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Extraction of Fixed-in-Space photoionization cross section and phase with high-order harmonic generation from aligned molecules[1]

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It has been known for over two decades that high-order harmonic generation (HHG) process occurs when an electron released earlier from an atom/molecule by an intense laser field returns to recombine with the parent ion. This relation between photo-recombination (or its time-reverse, photoionization process) and HHG has been firmly established on the quantitative level recently by the quantitative rescattering theory (QRS) [1]. According to the QRS, HHG signal can be expressed as a product of a returning electron wave packet and the *laser-free* photo-recombination differential cross section. The QRS has been carefully tested against available exact numerical solutions of the time-dependent Schroedinger equation. Here we report comparisons with recent experimental data for magnitude, phase, polarization state, and ellipticity of the emitted harmonics for aligned molecules, from which the molecular frame photoionization cross sections and phases can be probed in great details [2]. We also address the issue about the contribution from inner molecular orbitals [3,4]. Finally, we will discuss the possibility for future dynamic chemical imaging with femto-second temporal resolution on an example of probing molecular dynamics of vibrationally excited N_2O_4 [5]. In collaboration with C.D. Lin, R.R. Lucchese, T. Morishita, C. Jin.

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