Probing orientation degrees of polar molecule from high-order harmonic generation: the case of partial alignment

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Abstract. Recently, we have proposed a general method to probe the whole range of degrees of orientation based on the time-frequency profile of high-order harmonic generation (HHG) but for perfectly aligned polar molecules only. In this study, we expand the previous results by considering the imperfect alignment of an ensemble of polar molecules. The “experimental” HHGs with molecular alignment distribution are simulated by numerically solving the time-dependent Schrödinger equation. The results show that, for high and moderate degrees of alignment, the proposed method is still effective and robust. For an ensemble with low degrees of alignment, the systematic error is significant due to the inaccuracy caused by the molecules aligned at large angles to the laser electric field.

1. Introduction
For polar molecules, orientation plays an important role in strong-field phenomena. Because of the necessity of the orientation information, many efforts have been made focusing on measuring the degrees of orientation. In practice, there are different ways to directly measure this variable, such as imaging the angular distribution of fragment ion after the induced-laser Coulomb explosion [1, 2], or the free-induction decay [3, 4]. Recently, some nondestructive methods adopting high-order harmonic generation (HHG), a product of the interaction between molecules and intense laser, have been proposed to probe the orientation degrees of polar molecules [5–10]. Frunker et al. [5–8] suggested using the ratio of intensities of even to the adjacent odd harmonics to infer the degrees of orientation. Then, Shi et al. [9] developed another method for probing degrees of orientation based on calibrating HHG yield emitted from strongly polar molecules. However, these methods require strict conditions for the duration and pulse shape of the laser. The laser pulse must last either many cycles [5–8], or just a few optical cycles [9]. In addition, the former method, using the even-to-odd ratio, can retrieve only the magnitude, but not the sign of the orientation degrees.
In our recent study, we have proposed a general method to probe the full-range degrees of orientation of an ensemble of polar molecules using the time-frequency analysis of HHG [10]. The method can be applied regardless of the laser shape and laser duration. However, in Ref. [10], the polar molecules are assumed to be perfectly aligned. In practice, the perfect alignment is hard to be achieved [11–13]. To date, available alignment techniques can gain high degrees of alignment, about 0.6–0.8 [2, 14], compared to 1 for perfect alignment. Even so, considering the partial alignment of molecules is meaningful.

In this paper, we examine the effectiveness of our method proposed in Ref. [10], when considering the partial alignment of polar molecules. For this purpose, we generate the “experimental” HHG spectra by solving the time-dependent Schrödinger equation (TDSE). Here, the HHGs of polar molecules are convoluted with a distribution of spatial alignment and orientation. Then, using these convoluted HHGs, we applied the method to extract the degree of orientation.

The rest of the paper is organized as follows. Section 2 presents the main points of the TDSE method for simulation of convoluted HHG. Besides, a brief review of the procedure of probing the degrees of orientation by the time-frequency profile is also presented. Section 3 demonstrates the results and discusses on the validity of the method of probing the degree of orientation for a partially aligned molecular ensemble. We conclude the paper in Section 4.

2. Theoretical background
In this section, we present the procedure for extracting the degrees of orientation using the experimental HHG from polar molecules. However, since the experimental data with various alignment and orientation degrees are not readily available, we generate “experimental” HHG by simulation using the TDSE method. Hence, we first briefly review the TDSE method for solving the HHG of polar molecules, which is presented in detail in our previous studies [10, 15–17]. We choose the CO molecule as a case of polar molecules. The atomic units with \( \hbar = m_e = e = 1 \) are used unless stated otherwise.

2.1. Simulation of HHG by the TDSE method
The presentation of the CO molecule in the molecular frame \( Oxz \) is exhibited in Fig. 1, where its permanent dipole \( \mathbf{D} \) is chosen to be parallel or antiparallel to the \( Oz \) axis. For the simulation of the HHG, the time-dependent wavefunction \( \psi(r, t) \) of the CO molecule is solved from the TDSE, which has the following form

\[
\hat{t} \frac{\partial}{\partial t} \psi(r, t) = \left( -\frac{1}{2} \nabla^2 + V_{SAE}(r) + \mathbf{r} \cdot \mathbf{E}(t) + V_p(r, t) \right) \psi(r, t),
\]

where \( \mathbf{r} \) is the coordinate of an active electron; \( V_{SAE}(r) \) is the single-active-electron (SAE) potential; \( V_p(r, t) \) is the dynamic core-electron polarization potential induced by the laser field. The construction of the SAE model and the formula of the polarization potential is given in detail in Refs. [10, 15–17]. \( \mathbf{E}(t) \) is the laser electric field whose direction makes an angle \( \theta \) with the molecular axis, \( Oz \), as shown in Fig. 1.

After getting the time-dependent wave function, the induced dipole acceleration is determined as

\[
a(t) = \frac{d^2}{dt^2} \langle \psi(t)|\mathbf{r}\psi(t)\rangle.
\]

The HHG intensity is proportional to the square of the Fourier transform of the time-dependent acceleration dipole

\[
S(\omega) \propto \left| \int_0^T \hat{\mathbf{n}} \cdot \mathbf{a}(t)e^{-i\omega t} \, dt \right|^2,
\]

where \( \mathbf{a}(t) \) is the induced dipole acceleration.
Figure 1. The molecular frame $Oxz$ of the parallel and antiparallel CO molecules whose permanent dipole $D$ is directed along the $Oz$ axis. The electric field of the laser $E(t)$ lies in the plane $Oxz$ and makes an angle $\vartheta$ with the molecular axis.

where $\omega$ is the harmonic frequency; $\tau$ is the time duration of the laser pulse; and $\hat{n}$ is a unit vector. To calculate the time-frequency profile of HHG, we perform the wavelet transform of the induced acceleration dipole as

$$S(\omega,t) = \int_0^\tau a(t')\sqrt{\omega}W(\omega(t' - t))dt',$$

where the Morlet wavelet has the form $W(x) = (1/\sqrt{\sigma})e^{ix}e^{-x^2/2\sigma^2}$ [18]. Our examination shows that the time-frequency profile is stable for $\sigma$ ranging from 5 to 30. In our calculation, $\sigma = 20$ is chosen.

So far, the above-mentioned calculation focuses only on the single-molecule response with perfect alignment and orientation. In reality, for a molecular ensemble, the molecules are randomly aligned in the space, thus washing out the molecular-structure imprint in HHG due to the average effect. Therefore, before interacting with the intense laser pulse (probe pulse), the molecules are aligned by a weak laser pulse (pump pulse) [13]. In this way, the molecular axes are preferentially located inside a cone around the polarization vector of the aligning laser with a distribution $f_s(\vartheta')$, where $\vartheta'$ is the angle between the molecular axis and the electric field of the aligning laser. In this study, we approximate the alignment distribution by an analytical formula as used in Ref. [19]

$$f_s(\vartheta') = A\frac{\alpha}{2\pi\ln\frac{\alpha+1}{\alpha-1}}\frac{1}{\alpha^2 - \cos^2\vartheta'},$$

where the parameter $\alpha$ controls the alignment distribution; and $A$ is the normalizing constant. In reality, the alignment quality is commonly characterized by the degree of alignment, calculated by $\beta = \langle \cos^2\vartheta' \rangle = \langle f_s(\vartheta')|\cos^2\vartheta'|f_s(\vartheta')\rangle$. The notation “$s$” means that this alignment distribution is symmetric about $\vartheta' = 90^\circ$.

For an ensemble of polar molecules, besides alignment, the information on the orientation is also crucial. As presented in Fig. 1, we distinguish two molecular orientations: parallel if the permanent dipole $D$ is in the positive direction of the $Oz$ axis. Vice versa, the antiparallel molecule has permanent dipole pointing in the negative direction of $Oz$. The degrees of orientation is defined as $\eta = P_u - P_d$, where $P_u$ and $P_d$ the probabilities of parallel and antiparallel molecules, respectively. Incorporating with the imperfect alignment, the distribution of molecules is

$$f(\vartheta') = P_uf_s(\vartheta' < \pi/2) + P_df_s(\vartheta' > \pi/2).$$
After the alignment and orientation process, for generating HHG, the probe laser pulse, whose polarization vector makes an angle $\theta$ with the alignment laser’s one, is irradiated on the ensemble of molecules. Transforming to the frame associated with the probe laser pulse, the molecular distribution Eq. (6) is given by

$$f_\theta(\vartheta, \phi) = f(\vartheta'(\vartheta, \phi)),$$

where $\vartheta$ is the angle between the molecular axis, i.e. $Oz$, and the vector polarization $E(t)$ of the probe laser pulse, and $\phi$ is the azimuthal angle. The total induced acceleration dipole of the molecular ensemble is calculated as

$$a(t) = \int\int a(\vartheta, \phi, t) f_\theta(\vartheta, \phi) \sin \vartheta \, d\vartheta d\phi,$$

where $a(\vartheta, \phi, t)$ is the induced acceleration dipole of a single molecule, which is simulated by Eq. (2). The convoluted HHG is calculated by Eq. (3), i.e., through the Fourier transform of the total acceleration dipole Eq. (8).

### 2.2. Method of retrieving degrees of orientation by time-frequency profile

In this subsection, we briefly present the method of retrieving degrees of orientation proposed in our recent work [10]. The method is based on the scaled time-profile of HHG, which in turn, can easily be constructed from the HHG measurements, i.e., harmonic amplitude and phase. Recently, these quantities are commonly measured by RABITT (the reconstruction of attosecond beating by interference of two-photon transitions) [20–22], or FROG-CRAB (frequency-resolved optical gating for complete reconstruction of attosecond bursts) [23, 24] methods.

To apply the method of probing degrees of orientation, we need to measure HHGs from both the ensemble in interest, and the one with random orientation $\eta = 0$. Then, their induced dipole accelerations are calculated via the inverse Fourier transform

$$a(t) \propto \int \sqrt{|S(\omega)|} e^{i\Phi(\omega)} e^{i\omega t} d\omega,$$

where $S(\omega)$ and $\Phi(\omega)$ are the harmonic intensity and harmonic phase, respectively.

After that, the time-frequency profile is obtained by the time-frequency transform, for example, the wavelet transforms in Eq. (4). We plot the time profile $S(\omega, t)$ of a selected harmonic to determine the prominent peak and its corresponding emission time $t_r$. Then the time profile $S(\omega, t_r)$ is scaled to that of the ensemble of random orientation $S_R(\omega, t_r)$. The symbol “$R$” notates random ensemble. This quantity is called scaled time-profile.

Finally, the degree of orientation is inferred from the scaled time-profile intensity by comparing it to one of the two analytical expressions

$$\frac{S(\omega, t_r)}{S_R(\omega, t_r)} = \begin{cases} (1 + \eta)^2, \\ (1 - \eta)^2. \end{cases}$$

To determine which analytical expressions Eqs. (10a) or (10b) are used, we base on the electric direction of the probe laser field at the ionization time $t_i$. Indeed, the emission peak at the instants $t_r$ in Eqs. (10a) and (10b) is caused by the recombination of the electron that is released at the ionization time $t_i$. If at the instant $t_i$, the electric field is negative, then the ionization, and also the emission, from the parallel molecules dominate the antiparallel ones. Therefore, in this case, the Eq. (10a) is utilized. Vice versa, if at the instant $t_i$, the electric field is positive, the emission from the antiparallel molecules prevails the parallel ones. As a consequence, Eq. (10b) is applied. We note that the mapping between the returning time $t_r$ and the ionization time $t_i$ can easily be obtained from the classical simulation [25, 26].
3. Results and discussion

3.1. Retrieving degrees of orientation from partially aligned polar molecules

Now, we verify the validity of the method of retrieving degrees of orientation from partially aligned CO molecules. For this purpose, firstly, we generate the “experimental” HHG by the TDSE method as presented in subsection 2.1. In this work, we use a linearly polarized laser probe pulse whose electric field $E(t)$ has the following form

$$E(t) = E_0 \sin^2 \left(\frac{\pi t}{\tau}\right) \sin(\omega_0 t + \varphi),$$

(11)

where $E_0$, $\omega_0$, and $\varphi$ are the peak amplitude, carrier frequency, and the carrier-envelope phase (CEP) of the laser, respectively; and $\tau$ is the duration of the laser pulse. Throughout the paper, we use the laser pulse with parameters of $E_0 = 0.0755$ a.u., $\omega_0 = 0.057$ a.u. (800 nm), $\varphi = \pi$, and $\tau = 6T_0$, where $T_0$ is an optical cycle. We assume that the angle between the electric directions of the pump (aligned) and the probe lasers is $\theta = 0^\circ$.

![Figure 2](image_url)

**Figure 2.** HHGs from parallel ($\eta = +1$) and antiparallel ($\eta = -1$) CO molecules with alignment degrees of $\beta = 1$ (a) and $\beta = 0.5$ (b). The laser parameters are $E_0 = 0.0755$ a.u., $\omega_0 = 0.057$ a.u. (800 nm), $\varphi = \pi$, and $\tau = 6T_0$.

Figure 2 presents the HHG spectra from ensembles of CO with perfect alignment (a) and imperfect alignment with alignment degree $\beta = 0.5$ (b). In both cases, the cutoff is about 37th order, which is consistent with the cutoff law $1.3I_p + 3.17U_p$, where $I_p$ and $U_p$ are respectively the ionization potential and the ponderomotive energy. Besides, as mentioned in our previous work [10], in the case of perfect alignment, shown in Fig. 2(a), the opposite molecules contribute to the HHGs with an insignificant difference in intensity. The same behavior is observed for the case of imperfect alignment, as exhibited in Fig. 2(b). On the other hand, the figures also show the difference in the shape of the HHG spectra in the two cases of imperfect and perfect alignments. This difference can be easily understood since the HHG from imperfectly aligned molecules is averaged over many molecular angles.

In the next step of the procedure of retrieving degrees of orientation by the time-frequency analysis, one needs to plot the time profile of a selected harmonic. In Fig. 3, the time profiles of 33rd harmonic (H33) for the two cases of alignment degrees with $\beta = 1$ (a) and $\beta = 0.5$ (b) are illustrated. In both cases, the time profile has four peaks, in which peaks P2 and P3 are the most pronounced. The figures also reveal the sensitivity of the time profile to the degrees
Figure 3. The time profile of H33 of CO molecules with alignment degrees of $\beta = 1$ (a) and $\beta = 0.5$ (b). The electric field with indicated ionization instants of electrons that converts the emissions $P_2$ and $P_3$ is presented in Fig. (c). The used laser pulse has the same parameters as in Fig. 2.

The variation of the time profile intensity with varying orientation degrees is less pronounced than the case of $\beta = 1$. This weaker sensitivity may reduce the accuracy of the method of probing orientation degrees.

Table 1. The extracted versus “input” degrees of orientation from CO ensembles with various alignment degrees $\beta$.

| $\beta$     | 1   | 0.7 | 0.5 |
|-------------|-----|-----|-----|
| “Input” $\eta$ | -1  | -0.8 | -0.5 |
| Extracted $\eta$ | -1.08 | -0.86 | -0.54 |

To continue, we calibrate the time profile at peaks $P_2$ and $P_3$ against that of the unoriented ensemble $\eta = 0$, i.e., calculate the scaled time-profile. We then apply the analytical formulae Eqs. (10a) and (10b) to obtain the degree of orientation. As indicated in Fig. 3(c), the burst $P_2$ at about $2.87T_0$ is caused by the recombination of electron ionized at $t_i(H33, P_2) = 2.34T_0$ where the electric field is negative. Therefore, Eq. (10a) is adopted. Meanwhile, the emission of
peak $P_3$ corresponds to the ionization at the instant $t_i(H33, P_3) = 2.84T_0$, where the electric field is positive. As a consequence, Eq. (10b) is utilized. We note that the relation of the emission time and ionization time is taken from the classical simulation [25, 26].

Figure 4. The extracted degrees of orientation from the CO ensemble with various alignment degrees $\beta$. The extraction uses the scaled time-profile of H33. The dashed line shows the benchmark values. The used laser pulse has the same parameters as in Fig. 2.

Figure 4 illustrates the extracted degrees of orientation by the scaled time-profile of HHG from an ensemble of CO molecules with given orientation and alignment degrees. The step of the orientation degree is set to be 0.1. For convenience, we show some data points in Table 3.1. The results for H33 show that, for moderate and high degrees of alignment, the degrees of orientation can still be retrieved from the scaled time-profile. Indeed, for perfect alignment $\beta = 1$, the average deviation between the extracted and the “input” orientation degrees is 4.7%. For partial alignment, the average deviation unsurprisingly increases. In particular, for the ensembles with $\beta = 0.6, 0.7$, and $0.8$, the average deviations are respectively 14.7%, 10.5%, and 8.6%. Since available alignment techniques can achieve a high alignment degree, about $0.6 - 0.8$ in Refs. [2, 14], our proposed method can still yield satisfactory results. Furthermore, for general linear polar molecules, we can also apply our method to probe the degree of orientation. However, we have to examine the range of the degree of alignment within which our method can get satisfactory results.

3.2. Explanation for accuracy reduction for low alignment degree
To clarify the reason for the lower accuracy of the method of probing orientation degrees by the time profile for low alignment degrees, we study insight the alignment distribution of molecules for the case of alignment angle $\theta = 0^\circ$. In perfect alignment cases, all molecules in the ensembles align along the electric field of the probe laser. However, for an ensemble with partial alignment, as shown in Fig. 5, the molecules distribute around the electric polarization of the aligned laser with angle $\vartheta$, and as a consequence, around the electric field of the probe laser with angle $\vartheta$. The lower the alignment degree is, the more weight of the larger angle $\vartheta$ is. Therefore, in the following, we consider applying the method of probing degrees of orientation for the case of single-molecule with various angles $\vartheta$.

Figure 6 presents the time profile of H33 when the CO molecule aligns along (a), and make an angle $\vartheta = 60^\circ$ with the electric field of the probe pulse (b). Figure 6(b) shows that for $\vartheta = 60^\circ$, the time profile is less sensitive to the degrees of orientation. This weaker sensitivity leads to
more error between the extracted and the “input” degrees of orientation, as shown in Fig. 7. The average deviations for \( \vartheta = 0^\circ \), 30\(^\circ\), and 60\(^\circ\) are respectively 4.7\%, 16.1\%, and 35.2\%. In the other words, with increasing the angle \( \vartheta \), the systematic error caused by the method is larger.

Figure 5. The alignment distributions for alignment degrees \( \beta = 0.5 \) and \( \beta = 0.6 \).

To unravel the reason for the worse performance of the method for large angle \( \vartheta \), we revisit the foundation of the method of probing orientation degrees by the time profile. This method based on the asymmetry of the ionization, which is caused by the different Stark shift when the permanent dipole is parallel or antiparallel to the laser field [27, 28]. As seen in Fig. 1, the permanent dipole aligns along the \( Oz \) axis, where only the \( Oz \) projection of the electric field, i.e., \( E(t) \cos \vartheta \), acts on the permanent dipole. This projection is maximum when \( \vartheta = 0^\circ \), and reduces with increasing \( \vartheta \). Therefore, the emission asymmetry of oppositely oriented molecules for a large angle \( \vartheta \) is considerably weak, leading to the lower accuracy of the method of retrieving degrees of orientation. As a result, it reduces the accuracy of the method for ensembles with low alignment degrees. However, as indicated in the previous subsection, by recent alignment techniques, a high alignment degree, about 0.6—0.8, can be achieved; thus, our proposed method can still yield reliable results.

Finally, it should be noted that the above-mentioned HHG only considers the microscopic laser-molecule response. In fact, one has to incorporate the macroscopic propagation of the laser...
The extracted degrees of orientation for molecule CO with different molecular angles $\theta = 0^\circ$, 30$^\circ$ and 60$^\circ$. The extraction uses the scaled time-profile of H33. The used laser pulse has the same parameters as in Fig. 2.

Many studies have demonstrated that a good phase matching washes out the electron long trajectories due to their strong phase dependence on the laser intensity [29, 31]. Therefore, to artificially mimic the macroscopic HHG, we truncate the long trajectories by shortening the absorbing boundary [10, 32]. Then, we apply the same extracting method but to the data simulated by removing the long trajectories. The results show, the average error when use H33 is 2.2%, lower than 4.7% when including the long trajectories. For lower harmonic orders where the long trajectories may become prominent, for example, using H28, the average errors are 3.3% and 5.2% for including and removing long trajectories, respectively. Therefore, long trajectories do not affect significantly the efficiency and robustness of the method. This consequence can be understood since the analytical equations Eqs. (10a) and (10b) are applied at specific instants corresponding to the emission time of mostly short trajectories. Therefore, the existence of the long trajectories at other emission instants does not disturb the time profile of the short trajectories.

4. Conclusion
In this paper, we have verified the validity of the method of retrieving the degrees of orientation of partially aligned polar molecules by the time-frequency analysis. The HHGs of polar molecules convoluted with various degrees of alignment and orientation are generated by the TDSE method.

We have demonstrated that for an ensemble with high and moderate degrees of alignment, our proposed method still works effectively. The average deviation is 14.7% for the ensemble with the degree of alignment of 0.6. This systematic error is acceptable since the available alignment techniques can reach higher alignment degrees. Moreover, we have also explained the reason for the reduced accuracy of the proposed method when lowering the alignment degrees. For the case of a small degree of alignment, the contribution of molecules, whose molecular axis makes a large angle to the laser electric field, becomes significant. In turn, these molecules lower the overall sensitivity of the time profile of HHG to the molecular orientation.

Besides, we also analyze the effect of macroscopic propagation, which leads to the removal of the electron long trajectories, on the accuracy of the method. The benchmark calculation shows that the long trajectories do not affect the efficiency and robustness of the method.
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