Reveal multi-channel dynamics from high-harmonic and terahertz-wave spectroscopy (HATS)

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Synopsis Synchronizing high-harmonics and terahertz-wave spectroscopy (HATS) has been demonstrated as an efficient approach in revealing the molecular structure and the underlying electron dynamics. By tracing the yields from carbon dioxide at different dual-color relative phases (DRP) and different aligned angles, we find the alignment dependent terahertz generation dose not vary with DRP, while the alignment dependent even harmonics' yields evolve strongly with DRP, indicating the participation of inner orbitals for high harmonic generation, such as the HOMO-1 and HOMO-2.

Synchronizing measurement on high-harmonics and terahertz-wave spectroscopy (HATS) has been well demonstrated as an efficient and promising tool in revealing the complex dynamics of atoms and molecules in strong fields, due to the enormous spanning magnitude of the time scale and space scale. [1] Liberated by the strong laser field, electrons may escape or recombine to its parent core. The recombined electrons contribute to the high harmonic generation (HHG), while parts of the rest, namely the directly escaping and backward scattering ones, form the drift current causing the terahertz (THz) emissions.

By varying the dual-color relative phase (DRP), the yields of both the even harmonics and the THz generation can be modulated. In Figure 1, we show the alignment-angle and DRP dependent yields of THz wave generation and even harmonics from order 20th to 34th. Every vertical line within the contour plots is normalized to its peak intensity, which means the DRP dependent modulation of one order of harmonic or THz emission is normalized at one aligned angles. The optimal DRP of different harmonics and THz are indicated by the red dots, exhibiting little variation for even harmonics at different alignment angles with harmonic order lower than 26th . However, the optimal DRP for harmonics from order 28th to 34th show significant phase jump between 0 and 90 degrees aligned angles. In Figure 2, we show the optimal DRPs of different even order harmonics (relative to the THz optimal phase). The phase jump indicates the participation of HOMO-2 in harmonic generation process around the cutoff, when the laser field is polarized along the molecular axis.

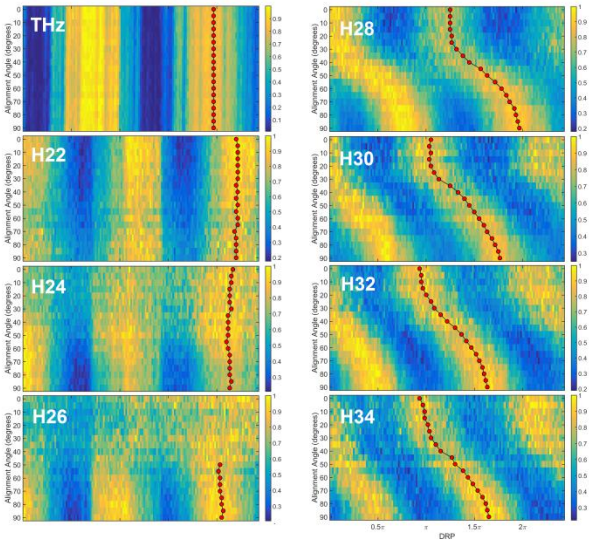

Figure 1. 2D-scanning (the alignment-angle and DRP dependence) of HATS. Each contour plot is normalized at each alignment angle (Top: 0 degree; bottom: 90 degree). The black lines with red dots indicate the optimal phase of emissions.

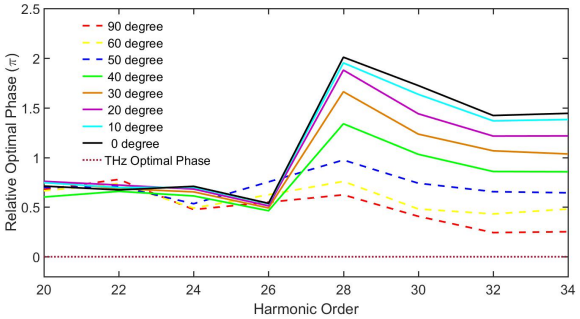

Figure 2. The optimal DRPs of different harmonic orders at different aligned angles.

References

[1] Y. Huang *et al.* 2015 Phys. Rev. Lett. **115** 123002

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