. Angle $\varphi \in [0, 2\pi]$ is the angle between the $Y$ axis and the $x_2$ axis, lying in the $YZ$ plane [see Fig. 2(b)]. The aligned molecules are uniformly distributed in $\varphi$. The interaction potential, $U$ and torque, $T$ induced by a non-resonant optical field are given by

$$U = -\frac{1}{2} \langle d_{\text{ind}} \cdot \mathbf{E} \rangle \quad T = \langle d_{\text{ind}} \times \mathbf{E} \rangle,$$

where the angle brackets denote time averaging over the optical cycle, $d_{\text{ind}} = \alpha \mathbf{E}$ is the induced dipole, $\alpha$ is the polarizability tensor, and $\mathbf{E}$ is the vector of the electric field. The duration of the laser pulses is assumed to be short as compared to the typical rotational periods of the chiral molecules, therefore the effect of the second pulse is considered in the impulsive approximation, $\Delta \mathbf{L} \propto \mathbf{T}$.

The second pulse (twisted with respect to the first one) induces unidirectional rotation in the $XY$ plane resulting in orientation of $\langle \ell \rangle$ along $Z$, which constitutes the first criterion for the long-lasting orientation. In addi-
tion, there is an orienting torque acting on the aligned most polarizable axis, $x_1$. To check whether the second criterion for the long-lasting orientation is satisfied, we evaluate the torques along the molecular $a$ and $c$ axes, $\tau_k (k = a, c)$

$$\tau_k \propto C_k \sin (2\phi) + D_k \sin (\phi + \phi_k),$$

(1)

where $C_k = (\alpha_{33} - \alpha_{22}) R_{k3}/2, D_k = \sqrt{A_k^2 + B_k^2}, A_k = (\alpha_{11} - \alpha_{33}) R_{k2}, B_k = (\alpha_{11} - \alpha_{22}) R_{k3},$ and $\cos (\phi_k) = A_k/D_k$. Here, $R_{nm}$ are the elements of the orthogonal rotation matrix relating the principal polarizability and inertia frames. (See Supplementary Materials for details.)

For chiral molecule, the two frames do not align [see Fig. 1(a)] and the off-diagonal elements of $R$ differ from zero. In this case, the subdomains of positive and negative $\tau_k$ in the interval $\varphi \in [0, 2\pi]$ are not equal, which is precisely the required $T_{11}^k$ vs $T_{12}^k$ asymmetry. When the two frames do align (as in non-chiral molecules), $R$ becomes diagonal resulting in torques $\tau_a = C_a \sin (2\varphi)$ and $\tau_c = A_c \sin (\varphi)$. In this case, the $T_{11}^c$ vs $T_{22}^c$ symmetry is preserved.

It may be shown that Eq. 1 remains the same for molecules with the most polarizable axis, $x_1$ pointing along $-X$. For the complimentary enantiomer ((S)-PPO), signs of some of the elements of $R_{nm}$ are reversed, resulting in the opposite orientation direction.

### III. NUMERICAL SIMULATIONS

In this section, we present the results of numerical simulations of the laser driven orientation dynamics of (R)-PPO molecule. We consider two implementations of laser fields with twisted polarization, a pair of delayed cross-polarized pulses [26–28] and an optical centrifuge [38–42]. The behavior of an ensemble of $N > 1$ molecules is investigated using both classical and quantum mechanical tools. The chiral molecule is modeled as a rigid asymmetric top having anisotropic polarizability and a dipole moment. Supplementary Table 1 summarizes the molecular properties used.

For the classical simulations, the behavior of a thermal ensemble was simulated using the Monte Carlo approach. Our numerical scheme relies on solving the Euler equations for angular velocities and parametrizing the rotations by quaternions [46]. The details of the scheme may be found in [25]. Before the excitation, the molecules are isotropically distributed, and the angular velocities are assigned according to the Boltzmann distribution for a gas at a temperature $T$. The rotational temperatures $T'_{\text{rot}}$ of both the transient and the persistent dipole are temperature dependent and are lower than the temperature of the gas. The sign of orientation is opposite for the second enantiomer (S)-PPO. The classical mechanism explaining the induced enantioselective transient orientation is described elsewhere [24–25]. The maximal amplitudes of both the transient and the persistent dipole are temperature dependent and become lower with increasing the temperature. The long-lasting dipole signals shown in Figure 3 exhibit small-amplitude beats about the asymptotically constant values. These beats stem from the statistical nature of the simulations, and they are inversely proportional to $\sqrt{N}$.

To access the influence of the quantum effects on the permanent orientation, we carried out fully quantum mechanical simulations of the dynamics of (R)-PPO molecules kicked by the two pulses. The pulse envelope used in the quantum simulations, is given by Sup. Eq. 6. The difference between this envelope and the Gaussian one used in the classical simulation is insignificant, as the pulses are short and the difference in their integrals is negligible ($< 1\%$). The rotational temperature was set to $T = 5 \text{ K}$. The delay of the second pulse was adjusted to the moment of maximal alignment of the most polarizable axis towards the $X$ axis, as measured using the alignment factor (see Sup. Eq. 23). Depending on the initial conditions, the pulses excite the molecules to the states with $J$
FIG. 3. Classical ensemble averaged projection of the molecular dipole on $Z$ axis, $\langle \mu_Z \rangle (t)$ for the case of $(R)$-PPO molecules excited by a pair of cross-polarized pulses. Here $N = 6 \times 10^5$. The curves span the range of temperatures $T \in [10, 100]$ K, in steps of 10 K. Parameters of the pulses are: $I_0 = 0.5 \times 10^{14}$ W/cm$^2$, FWHM = 0.10 ps. First pulse is centered at $t = 0$, while the delay of the second pulse is optimized for each $T$ (see text). Horizontal line is set at $\langle \mu_Z \rangle = 0$. The inset is an amplified version of the figure.

FIG. 4. Quantum expectation value of projection of the molecular dipole on $Z$ axis, $\langle \mu_Z \rangle (t)$ for the case of $(R)$-PPO molecules excited by a pair of cross-polarized pulses. Here $N = 6 \times 10^5$. The curves span the range of temperatures $T \in [10, 100]$ K, in steps of 10 K. Parameters of the pulses are: $I_0 = 0.5 \times 10^{14}$ W/cm$^2$, FWHM = 0.10 ps. First pulse is centered at $t = 0$, while the delay of the second pulse is optimized for each $T$ (see text). Horizontal line is set at $\langle \mu_Z \rangle = 0$. The inset is an amplified version of the figure.

FIG. 5. Classical ensemble averaged projection of the molecular dipole on the $Z$ axis, $\langle \mu_Z \rangle (t)$ for the case of $(R)$-PPO molecules excited by the optical centrifuge. The curves span the range of temperatures $T \in [10, 150]$ K, in steps of 10 K. Here $N = 9 \times 10^7$. Parameters of the pulse are: $I_0 = 5 \times 10^{12}$ W/cm$^2$, $\beta = 0.065$ ps$^{-2}$ and $t_{\text{on}} = t_{\text{on}} = 2$ ps and $t_f = 20$ ps. The dashed curve represents the scaled $\varepsilon^2(t)$, see Eqs. 2 and Sup. Eq. 7. The horizontal line is set at $\langle \mu_Z \rangle = 0$.

We model the electric field of such a pulse by

$$
\mathbf{E} = \varepsilon(t) \left[ \cos \left( \beta t^2 \right) \mathbf{e}_X + \sin \left( \beta t^2 \right) \mathbf{e}_Y \right] \cos (\omega t),
$$

where $\beta$ is the angular acceleration and $\varepsilon(t)$ is the envelope (dashed curve, Fig. 5) for explicit expression see Sup. Eq. 7. As in the previous case, we start with the results of the classical simulations. Figure 5 shows the ensemble-averaged projection of the molecular dipole on the $Z$ axis as a function of time, $\langle \mu_Z \rangle (t)$ resulting from excitation by the optical centrifuge. The temperature changes in steps of 10 K in the range of $T \in [10, 150]$ K. Akin to the double pulse scheme, excitation by the optical centrifuge results in enantioselective dipole signal. The signal drops down when the centrifuge is switched off after 20 ps. However, the orientation signal does not vanish completely, but levels off at a non-zero value. Although the amplitude of the transient signal is higher in the case of the double pulse excitation, the amplitudes of the long-time signals are higher in this scenario, making the optical centrifuge more efficient in inducing the permanent dipole orientation. The amplitudes of the signals, both during the driven and field-free dynamics periods are temperature dependent and decrease with temperature.

To investigate the long-lasting dynamics of the induced dipole moment, we carried out a fully quantum mechanical simulation of the centrifuge-driven $(R)$-PPO rotational dynamics. Centrifuge pulse had the same parameters as shown in the caption to Fig. 5, while the rotational temperature was set to $T = 5$ K. Such a pulse excites angular momenta of up to 40th, with the mean excitation of about 30h, which is significantly above the average thermal value of 3.5h. Higher angular momentum results in a better quantum-classical agreement, as compared to the double pulse excitation. The short-time dipole signal is shown in Fig. 5(a). During the pulse, the polarisation peaks at about 0.15 D, and it stays practically constant at the level of $\approx 0.03$ D after the end of the pulse, with a small-amplitude beating structure present at long times. For comparison, the classical signal obtained under the same conditions is plotted and the correspondence with the quantum results is very good up to $t \approx 60$ ps. On the nanosecond time-scale [Fig. 5(b)], one

Optical centrifuge excitation. Optical centrifuge is a laser pulse, whose linear polarization undergoes an accelerated rotation around its propagation direction. Optical centrifuge is a laser pulse, whose linear polarization undergoes an accelerated rotation around its propagation direction. Optical centrifuge is a laser pulse, whose linear polarization undergoes an accelerated rotation around its propagation direction.
can observe multiple beats, however the coarse-grained
time-averaged quantum signal remains positive, and
almost constant as the classically predicted one.

**Dynamical Tunneling.** Due to the coupling of dif-
ferent $K$-states by the rotational Hamiltonian (see Sup.
Eq. 8), the quantum mechanical rigid asymmetric top
does not have eigenstates with the angular momentum
being oriented within the molecular frame. As a conse-
quence, any state that is internally oriented at some point
in time can not be an eigenstate and will oscillate be-
tween being oriented and anti-oriented, an effect known
as dynamical tunneling [49]. In other words, a clock-
wise internal rotation would eventually become counter-
clockwise, and vice versa. This leads to a breakdown of
quantum-classical analogy, as the classical model intro-
duced in Sec. 1 assumes a strict separation of clockwise
and counter-clockwise rotation (no interchange between
$T^+_K$ and $T^-_K$). Therefore, one would expect no perma-
nent orientation after turn-off of all external fields for a
quantum mechanical chiral rotor.

Yet, in spite of the dynamical tunneling, our simula-
tions show a significant long-time orientation of the quan-
tum rotors, equal in magnitude to the classical model.
The reason is that the dynamical tunneling time grows
very fast with the angular momentum [50], being larger
the more the angular momentum is (anti-)oriented along
the $a$ or $c$ axis. Indeed, already for $J = 10h$, the tunnel-
ing time may exceed microseconds [50], explaining why
orientation was observed in the quantum simulations on
a nanosecond time-scale. As a matter of fact, the slight
decrease of the base line visible in Fig. 6 (thick red line)
is possibly a signature of the dynamical tunneling.

At this point one should also note that whilst dynam-
ical tunneling would be of greater importance on longer
time-scales than the ones presented here, other effects
(neglected here), like rovibrational couplings, hyperfine
structure [51] and intermolecular collisions are likely of
higher importance and would probably bury any signs of the dynamical tunneling in an actual experiment.

**Extrapolation to room temperature.** Due to the
statistical nature of our classical simulations on one hand
and basis size limitations of the quantum simulation on
the other, direct simulations at room temperature are
highly numerically demanding, therefore we resort to ex-
trapolation. Figure 6 shows the average long term value
of the classical calculated $\langle \mu_Z \rangle$, denoted by $\langle \mu_Z \rangle$ as a function of temperature on the double logarithmic scale. The error bars for log
$\mu_\langle Z \rangle$ are obtained using the standard error propagation for-

mula [52]. Solid lines are the fit function,

$$\text{Fig. 6. Quantum expectation value of the projection of the}
\text{molecular dipole on the Z axis, } \langle \mu_Z \rangle (t) \text{ for the case of (R)}-
PPO molecule excited by the optical centrifuge pulse. Par-
\text{ameters are similar to those of Fig. 6 (a) The short-time dipole}
\text{signal (solid) and its classical counterpart (dotted). The dashed}
\text{curve represents the scaled } z^2(t), \text{ see Eqs. 2 and Sup. Eq. 7.}
\text{(b) Long-time dynamics. Solid red - 100 ps time-average,}
\langle \mu_Z \rangle (t) = \frac{1}{\Delta t} \int_{t-\Delta t/2}^{t+\Delta t/2} \langle \mu_Z \rangle (t') \, dt'$

sponding light-induced voltage) favor its detection using
high-speed nanosecond electronics.

The appearance of long-time orientation in a gas sam-
ple can be detected by means of Coulomb explosion
[7, 8, 14, 17, 18, 43] and nonlinear optics through high
harmonic generation of various orders [54, 57], especially
by means of the second harmonic generation. Another
approach to detecting the induced orientation is by mea-
suring THz emission due to free induction decay of co-
herently oscillating molecular dipoles [58, 59].

**IV. CONCLUSIONS**

Summarizing, we have demonstrated long-lived ori-
entation of chiral molecules by laser fields with twisted
polarization using both classical mechanics and fully
quantum-mechanical treatments. The problem was analyzed using propylene oxide molecules, as an example, and considering two different implementations of the twisted optical field: (i) a pair of delayed cross-polarized laser pulses, and (ii) an optical centrifuge. We found very good agreement between the classical and quantum approaches over a wide range of experimentally relevant parameters, and over an extended time range. Significantly, we demonstrated the novel chiral phenomenon of persistent molecular orientation lasting long after the end of the exciting pulses. This counterintuitive effect was first conjectured in our previous papers [21,22] utilizing a classical approach, but its validity is now verified on a much longer time scale (several orders of magnitude longer) where the use of quantum treatment is unavoidable. The sign of the oriented dipole moment depends on both the sense of polarization twisting and the handedness of the molecule. This long-lasting orientation provides new modalities for detecting molecular chirality with the help of optical harmonics generation, or direct high-speed measurements of electric fields caused by laser-induced macroscopic polarization of the gas. Moreover, it is well known that optical pre-orientation of molecules affects their deflection by inhomogeneous fields (see e.g. [61,63], references therein, and recent reviews [61,65]). Enantioselective orientation effects considered in this paper may open up new avenues for separation of molecular enantiomers.

We note that ongoing experiments in Milner’s group show initial signals of the long-time enantioselective orientation, supporting our theoretical predictions. A report on these results and the corresponding theoretical analysis is in preparation [67].

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