Molecular Frame High Harmonic Dipole Amplitude and Phase Measurements

Julien B. Bertrand*[,1], Hans Jakob Wörner, David M. Villeneuve and Paul B. Corkum

[*] Joint Laboratory for Attosecond Science, National Research Council Canada and University of Ottawa, 100 Sussex Drive, Ottawa, K1A 0R6, Canada

Synopsis: We use field-free alignment to generate high-order harmonics in the molecular frame. We perform parallel measurements of the harmonic yield and the ionization probability as a function of molecular alignment. Assuming the degree of alignment achieved in the experiment, we deconvolve both the molecular frame angular ionization and harmonic yield from the alignment distribution. We further discuss the coherent retrieval of the high-order harmonic dipole in the molecule's reference frame using additional two-source [4] phase measurements.

The XUV light emitted in the High Harmonic Generation (HHG) 3-step process contains information about the interference (step 3) between a laser-driven recolliding electron wave packet (step 2) and the bound state from which it was initially tunnel-ionized (step 1)[1]. In molecules, the process can be highly sensitive to the molecular alignment. This is the basis of quantum tomographic imaging where HHG spectra were taken at different alignment with respect to the driving laser field (linearly polarized) and used to reconstruct the σ_g valence orbital of N_2 [2].

We present measurements where the ionization probability and the high harmonic intensity as a function of molecular alignment are measured in parallel, in the same experiment. For N_2 and Br_2 and CO_2 molecules, we observe that the measured angular-dependent ionization probability reflects, respectively the σ_g and π_g character of their highest occupied molecular orbitals (HOMO). The high harmonic signal modulates together with the ionization rate for the case of N_2 but anti-correlates in the case of Br_2 and CO_2.

Assuming the degree of alignment achieved in the experiment, we perform a deconvolution [3] of the ionization and harmonic signals from the distribution of molecular alignment to obtain the true molecular frame profiles. In fig. 1, we show the measured harmonic signal (a) and the ionization rate (b, scatter). After deconvolution, we obtain the molecular frame ionization probability (b, green solid curve) and harmonic yield (c). Since, the harmonic signal contains necessarily the contribution of the ionization (step 1), we perform another deconvolution of the harmonic signal but now incorporating the retrieved molecular frame ionization probability, it now represents the squared recombination dipole (d). The result is similar to the direct lab measurement (a). However, in Br_2 and CO_2, the deconvolution incorporating the ionization profile results in strong angular modulation not apparent in the raw experimental measurement.

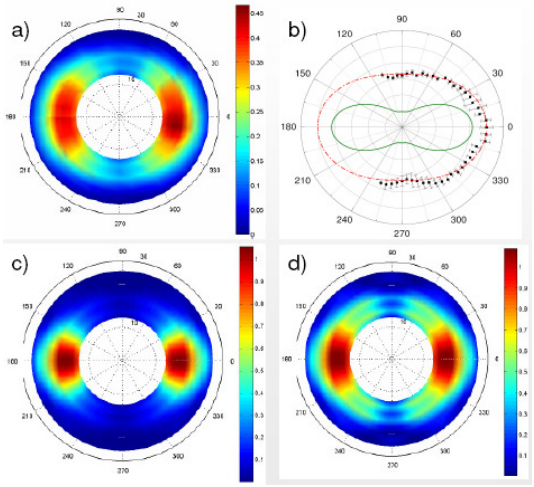

Fig. 1. a) N_2 High harmonic signal (radius: q=13 to 27, angle: Lab. molecular alignment), b) Ionization rate (scatter:exp, solid line: retrieved molecular frame ionization rate), c)Deconvoluted Squared Dipole , d) deconvoluted Squared Dipole including the measured ionization. I_{HHG}~$1.5x10^{14}$W/cm^2 and I_{Align}~$I_{HHG}/3$.

We will finally present two-source angular phase measurements in the above molecules and discuss the coherent deconvolution of the molecular frame complex dipole amplitude and phase.

References

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[1] E-mail: Julien.Bertrand@nrc.ca