

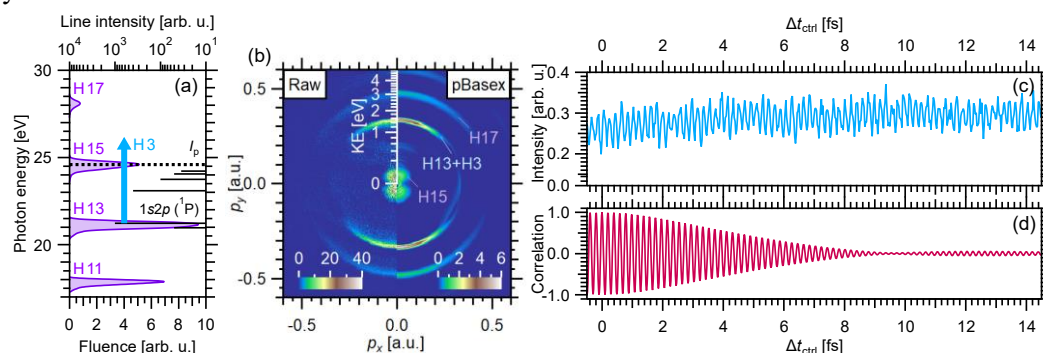
Attosecond optical and Ramsey-type interferometry using high-order harmonics

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An investigation of coherent superpositions of electronic states in atoms, i.e., electronic wavepackets (EWP), gives us an opportunity to deepen our understanding of the charge dynamics and achieve the control of quantum states. Ramsey-type interferometry, by which we monitor the quantum interference between two EWP created by a pair of phase-locked pulses, has been developed recently using extreme ultraviolet (XUV) pulses whose delay time is controlled in attosecond time scale.¹⁻³ In the present study, we demonstrate attosecond Ramsey-type interferometric measurements using ~ 20 fs XUV pulses and the split-and-delay interferometer⁴ after the generation of XUV pulses as high-order harmonics (HHs) of near-IR femtosecond laser pulses.

The spectrum of the generated XUV pulses is shown in Fig. 1(a) with the absorption lines of He. The irradiation of a pair of the 13th harmonic components in the XUV pulses generates a coherent superposition of the ground $1s^2$ state and the excited $1s2p$ state in He. A third harmonic pulse (266 nm) passing through a Mach-Zehnder interferometer is used as a probe pulse, by which He in the excited $1s2p$ state is ionized. The momentum image of the resultant photoelectrons is shown in Fig. 1(b). The yield of the electron plotted as a function of the delay time between the two XUV pulses Δt_{ctrl} in the range between -0.5 and 14.5 fs (Fig. 1(c)) exhibits an oscillation with the period of 195 as, corresponding to the excitation energy from the ground $1s^2$ state to the $1s2p$ state. The oscillation after $\Delta t_{\text{ctrl}} \sim 10$ fs can be interpreted as the Ramsey-type quantum interference because the optical interference disappears beyond the delay time of $\Delta t_{\text{ctrl}} \sim 10$ fs as in Fig. 1(d). The interferometric method we have developed in the present study can also be applied to the investigation of ultrafast dynamics of a vibrational and electronic wavepacket of molecular systems.



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