

SEMINA

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Photoelectron diffraction and molecular-frame photoelectron angular distributions

The molecular frame, i.e., the molecule-fixed coordinate system, is the natural reference frame for the study of molecules and their interaction with electromagnetic radiation or charged particles. Almost all theoretical approaches start in this molecular coordinate system. Unlike the angular distributions of electrons (and ions) in the laboratory frame, molecular-frame photoelectron angular distributions (MFPADs) often exhibit a rich structure and allow access to an unprecedented level of detailed information, such as phases of photoelectron waves [1], localization of core holes [2], and double-slit interference [3].

When interpreting MFPADs in terms of photoelectron diffraction [4,5], information on the geometric and electronic structure of the molecule can be obtained. Combining the photoelectron diffraction technique with short-pulse VUV and X-ray sources thus opens the door towards studying chemical reaction dynamics on isolated molecules with femtosecond temporal and Angstrom spatial resolution.



However, in most gas-phase experiments, the molecular frame is not directly accessible due to the random orientation of the molecules in the sample. After a brief introduction into the fundamentals of photoelectron angular distributions in the molecular frame, I will review the two most common experimental techniques for determining MFPADs (namely the angle-resolved photoelectron-photoion coincidence technique [1–5] and adiabatic or impulsive laser alignment [6]) and present various applications of both techniques that have led to new insights into the photoionization process. Finally, I will discuss first results from recent time-resolved photoelectron diffraction experiments performed at FLASH and LCLS.

References

- [1] O. Geßner et al., Phys. Rev. Lett. 88, 193002 (2002).
- [2] M. S. Schöffler et al., Science 320, 920 (2008).
- [3] D. Rolles et al., Nature 437, 711 (2005).

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- [4] A. Landers et al., Phys. Rev. Lett. 87, 013002 (2001).
- [5] B. Zimmermann et al., Nature Phys. 4, 649 (2008).
- [6] H. Stapelfeldt, T. Seideman, Rev. Mod. Phys. 75, 543 (2003).



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