SPACE SCIENCES

Chiroptical activity of gas-phase propylene oxide predicting the handedness of interstellar circular polarization in the presolar nebula

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Propylene oxide, the first chiral molecule recently detected in the interstellar medium, has once again raised the question whether biomolecular chirality might have cosmic origins. However, accurate chiroptical properties of propylene oxide in the ultraviolet spectral range necessary to suggest possible asymmetric synthetic routes in the gas phase are scarce. Here, we report on the first experimental measurements of the anisotropy spectra of gas-phase propylene oxide in the vacuum ultraviolet spectral range. Our experimental results provide novel insights into the handedness of interstellar circular polarization at the dawn of molecular evolution of our star- and planet-forming region. Besides the fundamental importance of this new investigation for understanding the origin and evolution of homochirality on Earth, our high-resolution experimental electronic circular dichroism data will inspire new efforts in quantum computational spectroscopy.

INTRODUCTION

Propylene oxide, also known as 2-methyl oxirane, has recently regained considerable attention being the first chiral molecule detected outside of our Solar System (1). Chiral molecules are extremely important in chemistry, physics, biology, and drug discovery (2–8). Despite their variable stereoselectivity in various chemical reactions, the homochiral nature of all chiral biomolecules is a decisive prerequisite for an enormous range of cellular functions (9). While homochirality is evolutionarily advantageous, it is still unknown how the molecular single-handedness of the building blocks of the complex triinity—DNA, RNA, and proteins—came about (10). Several asymmetric processes have been experimentally tested to induce chirality in molecular systems (11–20), but those focusing on circularly polarized light (CPL) appear to us to be the most encouraging, especially given the results reported on CPL-induced molecular chirality in amino acids (21–27). Chirality transfer from photons to matter and vice versa (28–30) is not only of fundamental importance to understanding one of the most puzzling questions of the early origins of life (31, 32) but also of practical importance as a preparative method for synthesis of chiral compounds (33–36) or next-generation photonic devices (37–39).

Experiments that use CPL illumination to induce a chiral bias require prior knowledge of the chiroptical response of the targeted matter to tune desired enantiomeric excesses (ee). Circular dichroism (CD) spectroscopy, based on the difference in absorption of left and right CPL (l- and r-CPL) of chiral molecules, remains the preferred technique to study chiral discrimination, because of its straightforward operation, high accuracy, and efficiency (40). The intrinsic enantioselectivity toward CPL of a given handedness and wavelength can be then quantified by the so-called anisotropy or dissymmetry factor $g = \Delta \epsilon / \epsilon$, the ratio between the differential extinction coefficient $\Delta \epsilon$, and the extinction coefficient $\epsilon$ (41–43).

Propylene oxide (Fig. 1A), a relatively simple chiral molecule, has recently been detected in the interstellar medium toward the high-mass star-forming region Sagittarius B2(N) (1). At the time, the observations did not allow the determination of whether there is a difference between the relative abundance of the two propylene oxide enantiomers. However, high-precision polarization measurements could provide a definite answer in the future.

The spectroscopic properties of propylene oxide and its derivatives have been extensively studied in the past (44–48). Because of the minimal conformational freedom of propylene oxide, it is, moreover, an ideal model compound for theoretical method development for chiroptical spectroscopy (49–51). However, the only low-resolution experimental CD spectra of propylene oxide previously reported in the ultraviolet (UV) (46) and vacuum UV (VUV) wavelength range (44, 45) show only very weak and inconsistent features for the higher-lying electronic states with no mirror symmetry confirmation of relative intensities, signs, and band positions of the two enantiomers. These experimental data are thus inadequate for validation of theoretical approaches. We have recently refreshed the field of gas-phase CD and anisotropy spectroscopy with a new experimental setup that combines an entirely new gas cell with synchrotron radiation from the AU-CD beam line at ASTRID2, Aarhus (Denmark) (52).

Here, we report on the first experimental measurements of the anisotropy spectra of $R$-$(+)$- and $S$-$(−)$-propylene oxide along with synchrotron CD and absorption spectra in the VUV spectral region. The concerted experimental and in vacuo theoretical experiments ultimately provide structural information on the electronic and vibronic transitions as well as the oscillator and optical rotatory strengths. Quantitative models based on these newly measured anisotropy spectra revealed correlations between the $R$-$(+)$-enantio-mer–enriched propylene oxide derivative detected in the Murchison meteorite (53) and the helicity of CPL in our presolar

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cloud with far-reaching consequences in understanding the origins of biomolecular homochirality.

RESULTS

Gas-phase chiroptical spectra

The anisotropy factor $g$ as a function of wavelength was studied by recording simultaneously the CD $\Delta \varepsilon$ and absorption $\varepsilon$ spectra of each enantiomer of propylene oxide using a gas of analytes in a dedicated sample cell with a length of 500 mm at the AU-CD beam line on ASTRID2 at Aarhus University (52). Figure 1B shows the CD and associated absorption spectra of $S$-$(\cdot)$-propylene oxide along with the most dominant resolved experimental electronic and vibronic transitions highlighting the high resolution of gas-phase synchrotron CD spectroscopy. To provide accurate anisotropy data of both enantiomers, the CD (fig. S1) and absorbance spectra (fig. S2 and table S1) were measured at various gas flow rates. Individually recorded CD spectra could be scaled directly with the cell pressure during measurements to yield spectra of similar shape and magnitude. The absolute CD signal, corrected for gas density and path length, is given in units of molar CD (per molar per centimeter) often referred to as Delta epsilon units as used in Fig. 1.

CD spectroscopy of both enantiomers of propylene oxide yielded two almost perfectly mirrored spectral curves (Fig. 1C), revealing the best resolved vibrational fine structure to date (fig. S1 and table S2). $S$-$(\cdot)$-propylene oxide displays the most pronounced maxima at around $\lambda = 173$ (CD band I) and 145 nm (III) and minima at around 159 (II) and 134 nm (IV), while $R$-propylene oxide shows mirror symmetry in CD signs and relative intensities.

To compute the CD spectrum of $S$-$(\cdot)$-propylene oxide, the first 16 time-dependent density functional theory (TD-DFT) vertical excitations were convoluted with Gaussian broadening functions with full width at half maximum (FWHM) of 0.12 eV. Vertical excitation energies and rotatory strengths $R$ computed with different functionals and basis sets are summarized in table S4 and compared with former theoretical results (table S5). Best agreement with our experimental data was found using the CAM-B3LYP functional, which was tailored explicitly to treat Rydberg-type and diffuse excitations (54). In combination with the aug-cc-pVQZ basis set, the convergence for the low-lying computed excitation energies and rotatory strengths is reasonably good, while larger deviation occurs for CD bands III and IV. According to previous literature (44, 45, 50, 51, 55), the CD transitions were assigned as the Rydberg transitions from either $\pi(O)$, the lone pair orbital on the oxygen atom, or the $\sigma$-type orbital localized along the oxirane ring bonds (table S5, A and B). The often mixed $s$- and $p$-characters of involved Rydberg-type orbitals in combination with the fine structure of our experimental gas-phase CD spectra attributable to vibrational modes—not considered in our theoretical model—make definite electronic assignments of the experimental excitation energies rather challenging.

Apart from a spectral shift of CD band III, the sign and spectral position of the calculated CD bands match those of our measured spectrum (Fig. 1D), further supporting the consistency of our data. The discrepancies in the energy range of 8 to 9 eV obviously
highlight the challenges in predicting CD spectra of oxiranes, particularly where the overall signal is a result of cancelation of vibronic transitions of opposite signs with distinct vibrational structures (50, 51, 56). Our first coherent high-resolution experimental absorbance and CD data of both propylene oxide enantiomers in the U.V. spectral region, therefore, open a plethora of avenues for profound optimization of computational methods that consider vibrational modes to accurately simulate chiroptical properties.

The intensity of the anisotropy bands follows the intensity pattern of the CD spectra. Band I is the most intense feature in both CD and anisotropy spectra, followed by bands II, IV, and III (Fig. 2A). Consequently, the highest %ee can be induced in the wavelength range of 165 to 200 nm with values predicted to be about 6% for a reaction rate of 0.9999 and $\xi_{14\text{nm}} = 1.3 \times 10^{-2}$ based on the relation reported in (43) and recapped in the Supplementary Text. The predicted ee curves over the entire anisotropy wavelength range are shown as thin lines in Fig. 2A for two selected photolysis rates $\xi$ of 0.999 (99.90%) and 0.9999 (99.99%), respectively. Further anisotropy values and their calculated minimum inducible %ee by asymmetric photolysis at the extent of reaction 0.9999 at energies corresponding to the extrema of anisotropy bands I to IV are given in table S3.

**Asymmetric photochemistry at the dawn of molecular cloud evolution**

The enantiomeric enrichment of naturally occurring $l$-amino acids (57, 58) and $d$-sugar derivatives (59) found in carbonaceous meteorites offers the sole example of asymmetry outside of the biosphere and has raised the hypothesis of a possible link between astrochemical evolution and the origin of homochiral biomolecules. However, both the origin of this extraterrestrial chiral bias and the emergence of the first chiral molecule itself are still unanswered. Asymmetric photochemistry by stellar CPL is arguably the most compelling explanation behind the chiral bias found in meteorites. This idea gained support when CP radiation in the near-infrared was found in the active sites of star-forming regions of the Orion Nebula (60–62) and in NGC 6334-V (63). Considering that a primordial chiral bias originated in the interstellar medium and was at least partially incorporated into planetesimals and planets, extraterrestrial chiral molecules could have thus contributed to the chiral choice of homochiral life on Earth. Interstellar ice simulation experiments revealed a formation mechanism of racemic propylene oxide via galactic cosmic ray–driven nonequilibrium chemistry involving suprathermal oxygen at very low temperatures and its subsequent sublimation during temperature-programmed desorption at 100 K (64). While the predominant photodecomposition of gas-phase propylene oxide upon UV radiation is known to proceed via a ring opening followed by a 1,2-hydrogen shift to give propanal and to a minor extent acetone (65), our anisotropy data predict that chiral discrimination via asymmetric photolysis of racemic propylene oxide in the gas phase could lead to an enantiomeric enrichment when the photolysis is incomplete. No chiroptical data are now available for propylene oxide in the solid state, simulating the surface of interstellar dust grains. A derivative of propylene oxide has recently been detected in the ethanol extracts of the Murray meteorite (53). The enantioselective analysis revealed a large ee of about 10%, i.e., an excess of the $R$-$(+)$-enantiomer.

Under realistic astrophysical conditions, natural sources of circularly polarized radiation are spectrally broadband in the pertinent UV spectral region (60–62), and photochemical activation of multiple CD/anisotropy bands of opposite signs is expected. To investigate the net effect of asymmetric photolysis of nonmonochromatic CPL emission, integration of each wavelength interval corresponding to an anisotropy band and integration over the entire investigated wavelength range have been performed (Table 1 and fig. S3). The results show that broadband UV irradiation by stellar $l$-CPL with a constant flux in the measured...
wavelength range would induce an ee in favor of the R-enantiomer of gas-phase propylene oxide of at least 1%. The calculated net %eeR over the range of 138 to 202 nm can be thought of as a lower limit considering the reduction of UV photon flux in the far UV range (60), resulting in a weaker impact of the g bands between 132 and 164 nm compared to the most intense g band spreading from 164 to 202 nm. The most pronounced anisotropy band 1 coinciding with the dominant UV emission wavelength range of most of the stars (60) would thus dictate the handedness of UV CPL inducible enantiomeric excess (Fig. 2B).

Although we cannot exclude the impact of asymmetric solid-state photolytic reactions on the surface of icy grains, which may differ due to the influence of a surrounding polar solvent on the chiral activity of propylene oxide, we assume that its overall impact on the total inducible ee is rather limited. The lowest energy anisotropy band attributed to the nh2 → R(3s) transition is expected to be in the far UV even in the solid state where the ice matrix dominates the stellar CPL absorption with energies above 6.5 eV. Nevertheless, future studies of the influence of a surrounding water matrix on the excitation energy and the sign of CD band 1 recorded in the gas phase between 7 and 7.5 eV by a surrounding water matrix are envisaged.

**DISCUSSION**

If our Solar System was formed in a high-mass star-forming region, which is supported by the presence of short half-life radionuclides such as 60Fe and 26Al in primitive meteorites (66)—according to our anisotropy-based prediction—it may have been irradiated with left-handed CPL to result in an excess of R(+)-propylene oxide and its derivatives in the Murchison meteorite. While our current prediction of the inducible %eeR in propylene oxide should be considered as the lower estimate due to the nonlinear decline of the stellar CPL in the VUV (60), desorption-condensation cycles connecting the solid interstellar ices with the surrounding gas may have been crucial for an ee amplification of interstellar propylene oxide starting from a few percent, as shown here in the gas phase, to about 10% eeR as identified in the Murchison meteorite (53). Besides, it is a common view that not only meteoritic amino acids but also their chiral bias may have originated from their precursor structures via Streecker-type synthesis during parent body aqueous alteration (53). Note that R(+)-propylene oxide can, in two steps, lead to the proteinogenic amino acid l-alanine. The nucleophilic attack of ammonia (NH3) on the stereogenic carbon of the R(+)-propylene oxide under acidic conditions leads to S- (+)-2-amino ethanol, the reduced form of the proteinogenic amino acid l-alanine (Fig. 3). Given the evidence of ammonia in the interstellar medium, this reaction could have occurred on interstellar icy dust particles, and the resulting amino alcohols could have been subsequently embedded in comets and asteroids. During aqueous alteration of the parent body, an oxidation process might have allowed the formation of l-alanine. Thereby, the discovery of an enantiomeric excess of the R(+)-enantiomer of a propylene oxide derivative (53) could be related to the reported ee of the l-enantiomer of some meteoritic amino acids (67, 68).

Besides the substantial improvement of our CD and anisotropy spectra in terms of resolution and mirroring effect down to the far UV region, we believe that our data will stimulate future studies on establishing a unified view of the CPL scenario considering (i) the chirality properties of key biomolecules, (ii) the hypothesis that our Solar System encountered I-CPL radiation at the time of its formation, and (iii) the detection of chiral molecules in meteorites and samples returned from the asteroid sample-return missions Hayabusa2 and OSIRIS-REx.

**MATERIALS AND METHODS**

**Chemicals**

Enantiopure standards of R(+)-propylene oxide (≥98.5%) and S-(−)-propylene oxide (not specified) were purchased from Sigma-Aldrich (Germany). Both standards were of high optical purity (99% ee) and were used without any further purification. R- (+)-camphor (98%) used for calibration purposes of the gas cell before recording CD spectra was purchased from Sigma-Aldrich (Germany).

**Experimental design**

Gas-phase absorption spectra, electronic CD, and anisotropy spectra of individual enantiomers were recorded at the AU-CD beam line on ASTRID2 at Aarhus University (Denmark) (69, 70). A temperature- and pressure-controlled gas cell has been built for this purpose and is described in detail elsewhere (52). The cell is made of stainless steel with an optical path length of 500 mm and an inner diameter of 18 mm. The vapor pressure of the sample was monitored by a type 631D Baratron heated manometer (MKS Instruments). CaF2 windows used at the extremities of the gas cell connected the apparatus with the photoelastic modulator to produce the CPL at one end and a photomultiplier tube detector at the other. Heating tapes and cartridge heaters allow heating of the gas cell while maintaining a constant pressure of 1 mbar. 12 (Table 1).

**Table 1. Inducible ee values for propylene oxide.** Integrated anisotropy g bands and inducible minimum enantiomeric excess ee through asymmetric photolysis of racemic propylene oxide (PO) at the extent of reaction ξ = 0.9999 and broadband UV I-CPL with uniform flux density in the UV range.

| λ/nm  | Average g (×10−3)* | Average %eeR | Average %eeρ | Average %eeρ S−R |
|-------|--------------------|--------------|--------------|------------------|
| 164–  | 0.51               | 2.4          | 0.51         | 0.51             |
| 202   |                    |              |              |                  |
| 149–  | −0.29              | −1.4         | 0.29         | 0.29             |
| 164   | 0.08               | 0.4          | 0.08         | 0.08             |
| 138–  |                    |              |              |                  |
| 149   | 0.18               | −0.8         | 0.18         | −0.8             |
| 138   |                    |              |              |                  |

*Average anisotropy g(λ) of each active g band of S-PO. †Average anisotropy g(λ) over the entire g spectra of S-PO. ‡Average inducible ee of each active g band. §Average inducible ee over the entire g spectra.

**Fig. 3. Gas synthetic pathway from R(+)-propylene oxide to l-alanine.** First step: Epoxide opening in acidic condition. Second step: Over-oxidation of S- (+)-2-amino ethanol.
needed for earlier measurements on amino acids, but the vapor pressure of propylene oxide was high enough to allow operation of the gas cell at room temperature. However, differential heating of the windows to higher temperatures compared to the main body of the gas cell avoided local condensation of analytes. Therefore, throughout all measurements, the temperature of the CaF$_2$ windows was set to 30°C. Two all-metal valves are mounted on the main gas cell tube: one connects, via a flexible hose, to a Varian V70 turbo pump, while the other valve connects to the sample reservoir, a 25-ml single-neck round-bottom flask, connected at the center of the gas cell. This allows the pumping speed and sample inlet rate to the gas cell to be adjusted. Measured pressures increased rapidly above 0.8 mbar without sample flow, while it was kept in between 0.03 and 0.16 mbar for $R$-($+$) and $S$-($-$) propylene oxide during absorption and CD measurements.

**Gas-phase synchrotron radiation CD and anisotropy spectroscopy**

The operation of the experimental setup was confirmed through measurement of $R$-($+$)-camphor, for which the CD spectrum in the gas phase with a characteristic transition at about 300 nm is known. The absorption $\varepsilon$ and CD spectra $\Delta \varepsilon$ of each propylene oxide enantiomer were recorded simultaneously in the gas phase. The gas cell was operated under flow conditions, where the valves to the sample and the turbo pump were partially open, to ensure a continuous renewal of the gas in the cell. This was done to avoid a build-up of any photodecomposition products. We were not able to maintain constant pressure under continuous flow conditions of each enantiomer but observed a steady increase of the pressure throughout the sample scans. Comparing different sample scans with each other, however, confirmed that the increase in pressure was due to the sample itself and not from other impurities such as water or decomposition products. The sample pressure was recorded for each data point in a spectrum, and all spectra are pressure-corrected.

The anisotropy spectra $g(\lambda) = \Delta \varepsilon/\varepsilon$ could be obtained as the absorbance of the gas, $\varepsilon$, was simultaneously measured with the CD. Absorbance is calculated using the regulated high voltage applied to photodetector and the gain value for detector, which has been determined through measurement on a series of samples on both the AU-CD beamline and a calibrated photospectrometer (Evolution 300, Thermo Scientific). Approximately 10 ml of propylene oxide was added to a 25-ml pear-shaped glass container attached to the center of the gas cell by an all-metal valve. This amount of sample proved to be enough for multiple measurement runs. Before CD measurements, the sample was purged of dissolved gases by three freeze-pump-thaw cycles, and the valve was fully open to the cell to remove air or water vapor coming off the samples. The gas-phase spectra were recorded from 4.4 to 9.5 eV (280 to 130 nm). Sample vapor pressures were chosen so that the optimal optical density near 0.8 providing the best signal-to-noise ratio was obtained. Multiple baseline scans of the empty gas cell were recorded before and after each propylene oxide enantiomer. Fortunately, the gas cell was designed to quickly be taken apart, cleaned, and re-assembled as extensive white deposits within the cell and sample valve were observed. After extensive cleaning and remounting of the sample inlet for the gas cell, we pumped down the cell for several hours before running new baselines and the opposite enantiomer.

The CD spectra of propylene oxide enantiomers could be scaled directly with the cell pressure during measurements, and the absolute CD signal (corrected for gas density and path length) could be calculated. Thus, the CD spectra shown are in units of molar CD (per molar per centimeter) often referred to as Delta epsilon units. The CD spectra were mildly smoothed with a seven-point Savitzky-Golay filter.

**Computational details**

The initial molecular structure of propylene oxide was built using GaussView (version 6). As the molecule exhibits no internal degrees of freedom, solely one conformer was considered. This structure was further optimized with Gaussian16, using a combination of different methods and different basis sets. We performed a single-point energy calculation with zero-point corrected energies including frequency calculations to verify the nature of true minima. Three methods previously applied for our amino acid gas-phase spectra were evaluated: CAM-B3LYP, oB97X-D, and M06-2X.

TD-DFT calculations were completed on the optimized structure using the three functionals combined with different basis sets. Two hundred excited states were calculated for the optimized conformer. Last, the combination of the range-separated functional CAM-B3LYP method with the quadruple-$\zeta$ with polarization and diffuse functions: aug-cc-pVQZ leads to the best fit between experiment and theory. To compare the theoretical data with the spectral shapes of the experiments, the theoretical electronic CD transitions have been convoluted by means of summing rotatory strength weighted Gaussian distribution functions with FWHM of 0.12 eV.

**Supplementary Materials**

This PDF file includes:

- Supplementary Text
- Figs. S1 to S3
- Tables S1 to S5
- References

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