Extension of high harmonic spectroscopy in molecules by a 1300 nm laser field

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Abstract: The emerging techniques of molecular spectroscopy by high order harmonic generation have hitherto been conducted only with Ti:Sapphire lasers which are restricted to molecules with high ionization potentials. In order to gain information on the molecular structure, a broad enough range of harmonics is required. This implies using high laser intensities which would saturate the ionization of most molecular systems of interest, e.g. organic molecules. Using a laser at 1300 nm, we are able to extend the technique to molecules with relatively low ionization potentials $(\sim 11 \text{ eV})$, observing wide harmonic spectra reaching up to 60 eV. This energy range improves spatial resolution of the high harmonic spectroscopy to the point where interference minima in harmonic spectra of N₂O and C_2H_2 can be observed.

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1. Introduction

High harmonic generation (HHG) from molecules is currently being investigated as a means to track molecular dynamics with sub-femtosecond time resolution [1-10], including rearrangement of nuclei [6,7] and electrons [9,10]. These techniques rely upon measuring the broad spectrum of coherent radiation emitted by molecules aligned in space, when interacting with intense laser fields.

The key picture for understanding high harmonic generation is the three-step model consisting of: (i) tunnel ionization of a molecule, (ii) acceleration of the electron in the laser field, and (iii) recombination of the high energy electrons with the parent ion [11]. Recent theoretical [1,12] and experimental works [3,4,13,14] have established that high harmonic spectra from spatially aligned molecules encode the structure and symmetries of molecular orbitals. One of the key steps in observing these structures is controlling the angle between the molecular axes and the polarization of the driving laser field which determines the motion of the recolliding electrons. This can be achieved by non-adiabatic molecular alignment by a preceding laser pulse [15]. The use of the non-adiabatic alignment technique allows probing a sample of aligned molecules in the absence of an electromagnetic field. Then, the well-defined mapping of the time-delay between ionization and recombination onto the energy of the emitted harmonic allows the extraction of dynamical information on ultrafast structural rearrangements with ~100 attoseconds temporal resolution [6–10].

The spatial resolution of the HHG spectroscopic methods is set by the de Broglie wavelength of the recolliding electrons. Therefore, a harmonic spectrum reaching significantly more than 50 eV is required in order to record structural features arising from typical internuclear distances (in the 1 - 2 Å range). Using a standard 800 nm Ti:Sapphire laser, such harmonic spectra demand laser intensities larger than $\sim 3 \times 10^{14}$ W/cm², well above the ionization saturation intensity of most molecules. Therefore, the availability of experimental data has been limited to a few molecular systems with relatively high ionization

potentials (\geq 13 eV), typically N₂ and CO₂. This requirement also limits the capabilities of intensity-dependent studies, as the full structural contribution to the harmonic spectrum cannot be probed at the lower intensities [9,16]. More recently, the investigation of HHG in aligned molecules has been extended to organic polyatomic molecules using ~10 fs duration laser pulses: acetylene (C₂H₂), ethylene (C₂H₄) and allene (C₃H₄) [17,18]. However, the range of harmonics obtained from these molecules was very limited, with maximum photon energies of ~46 eV in C₂H₂ due to low saturation intensity ~1.5 × 10¹⁴ W/cm². Thus, a conventional 800 nm driving laser is not suitable to obtain comprehensive structural information and quantify possible effects of electron correlation and multi-electron dynamics encoded in HHG for most molecules.

The maximum photon energy in the harmonic spectrum follows the so-called cut-off law: $E_{MAX} = I_P + 3.17U_P$, where I_P is the ionization potential of the molecule and U_P is the ponderomotive energy given by: $U_P = e/m_e(E_0\lambda/4\pi c)^2$ (*e* and m_e are the charge and mass of the electron respectively, *c* is the speed of light, E_0 is the electric field amplitude and λ is the laser wavelength). Therefore, by increasing the fundamental laser wavelength we can extend the harmonic cut-off to higher photon energies whilst keeping the laser intensity below the saturation intensity. This is the approach we have adopted to extend the technique of HHG spectroscopy. However, a consequence of increasing the wavelength is that the laser cycle and hence the time the electron spends in the continuum between ionization and recombination increases in proportion to λ . As the electron spends more time in the continuum, the electronic wavepacket spreads more in the transverse direction, reducing the probability of recollision [19]. The harmonic signal scales with the laser wavelength as λ^{-5} or λ^{-6} [20–22], hence, a balance between extending the cut-off and losing harmonic efficiency must be struck.

We found in our experiments that a wavelength of 1300 nm offers a good compromise between a significant cut-off extension and adequate HHG generation efficiency. At this wavelength the maximum photon energy of the harmonic spectrum for a given laser intensity is increased by a factor of 2.6 relative to 800 nm.

2. Experimental setup

An illustration of the experimental setup is shown in Fig. 1. We used a 1 kHz Ti:Sapphire CPA laser system at 800 nm to pump an optical parametric amplifier delivering 1 mJ pulses of 40 fs at 1300 nm for high harmonic generation. Additional synchronized pulses of 1 mJ and 80 fs from the 800 nm pump were employed for non-adiabatic molecular alignment. The 800 nm beam was sent through a motorized delay stage in order to control the delay between the aligning and harmonic driving pulses, and their relative polarization angle Θ was controlled by means of a half-wave plate. The two beams were recombined by a normal incidence dichroic mirror and sent collinearly into a vacuum chamber where they were focused onto a continuous flow gas jet by a 30 cm focal length lens. The gas jet was produced by expanding the gas with a backing pressure of 2 bar through a 100 µm diameter nozzle into the chamber. The gas temperature at the interaction region was estimated as ~90 K, sufficiently cold to obtain a significant degree of molecular alignment. A telescope in the 1300 nm beam was used to compensate the chromatic aberration of the focusing lens. The intensities at the interaction region were typically 10^{14} W/cm² for the 1300 nm and 5 × 10^{13} W/cm² for the 800 nm. The latter was set in order to optimize the degree of alignment without generating any harmonics in the monitored region. The harmonics were spectrally resolved in a flat-field XUV spectrometer and focused onto a microchannel plate detector coupled with a phosphor screen. The images of the harmonic spectra were acquired with a CCD camera and transferred to a computer. Each spectrum was averaged over 10^4 laser shots typically.



Fig. 1. Experimental layout for high harmonic generation in aligned molecules.

3. Results and discussion

We examined several molecules: nitrogen (N_2) , carbon dioxide (CO_2) , acetylene (C_2H_2) , and nitrous oxide (N₂O), covering a variety of ionization potentials and electronic structures (see Table 1). The highest occupied molecular orbitals (HOMO) which characterize the dominant features of the HHG spectra are illustrated in Fig. 2 for all of the molecules investigated. These were calculated by DFT using GAMESS-UK with the 3-21G basis set. Figure 3 shows a series of harmonic spectra obtained from each molecule with 1300 nm, 40 fs pulses and intensities in the range $(1.0 - 1.7) \times 10^{14}$ W/cm² (see details in the caption). Photon energies up to 60 eV were detected in the spectra of N_2O and C_2H_2 , whereas in N_2 and CO_2 the maximum harmonic detected exceeded 80 eV at the highest intensities used. Also plotted in Fig. 3 are harmonic spectra of N_2 , CO_2 , and C_2H_2 previously recorded with 800 nm at higher intensities $(1.8 - 2.1) \times 10^{14}$ W/cm² [17,18]. The spectra acquired with 800 nm pulses have significantly lower cut-off energies. The most significant case is that of C_2H_2 , where the spectrum obtained at the ionization saturation intensity with 800 nm pulses has a cut-off at \sim 46 eV and the spectral plateau does not extend beyond \sim 40 eV, missing the region of interest for observation of interference features. In contrast, the spectrum obtained using the 1300 nm source reaches up to 60 eV with a laser intensity of only 10^{14} W/cm².

Molecule	T _{rot} (ps)	$I_{P}(eV)$	Internuclear distance (Å) HOMO symmetry	
N_2	8.4	15.6	1.1	σ_{g}	
CO_2	42.7	13.8	2.3 †	$\pi_{ m g}$	
C_2H_2	14.2	11.4	1.2 ‡	$\pi_{ m u}$	
N_2O	39.8	12.9	2.3 §	$\sim \pi_{\rm g}$	
† ‡ § between the	between between oxygen and the	outermost nit	the oxyge the carbo crogen atom.	n atoms. n atoms.	
N ₂	CO ₂		C_2H_2	N ₂ O	
0		••	~=0=0 −	• •••••••••	

Table 1. Relevant parameters for the molecules investigated in this study.

Fig. 2. Highest occupied molecular orbital (HOMO) of the molecules investigated, calculated by DFT using GAMESS-UK with the 3-21G basis set.

Molecular axis alignment is known to influence the intensity of the harmonics in a way which is characteristic of the instantaneous structure of the molecule [2,3,13,14]. The effect of non-adiabatic impulsive alignment in the molecular sample was observed as a modulation in the harmonic signal as a function of the delay between the harmonic generating (driving) pulse and the aligning pulse. These modulations correspond to revivals of the aligned state of the molecules occurring at times multiple of T_{rot} (full revivals) and $T_{rot}/2$ (half revivals), where T_{rot} is the fundamental rotational period of the molecule [15]. The delay between the driving and aligning pulses was set to correspond with a half revival at the time when the sample is maximally aligned around the aligning field. The degree of alignment, estimated by solving the time-dependent Schrödinger equation in the experimental conditions [23] was $\langle \cos^2\theta \rangle =$ 0.66 for N₂, $\langle \cos^2\theta \rangle = 0.62$ for CO₂, $\langle \cos^2\theta \rangle = 0.72$ for C₂H₂, and $\langle \cos^2\theta \rangle = 0.67$ for N₂O, where θ is the angle between the molecular axis and the aligning field. Then, a half-wave plate was used to vary the angle Θ between the polarizations of the driving and aligning fields, allowing us to measure the harmonic signal as a function of the alignment of the molecular ensemble relative to the driving field polarization. Accounting for the actual alignment distributions [23] we can estimate the most probable orientation angle of the molecules with respect to the driving field, $\langle \theta \rangle$. For $\Theta = 0^{\circ}$ and typical aligning conditions $\langle \theta \rangle$ is ~30°.

The harmonic intensity was measured as a function of the angle between the driving and aligning field polarizations for aligned samples of N₂, CO₂, C₂H₂, and N₂O (see Fig. 4). The signals recorded from the aligned ensembles are normalized against the unaligned ones. We can clearly observe that for $\Theta = 0^{\circ}$ a minimum in the normalized HHG spectrum is observed for CO₂, C₂H₂, and N₂O. Whilst this is a familiar result in CO₂ [3,4,9] and has very recently been recorded in N₂O [24] this is the first time, to our knowledge, that harmonic spectra with this feature have been observed in N₂O and C₂H₂.

In order to qualitatively interpret these new results we take the following simplified approach: The highest occupied molecular orbital in these molecules is expected to have a characteristic two-centre structure; contributions to the recombination amplitude from the two centers can interfere constructively or destructively [1] if the returning electrons have a momentum ($p = h/\lambda_{deB}$) corresponding to a de Broglie wavelength that satisfies the condition

$$\frac{n}{2}\lambda_{\rm deB} = R\cos\theta,\tag{1}$$

where *n* is an integer, *R* is the internuclear distance and θ is the angle between the internuclear axis and the laser polarization vector. For a symmetric orbital (such as the π_u HOMO of C₂H₂) the first destructive interference occurs when n = 1, i.e. when the two centers are separated by half a de Broglie wavelength in the direction of electron propagation (driving laser polarization). For the anti-symmetric π_g HOMO of CO₂ the first destructive interference is expected when the two centers are one de Broglie wavelength apart (n = 2). Note that the HOMO of N₂O does not strictly have a π_g symmetry, but this will be assumed for the purpose of the present simplified analysis as the π_g form captures the salient features of orbital geometry and symmetry relevant to the structural interferences in N₂O.

In both N₂O and CO₂, with a similar internuclear separation, the observed dip in the harmonic spectrum for $\Theta = 0^{\circ}$ is centered around 55 eV. Given that $\theta = 30^{\circ}$ is the most probable angle between the driving field and the molecular axis in this case, our data are compatible with destructive structural interference, expected around 52 eV.

The molecule of acetylene has a shorter internuclear distance than CO_2 and N_2O , however, it has a symmetric HOMO (as opposed to the anti-symmetric HOMO of CO_2 and N_2O). Therefore, the first destructive interference is expected at smaller photon energies (46 eV), in agreement with the measurement. This is the first time that evidence of interference has been reported in C_2H_2 as in earlier measurements made at 800 nm [17,18], the harmonic spectrum could not be extended beyond 40 eV, preventing the observation of the minimum.



Fig. 3. (Black) Harmonic spectra obtained from different unaligned molecular systems with 1300 nm, 40 fs pulses. N₂: 1.7×10^{14} W/cm²; CO₂: 1.4×10^{14} W/cm²; C₂H₂: 1.0×10^{14} W/cm²; (Red) Harmonic spectra obtained with 800 nm. N₂: 2.1×10^{14} W/cm²; CO₂: 2.1×10^{14} W/cm²; C₂H₂: 1.8×10^{14} W/cm². The harmonic intensity has been offset for clarity.



Fig. 4. Ratio of harmonic signal between aligned and unaligned samples of N₂, CO₂, C₂H₂, and N₂O as a function of photon energy (harmonic order) and polarization angle Θ of the aligning pulse for a driving laser pulse at 1300 nm. The laser intensities are N₂: 1.3×10^{14} W/cm²; CO₂: 1.0×10^{14} W/cm²; C₂H₂: 1.1×10^{14} W/cm²; N₂O: 1.0×10^{14} W/cm². The red arrows mark the position of the interference minima. Notice the reversed angle scale in the N₂ plot.

In contrast, the spectrum of aligned N₂ obtained at $\Theta = 0^{\circ}$ does not show any evidence of two-center interference. This result has been predicted in theoretical calculations and is due to the mixed symmetry of its HOMO [25,26]. The dependence of the harmonic intensity on the alignment angle of N₂ follows the angular dependence of its ionization yield, with a maximum at $\Theta = 0^{\circ}$ and a minimum at $\Theta = 90^{\circ}$ for all harmonic orders [3].

It must be pointed out that Eq. (1) is derived in the frame of a single active electron in the strong field approximation, that is, considering the recolliding electron wavefunction as a superposition of plane waves. In more accurate calculations, using the strong-field eikonal-Volkov approximation [27], including electron exchange correlation and taking into account

the angle dependent ionization probability, the interference minimum in CO_2 shifts to 60 eV [28], still within the range of our observed minima. Moreover, we have recently found that the minimum detected in CO_2 is the result of an interplay between the structural minimum [defined by Eq. (1)] and a *dynamical* minimum arising from interference between different ionization channels [16]. In fact, it is the suppressed contribution of the HOMO to the overall HHG signal due to the structural interference that increases the relative amplitude of additional ionization channels (lower lying orbitals, in the Hartree-Fock picture) giving rise to dynamical interferences. This interplay explains an observed oscillation of the position of the minimum in the harmonic spectrum of CO_2 around 55 eV as the laser intensity is varied [16]. It is indeed possible that the structural minima found in N₂O and C₂H₂ would also give access to the dynamics of ionization from lower lying orbitals. A systematic study of the HHG spectrum from N₂O and C₂H₂ with different laser intensities will elucidate the effect of dynamical interferences in these molecules.

4. Conclusion

Observation of spectral features characteristic of the molecular structure and dynamics is precluded by the onset of saturation effects that limit the range of harmonics that can be observed with a standard 800 nm Ti:Sapphire laser system. This has been overcome in the case of acetylene (C_2H_2) by using a 1300 nm driving field, which has enabled the study of a critical part of its harmonic spectrum for the first time. These results illustrate the power of using long wavelengths (in the 1000 nm – 1500 nm range) for HHG of molecules with relatively small ionization potentials. Many molecules of interest, e.g. organic molecules, have ionization potentials in the range 10-12 eV, therefore the proposed method makes these molecules accessible to HHG spectroscopy. Extending the driving field to even longer wavelengths is perfectly feasible and may allow HHG spectroscopy of molecules with still lower ionization potential. In this case the signal level will be reduced due to the wavelength scaling of efficiency and that may require longer data acquisition times and increased repetition rates to achieve the measurement.

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