Detecting chirality in spatially oriented molecules by Coulomb explosion imaging

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We present a novel method for detecting chirality in the gas phase: Chiral molecules are spatially aligned in three-dimensions by a moderately strong elliptically-polarized laser field. The momentum distributions of the charged fragments, produced by laser-induced Coulomb explosion, are recorded. They show distinct three-dimensional orientation of the enantiomers, when the laser polarization ellipse is rotated by a non-right angle with respect to the norm vector of the detector plane. The resulting image asymmetry is directly connected to the enantiomeric excess and to the absolute handedness of the underlying molecular sample. We demonstrated our scheme computationally for camphor (C_{10}H_{16}O), with its methyl-groups as marker fragments, using robust quantum-mechanical simulations geared toward experimentally feasible conditions. The combination of three-dimensional alignment with one-dimensional orientation, induced by an additional dc field, increases the asymmetry and sensitivity of the method. Our method is simple and robust. Its high sensitivity is comparable to other modern chiroptical approaches and can be readily optimized for any chiral molecule with an anisotropic polarizability tensor by adjusting the laser field configuration.

Chiral molecules exist in structural forms known as enantiomers, which are mirror images of one another that are non-superimposable by translation and rotation. The chemical behavior of molecular enantiomers can be profoundly different. Particularly in the pharmaceutical industry, methods to differentiate between them or to determine the enantiomeric excess (ee) of a chiral sample are important. In recent years, there have been considerable advances in gas-phase chiroptical techniques and a variety of such methods have emerged, for example, using phase-sensitive microwave spectroscopy[10] Coulomb explosion imaging with coincidence detection,[11] photoelectron circular dichroism (PECD)[2] chiral-sensitive high-harmonic generation,[5,6] or attosecond-time-resolved photoionization.[3] These approaches offer improved sensitivity and their success is based on exploiting electric-dipole interactions for chiral discrimination,[12] producing stronger signals than circular dichroism from magnetic-dipole interactions.

Coulomb explosion imaging is a powerful and efficient approach to retrieve the instantaneous absolute structures of complex molecules.[13,14] Applied to chiral molecules, coincident imaging of fragments emitted from the chiral center can be used to determine the handedness of their enantiomers, in the conceptually most straightforward way by coincident detection of all fragments attached to the stereocenter.[14] For axially chiral molecules, it has been demonstrated that it is sufficient to only correlate two different fragments, if the molecules are pre-aligned along their axis of chirality.[13]

Here, we explore the effect of spatial three-dimensional (3D) alignment of molecules in Coulomb explosion imaging in order to sensitively probe the ee and the handedness of a chiral sample with it. Using accurate computational procedures, we demonstrate that 3D alignment by an elliptically-polarized non-resonant field can break the symmetry in a fragments position and momentum distribution in the detector plane, if the polarization ellipse is tilted by an angle $0 < \beta < 90^\circ$ with respect to the norm vector of the detector. The asymmetry between the detector’s left and right halves gives access to the ee and handedness of chiral samples. This method is more robust than previous Coulomb-explosion-based approaches and our theoretical estimates for the sensitivity to the ee are comparable to other modern chiroptical techniques, such as PECD. To further enhance sensitivity we also explore the effect of one-dimensional (1D) orientation combined with 3D alignment.

Figure 1 illustrates the underlying idea of our approach, which is demonstrated for the prototypical chiral molecule camphor (C_{10}H_{16}O). A non-resonant elliptically-polarized laser field is applied to achieve 3D alignment. The most polarizable axis of the molecule $p$ is aligned along the major axis $Z_\text{L}$ of the elliptical field and the second most polarizable axis $q$ along the minor elliptical axis $X_\text{L}$. We chose the three distinct methyl (CH$_3$) groups in camphor as marker fragments to differentiate between the $R$ and $S$ enantiomers. Their flight directions can be observed experimentally as momentum distributions of the CH$_3^+$ ions resulting from multiple ionization followed by Coulomb explosion of the molecule[15,16].

We assume that two-body dissociation events produce
equal initial momenta for CH$_3$ fragments at three different molecular sites. By normalizing the size of the Newton sphere to one, the momenta distributions are given by the position distributions of the CH$_3$ groups. These methyl-group distributions in the detector plane are schematically plotted in Figure 1 for the idealized case of perfect 3D alignment. Fixed in the $X_L$, $Z_L$ laser polarization plane, the molecule orients itself in one of four equally preferred ways, which are related by 180° rotations about the $p$ and $q$ most polarizable axes of the molecule. Fixing the plane of elliptical polarization in the $XZ$ laboratory plane, the cartesian coordinates of an atom in the molecule projected onto the $YZ$ plane of the detector for all four possible spatial molecular orientations are given by

$$
\begin{align*}
  r &= (y, + (z \cos \beta - x \sin \beta)) \\
  r_p &= (-y, + (z \cos \beta + x \sin \beta)) \\
  r_q &= (-y, - (z \cos \beta + x \sin \beta)) \\
  r_{pq} &= (+y, - (z \cos \beta - x \sin \beta))
\end{align*}
$$

where $x$, $y$, $z$ denote the cartesian coordinates of an atom in the principal axis of polarizability frame of the molecule. The subscript indices $p$ and $q$ denote cartesian vectors obtained by 180° rotations about the respective molecular polarizability axes, which in the case of perfect 3D alignment coincide with the $Z_L$ and $X_L$ axes of the polarization ellipse. The angle $\beta$ is the angle between the major $Z_L$ axis of the ellipse and the norm ($\mathbf{e}_X$) of the detector. It describes the rotation of the polarization ellipse about the $Y$ axis.

The four different positions $r$, $r_p$, $r_q$ and $r_{pq}$ in the plane of the detector are plotted in Figure 1 for the three carbon atoms that belong to the methyl-groups for $R$ and $S$ camphor. Different enantiomers have opposite signs of the $Y$ component of each position vector in (1). When $\beta = n \cdot 90^\circ$, $n = 0, 1, 2, \ldots$, the four different positions in the plane of the detector for each atom produce an image, which is symmetric with respect to the inversion of both $Y$ and $Z$ axes, as shown in Figure 1 a. Since the position vectors for the $R$ and $S$ enantiomers differ only in the sign of the $Y$ coordinate, the resulting projections will look exactly the same for different enantiomers. However, when the $\beta \neq n \cdot 90^\circ$ the symmetry with respect to the inversion of the $Y$ axis in (1) will be broken. As a result, the sums of the four equivalent molecular spatial orientations will exhibit distinctly different projections on the detector plane for the $R$ and $S$ enantiomers, see Figure 1 b. The detector images of the enantiomers are unsymmetric with respect to the left and right parts and are in fact mirror images of each other for the enantiomers. This allows for the determination of the $cc$ and the handedness of a chiral sample. Notably, the present approach does not require coincidence measurements of different fragment species.

To benchmark our scheme we have performed quantum-mechanical calculations of the rotational dynamics of camphor using the accurate variational procedure Rich-Mol[22] which simulates the rotation-vibration dynam-
ics of molecules in the presence of external fields. The field-free rotational motion was modelled using the rigid-rotor Hamiltonian with the rotational constants \( A = 1446.968977 \) MHz, \( B = 1183.367110 \) MHz, and \( C = 1097.101031 \) MHz.\(^\text{15}\) Simulations of the field-induced time-dependent quantum dynamics employed wave packets built from superpositions of field-free eigenstates including all rotational states of the molecule with \( J \leq 40 \), where \( J \) is the quantum number of overall angular momentum. Only the vibrational ground-state was considered, reflecting the conditions in a cold molecular beam. The time-dependent coefficients were obtained by numerical solution of the time-dependent Schrödinger equation using the time-discretization method with a time step of \( \Delta t = 10 \) fs and a Lanczos-based approach for the time-evolution operator.\(^\text{19}\)

The field interaction potential was represented as a multipole moment expansion of order up to the polarizability tensor were calculated using the coupled cluster method CCSD(T) with the augmented-correlation-consistent basis set aug-cc-pVTZ\(^\text{20,21}\) in the frozen-core approximation. The calculations were performed at the experimentally determined molecular geometry\(^\text{18}\) using CFOUR.\(^\text{22}\)

The elliptically-polarized adiabatic laser field was represented as

\[
E(t) = E_0 \sqrt{4 \log 2/(\pi \tau^2)} \exp \left( -4 \log 2(t - t_0)^2/\tau^2 \right) \\
\times \left[ (\cos(\omega t) \cos \beta + \frac{1}{\sqrt{3}} \sin(\omega t) \sin \beta) e_X + (\cos(\omega t) \sin \beta - \frac{1}{\sqrt{3}} \sin(\omega t) \cos \beta) e_Z \right]
\]

with the parameters \( E_0 = 4 \times 10^9 \) V/cm, corresponding to a laser peak intensity \( I = 6 \times 10^{11} \) W/cm\(^2\), \( \omega = 800 \) nm, \( t_0 = 440 \) ps, and \( \tau = 250 \) ps. The calculations were performed for \( \beta \) angles ranging from 0 to 90°. For some calculations we added the interaction between the permanent molecular dipole moment and a static electric field of 1 or 5 kV/cm aligned along the detector norm vector \( e_X \). A hypothetical strong probe pulse, causing the Coulomb explosion, was applied at a time \( t = 440 \) ps corresponding to the peak intensity of the alignment field. Idealized simulations were performed at an initial rotational temperature of \( T = 0 \) K, and for experimentally realistic conditions at \( T = 0.2 \) K.\(^\text{22}\)

The degree of 3D alignment is characterized by \( \langle \cos^2 \theta_{pZ} \rangle = 0.84 \) and \( \langle \cos^2 \theta_{qX} \rangle = 0.76 \) for \( T = 0 \) K. For a finite initial temperature of \( T = 0.2 \) K we obtained \( \langle \cos^2 \theta_{pZ} \rangle = 0.64 \) and \( \langle \cos^2 \theta_{qX} \rangle = 0.50 \).

The distributions of the methyl-group fragments of camphor in the YZ detector plane were simulated as the probability density distributions of the corresponding carbon atoms. These were obtained as a normalized sum of contributions from the three individual methyl-group carbon atoms with equal weights. As the recoil axes, we chose vectors along the molecular bonds connecting the carbon atoms in the methyl groups with the backbone of the molecule. To account for non-axial recoil, the calculated probability density distributions were convoluted with a Gaussian function with a full-width at half maximum (FWHM) of 30°, which is near typical experimental values.\(^\text{20}\)

**Figure 2 a** shows the calculated 2D projections of the probability density distributions for the carbon atoms in the methyl groups of \( R \) and \( S \) camphor for different \( \beta \) angles and an initial rotational temperature of \( T = 0 \) K. As expected, for \( \beta = 0, 90 \)°, the 2D projections are symmetric with respect to inversion of \( Y \) and \( Z \) axes. Thus, their averages for the four orientation look identical for the different enantiomers. The 2D density projections become asymmetric with respect to inversion of the \( Y \) axis for intermediate values of the \( \beta \) angle. In **Figure 2 a** the results are shown for \( \beta = 30 \)° and 60°. For different enantiomers the distributions are exact mirror images of each other in the \( YZ \) plane. For racemic mixtures, the 2D density, and consequently the momentum projections of the methyl-group fragments, will be symmetric to inversion of the \( Y \) axis, and the presence of an asymmetry between the left and right halves of the detector will thus indicate the ee.

To identify the parts of the detector images, which have the largest asymmetry and are therefore most sensitive to the ee, we propose to define an asymmetry parameter as a normalized difference \( A(\theta) = [N_1(\theta) - N_2(\theta)]/[N_1(\theta) + N_2(\theta)] \) between sectors in the right and left halves of the detector. Here, \( N_1(\theta) \) is the intensity in an angular sector of fixed width \( \Omega \) at \( \theta = 0 \ldots 180 \)°, i.e., in the right half of the detector. Thus, \( N_1(\theta) \) is the corresponding intensity in the left half of the detector. The asymmetry \( A(\theta) \) for \( \Omega = 30 \)° for different \( \beta \) is shown in **Figure 2 b**. The largest value of \( A \) for the \( R \) and \( S \) enantiomers, respectively, are obtained as \( A \approx 0.22 \) for \( \beta = 30 \ldots 50 \)° and \( A \approx -0.3 \) for \( \beta = 90 \)°.

Generally, the asymmetry values \( A \) depend on the molecule, its marker fragments, and their recoil axes with respect to the alignment plane. In the case of a large number of indistinguishable fragments attached at various molecular sites, e.g. hydrogen atoms, the total probability density will look more isotropic, even for strong 3D alignment. The degree of angular asymmetry will also be lowered when looking at fragments dissociating in directions nearly parallel to the alignment plane.

In the present case, there are three indistinguishable \( CH_3 \) fragments attached at different sites of camphor. The optimal value of the \( \beta \) angle can be thought of as the one that maximizes the overlap of the 2D probability density distributions of different \( CH_3 \) fragments. This leads to a more anisotropic total density distribution and a better contrast with respect to variation of \( \theta \).

The magnitude of angular asymmetry \( A(\theta) \) also depends on the degree of 3D alignment. The lower degree of alignment for a 0.2 K sample leads to more diffuse 2D projections of the probability density distributions and,
The present estimates of the maximum asymmetry for values of $\lambda$ can be achieved, for instance, by rendering the four equivalent alignment orientations of the molecule and its (approximate) polarizability tensor. In a standard setup as is typically used for studying molecular alignment, employing elliptically polarized non-resonant laser pulses producing the effect of 3D alignment with 1D orientation. Camphor still allows for two of the four orientations producing the effect of 3D alignment with 1D orientation.

One may consider to increase the degree of asymmetry by rendering the four equivalent alignment orientations of unequal probability. This can be achieved, for instance, by applying a dc electric field along the norm vector of the detector plane, known as mixed-field orientation. We calculated the asymmetry $A(\theta)$ for dc field strengths of 1 and 5 kV/cm at $T = 0$ K, shown in Figure 4; note that $A(S) = -A(R)$.

As the dc field breaks the symmetry with respect to the inversion of $Y$ and $Z$ axes, although the simultaneous inversion of both axes is still symmetric, the non-zero asymmetry can be observed even at $\beta = 90^\circ$. The maximal degree of asymmetry increases up to $\pm 0.4$ with increasing dc field strength, the effect however quickly saturates at stronger dc fields. The absolute sign of the asymmetry, defined as the difference between the left and right halves of the detector, as well as the optimal values of $\beta$ and $\theta$ remain the same as for pure alignment. This is rationalized by the fact that the mixed-field orientation in camphor still allows for two of the four orientations producing the effect of 3D alignment with 1D orientation. The mixed-field orientation effect however can only be achieved for polar molecules with a non-vanishing projection of the dipole moment onto the $pq$ plane of the most polarizable axes.

In conclusion, we demonstrated a novel and robust approach for detecting chirality based on the Coulomb explosion imaging of 3D aligned molecules. The method employs elliptically polarized non-resonant laser pulses in a standard setup as is typically used for studying molecular alignment. The chirality is revealed by an asymmetry in the 2D projections of ion momentum distributions. This paves the way to the sensitive analytic use of Coulomb explosion imaging for detecting the ee with a sensitivity comparable to PECD. Any molecule with three different principal polarizability components can be investigated this way.

Although we found that for camphor the methyl-group fragments deliver sufficient asymmetry, one has to keep in
mind that these fragments could possibly exhibit larger non-axial recoil velocities as assumed here based on typical experimental values, which would produce an additional smearing effect on the structures in the ion momentum distributions. The present approach therefore is best suited for chiral molecules with nearly-axially-recoiling atomic groups or utilizing more general analysis approaches.

When compared to existing coincidence Coulomb explosion imaging techniques, our approach does not require the correlated detection of multiple different fragments. This enables much faster data acquisition, which is highly advantageous for ultrafast time-resolved studies. Furthermore, our approach can clearly distinguish between the left- and right-handed enantiomers without any correlated measurements. The external fields can be further optimized to improve the sensitivity. In particular, we have demonstrated that mixed-field orientation can be used to enhance the asymmetry in the ion momentum distributions and thus the ee sensitivity of the method. The approach could be combined with PECD in a COLTRIM setup to provide additional information on the handedness of the sample.

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The data that support the findings of this study are available from the corresponding author upon request.

FIG. 3. Computed 2D projections of the averaged probability density distributions for carbon atoms in the methyl groups of $R$-camphor and $S$-camphor at the peak of the alignment field and an initial rotational temperature of $T = 0.2$ K. On the bottom panel, the asymmetry parameter $A$ as a function of the $\theta$ angle for both enantiomers is displayed. The results are shown for the optimal value of $\beta = 40^\circ$.

FIG. 4. Effect of the dc field on the asymmetry of the 2D projections of the averaged probability density distributions for carbon atoms in the methyl groups of $R$-camphor, computed at the peak of the alignment field for different values of $\beta$ and an initial rotational temperature of $T = 0$ K.

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