# Alignment dependent spectral modulation in molecular high-order harmonic generation

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**Synopsis** By solving a 3D time-dependent Schrödinger equation with arbitrary alignment angles in non-Born-Oppenheimer approximations (NBOA), we investigated the high-order harmonic generation (HHG) process of$H_2^+$. The nuclear motion leads to redshifts in HHG spectra. The redshift unique in molecular HHG is founded to be robust in both tunneling and multiphoton ionization regimes. The redshift decreases with the increase of alignment anglesand is sensitive to the initial vibrational states.It can be used to extract the ultrafast electron-nuclear dynamics and image molecular structure.The spectral width of HHG increases in NBOA.

HHG has been used extensively in the dynamicimaging on its natural attosecond time scale as well as the generation of attosecond laser pulses. Because the nuclear motion modifies electron dynamics, it is crucial to study HHGs in NBOA.

In order to investigate the alignment dependence, we solve a 3D time-dependent Schrödinger equation (TDSE) in thevelocity gauge with an additional nuclear vibration freedom to describe the interaction. The electron motion is restricted in a 2D plane and the nuclear vibration is along the molecular axis.

Redshifts occur on the HHG in NBOA influenced by the nuclear motion. In Fig. 1, harmonic peaks in the fixed-nuclei approximation locate in the odd frequencies of the fundamental frequency exactly, while the photon energies of HHG decrease in NBOA. The wavelet analyses show that more harmonics generate on the falling part of the pulse, where electrons ionized later will get less energy than the previous laser cycledue to smaller laser amplitude [1]. This is because of the bigger ionization rate caused by the lager internuclear separation there. The laser is 500nm, $10^{14}$W/cm$^2$.

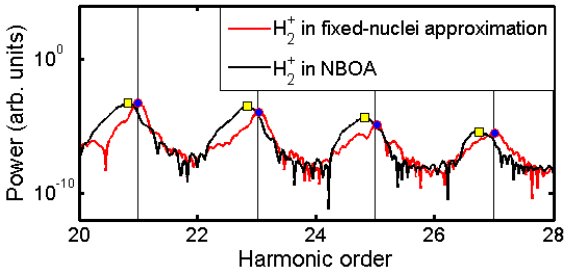


**Figure 1.** HHG spectra in fixed-nuclei approximation and NBOA respectively. [2]

The redshifts decrease as the alignment angles increase as shown in Fig. 2. The dissociation mainly comes from the dissociating first excited $2p\sigma_u$ state. When $\theta = 0°$, the coupling between the ground state $1s\sigma_g$ and $2p\sigma_u$ is the strongest. As $\theta$ increases, the transition probability to $2p\sigma_u$ state decreases since the electric field along the molecular axis becomes weak.

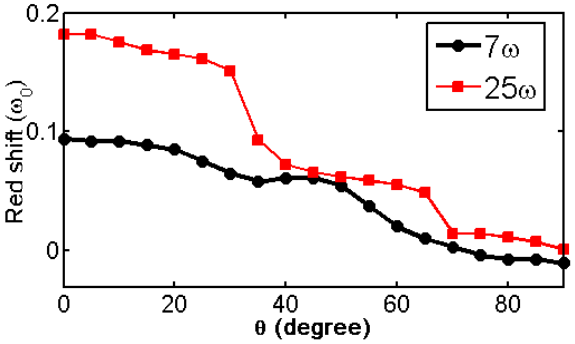


**Figure 2.** Redshifts of HH7th, 25th versus the alignment angle respectively. [2]

We also found that redshifts are sensitive to the initial vibrational state by investigating the HHGs in ν=0 and 1 respectively. The FWHMs of HHG peaks become wider in NBOA.

The redshift is a universal phenomenon during the dissociation of molecules. It can be used to detect the nuclear dynamics and probe electron-nuclei correlation in ultrashort time scale.

## References

[1] X. B. Bian and A. D. Bandrauk, 2014*Phy. Rev. Lett.***113**193901
[2] M. Z. Li, G. R. Jia and X. B. Bian2017*J. Chem. Phys.***146**084305

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