

Molecular Recollision Interferometry in High Harmonic Generation

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Abstract. Using extreme-ultraviolet interferometry, we measure π phase shifts in high harmonics generated from transiently aligned molecules. This data directly reflects the quantum interferences in the electron wave packet due to the two-center molecular structure.

There has been considerable recent interest in using high-harmonic generation (HHG) to observe molecular structure and dynamics. Lein et. al. introduced the two-center model which explains HHG from molecules as quantum interference of harmonic emission from two distinct regions in the molecule [1,2]. The two-center model has been used to explain the changes in HHG intensity as a molecule rotates [3]. Measuring the HHG phase would further test this model, which predicts π phase shifts in the harmonic emission above certain harmonic orders. In addition, tomographic imaging of molecules must currently assume information about the HHG phase [4]. However, only the HHG intensity has been measured in published measurements of HHG from molecules to date. In this work, we directly measure the phase of HHG from aligned molecules, by observing the quantum interference between HHG from regions of aligned and isotropic molecules. We unambiguously observe a π phase shift in the harmonic emission.

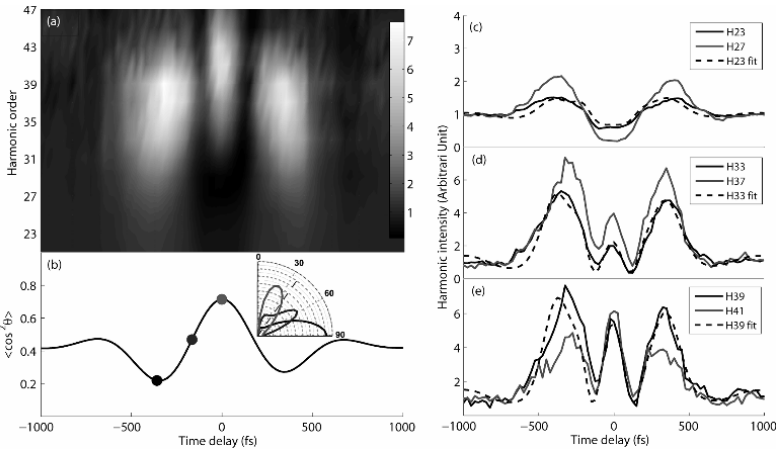


Fig. 1 (a) Measured intensity for harmonic orders 21-47 as a function of time delay. (b) Theoretical alignment parameter $\langle \cos^2\theta \rangle$. See text for the inset. (c) –(e) Lineouts of harmonic orders 23, 27, 33, 37, 39, and 41. The dashed lines show fits.

For our experiment, a pump beam transiently aligns the molecules in part of the sample, and a probe beam generates harmonics. We use two glass plates tilted at slight angles, and partially inserted into the focusing laser beam (2~3 cm after a lens of 30 cm focal length), to split the focus into two elliptical focal spots with diameters (full width at half maximum) of $\sim 80 \mu\text{m}$. The harmonics generated in the two regions then interfere in the far field. The pump beam is focused non-collinearly into the molecular gas jet at the position of one of the probe foci. Blocking the harmonic generating beam for the isotropic molecules allows for measurement of the intensity.

The net harmonic intensity for orders 21-47 as a function of time delay between pump and probe pulses for the $\frac{3}{4}$ revival in CO_2 is shown in Fig 1(a). To aid in understanding the relation between molecular alignment and harmonic intensity, Fig. 1(b) shows the alignment parameter $\langle \cos^2\theta \rangle$, while Figs. 1(c)-(e) show lineouts for each harmonic order. For harmonic orders 29 and below, the harmonic intensity follows an inverse of the alignment parameter. However, for harmonic orders 31 and above, the intensity goes through a minimum, increases when the molecules are best aligned, and goes through another minimum before increasing again. This previously unreported anomalous peak is strongly suggestive of a phase shift due to quantum interference.

The interference patterns observed when harmonics from aligned and isotropically distributed CO_2 molecules interfere in the far field confirm this phase shift. Figs. 2(a) and 2(c) show the interference patterns for harmonic orders 27 and 33 respectively. When strongly aligned, the fringes clearly shift for order 33; however, there is no such shift for order 27. The duration of the phase shift exactly corresponds to the duration of the anomalous peak for order 33 (Fig. 1(d)). The magnitude of the phase shift is determined by integrating the fringes from -100fs to 100fs and comparing with the integration of the fringes from outside this time window. Figs. 2(b) and 2(d) plot the results. The sine function fit yields HHG phase shifts of 0.035 ± 0.5 radians and 3.4 ± 0.3 radians for orders 27 and 33 respectively. We also investigated HHG intensity and phase at half revival of N_2O and N_2 molecules. For N_2O , a phase shift and anomalous peak occurred for order 31 and above during the $\frac{1}{2}$ rotational revival. However, in N_2 we observed no phase shift in the harmonics range from 19 to 31.

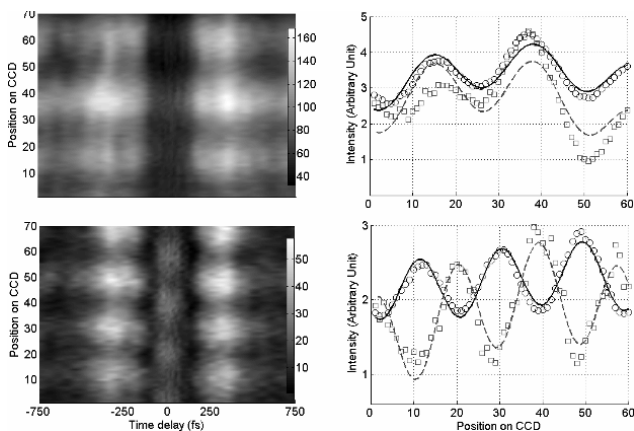


Fig. 2 (a), (c) Interference patterns as functions of time for harmonic orders 27 (a) and 33 (c). (b), (d) Intensity-scaled integrated fringes in the -100fs to 100fs interval (squares) and fit (dash line). Integrated fringes outside this time window are also shown (circles) with fit (solid line) for orders 27 (b) and 33 (d). A clear π phase shift is seen for order 33 when the molecules are aligned.

The inset of Fig. 1(b) aids in explaining the origin of the anomalous peak and the phase shift, by plotting the angular distributions for three different time delays. The dashed line corresponds to the critical alignment angle – where, according to the two-center molecular HHG model, the sign (phase) of the harmonic emission changes for order 33 due to quantum interferences in the radiating electron wave packet. When well aligned, most of the CO₂ angular distribution falls on one side of the critical angle. At the other two time delays, most of the CO₂ angular distribution falls on the other side of the critical angle. Consequently, the 33rd harmonic emission will undergo a phase reversal during a rotational alignment. For harmonic orders 29 and below, the critical angle is smaller. Under our alignment conditions, the entire molecular distribution never crosses the 29th order critical angle, and therefore no phase shift is observed. Finally, when the angular distribution is centered on the critical angle, HHG is minimized due to destructive interference. This explains the anomalous peaks in Fig. 1.

To further test the two-center interference model, we fit the CO₂ data to

$$HHG(t) = \left| \int \rho(\theta, t) A \sin(\pi R \cos \theta / \lambda) d\theta \right|^2 + C \quad (1)$$

where $\rho(\theta, t)$ is the time-dependent angular distribution calculated by numerically solving time dependent schrödinger equation, A is a normalization constant, R is the distance between the two centers, λ is the wavelength of the recolliding electron, and C is an offset constant. Fits for harmonic orders 23, 33 and 39 are shown in Figs. 1(c)-(e). In the least-square fitting process we use A, $B=R/\lambda$ and C as the fitting parameter. The data agrees well with the two-center model, strongly suggesting that quantum interferences during recombination determine the primary features of the angular modulation of HHG, and both the phase and amplitude of the returning electron wave packet are not sharply depends on the molecular orientation. From the fits, we extract the value of R/λ for each harmonic order, allowing us to determine the kinetic energy of the electron i.e. $E_k = n\hbar\omega - \delta I_p$, where n is the harmonic order and I_p is the ionization potential. A value for δ of 0 corresponds to a physical picture of the returning electron emitting harmonics as it returns to the edge of the molecular potential, whereas a value for δ of 1 corresponds to the emission occurring when the electron returns to the “bottom” of the molecular potential. We found that δ increases with increasing harmonic order, indicating a decreasing influence (dispersion) of the molecular potential on the recolliding electron for high order harmonics.

In summary, we have directly observed π phase shifts in harmonic emission from molecules for the first time. We also use the measured HHG phase and intensity to extract information about the effect of the molecular potential on the recombining electron in CO₂ [5].

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