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Attosecond pump-probe of doubly excited states of helium through two-photon interferometry

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Abstract We show that correlated dynamics in wave packets of doubly excited helium can be followed in real time by a pump-probe setup based on two-photon interferometry. This approach promises to map the evolution of the two-electron wave packet onto experimentally easily accessible non-coincident single electron spectra.

Recent advances in laser sources have allowed the production of isolated extreme ultraviolet (XUV) light pulses as short as 80 attoseconds [1]. This initiated a new field, *attosecond physics*, dedicated to exploring electronic dynamics in atoms, molecules and solids in the time domain (see [2] and references therein). Most measurement protocols realized or proposed up to now rely on the interplay of a few-cycle IR pulse with a duration of a few femtoseconds and the synchronized attosecond XUV pulses produced by it. To achieve sub-fs time resolution, nonlinear effects depending on the instantaneous IR field strength (such as tunneling or streaking) are exploited. However, the presence of the non-perturbative IR field can strongly influence the dynamics. Therefore, excitation of an electronic wavepacket by an attosecond pump pulse, followed by an attosecond probe pulse to take snapshots of the ensuing electronic motion, has been dubbed the "holy grail" of attosecond physics. One obvious difficulty is the limited intensity of most current attosecond pulse sources, although rapid progress in increasing their efficiency is being made [3, 4]. Accordingly, a first pump-probe experiment for autoionizing resonances in Xe on the one-femtosecond timescale has very recently been reported [5].

This experimental perspective challenges theory to identify possibilities for monitoring non-trivial correlated electronic motion in an XUV-XUV pump-probe setup.

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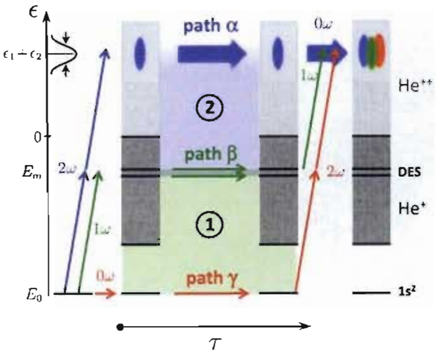

Fig. 1 Three-path interferometer for attosecond two-photon double ionization probing the coherent dynamics in DES. The three paths α, β, and γ are represented by blue, green, and red arrows, respectively (see text). Interference areas $\Delta E\tau$: Area 1 (light-green) is delineated by (quasi-) bound states and is stable under average over ε_1, ε_2. Area 2 (in light-blue) is delineated by the energy $E = \varepsilon_1 + \varepsilon_2$ of the two-electron continuum state and varies rapidly under variation of ε_1 or ε_2.

Hu and Collins [6] proposed to map out the wavepacket dynamics in *singly excited* helium. This requires a two-color pump-probe sequence and dominantly probes single-electron dynamics. Morishita *et al.* [7] showed that the correlated motion in a wavepacket among the *doubly excited* states (DES) of helium can be resolved by an XUV-XUV pump-probe scheme provided that the full six-dimensional momentum distribution is available.

We here summarize our recently proposed single-color XUV-XUV *interferometric* pump-probe protocol [8], which allows to follow the correlated two-electron motion in DES by observing only (relatively) easily accessible non-coincident observables. We exploit the interference between three two-photon double ionization pathways (see Fig. 1) in a fashion which greatly enhances the observable signal. To provide quantitatively correct results, we solve the time-dependent Schrödinger equation (TDSE) in its full dimensionality (see [9] for more details).

The attosecond two-photon pump-probe sequence (Fig. 1) can be viewed as a three-path interferometer, with the time delay τ between the pulses corresponding to the "arm length". Path α corresponds to two-photon double ionization by the pump pulse alone (see e.g. [9]), path γ is its replica induced by the probe pulse. The intermediate path β represents a proper pump-probe sequence where the first one-photon transition coherently excites a wavepacket of DES whose time evolution is then probed by double ionization by the second photon after the delay time τ. Two specific features of this three-path interferometer are key: First, path α represents a "fuzzy" slit, such that the interference phase $\Delta E\tau$ (represented by area 2 in Fig. 1) between path α and any other path rapidly varies over the Fourier width of the final energy in the continuum. Any partial trace over unobserved variables, e.g. the energy of one electron, will wipe out the interference fringes associated with path α and will result in an incoherent and τ-independent background contribution to the observed electron spectra. Second, the contribution from path γ can be seen as a delay-independent *reference wave* that the signal of interest from path β interferes with, enhancing the observable signal.

By choosing an appropriate spectral window between the two "sequential peaks" at $\omega - I_1$ and $\omega - I_2$ (where I_n is the nth ionization potential) in the single-electron

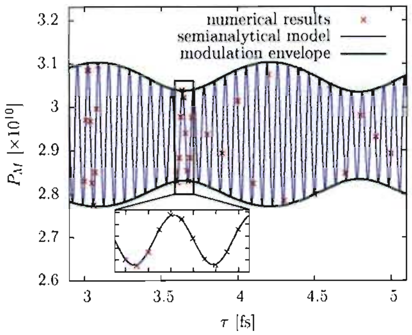

Fig. 2 Yield of restricted one-electron spectrum ($16.3\,\mathrm{eV} < \varepsilon < 28.6\,\mathrm{eV}$) resulting from double ionization of He by a pump-probe sequence of a 20 nm pump – 20 nm probe setup from the $1s^2$ singlet state as a function of delay time τ. Crosses: full numerical solution of the TDSE; blue line: semianalytical model including doubly excited resonances $|2snp^+\rangle$, $n = 2-5$, and $|2s3p^-\rangle$; green line: envelope of the modulation of the fast oscillation $A_M(\tau)$. Inset: magnification for detailed quantitative comparison.

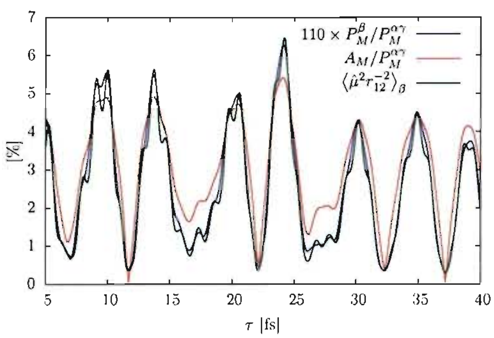

Fig. 3 Yield P_M^β from DES and modulation A_M, shown as ratios to the background yield $P_M^{\alpha\gamma} = 2\int_M |\gamma_\mathbf{K}|^2 d\mathbf{K}$ from paths α and γ, for the restricted one-electron spectrum ($17.7\,\mathrm{eV} < \varepsilon < 30.0\,\mathrm{eV}$), compared with the DES expectation value $\langle\hat{\mu}^2 r_{12}^{-2}\rangle_\beta$. The pulses ($\sin^2$ shape with 2 fs total duration, central wavelength 19 nm) coherently excite $|2snp^+\rangle$ ($n = 3-8$) with appreciable probability.

spectrum, contributions from unwanted paths[1] can be eliminated. We analyze the results using a simple semianalytical model (similar to [10, 11] used for XUV-IR setups). This exploits the fact that only the initial state $|\gamma\rangle \equiv |\psi_0\rangle$ and DES $|\beta^m\rangle \equiv |2snp^\pm\rangle$ within the pump pulse bandwidth[2] contribute to double ionization within the spectral window. Up to a global phase, the double ionization amplitude is $\langle\mathbf{K}|\psi_f\rangle = \gamma_\mathbf{K} + \sum_m e^{-i\Delta E_m \tau}\beta_\mathbf{K}^m$, where $\mathbf{K} \equiv (\mathbf{k}_1\mathbf{k}_2)$, $\Delta E_m = E_m^\beta - E_0^\gamma$, τ is the time between the pulses, and $g_\mathbf{K} = \langle\mathbf{K}|\hat{U}^{(2)}|g\rangle\langle g|\hat{U}^{(1)}|\psi_0\rangle$ (for $g = \beta, \gamma$). $\hat{U}^{(i)}$ is the time evolution operator associated with the ith pulse (1=pump, 2=probe).

Each of the mixed terms $\gamma_\mathbf{K}^*\beta_\mathbf{K}^m$ in the probability $P_\mathbf{K} = |\langle\mathbf{K}|\psi_f\rangle|^2$ oscillates with frequencies $\mathrm{Re}(\Delta E_m)$ corresponding to periods of ≈ 70 as. The superposition of several terms, $\widehat{\beta}_\mathbf{K}(\tau) = \sum_m e^{-i\Delta E_m \tau}\beta_\mathbf{K}^m$ leads to a modulation with frequencies $\mathrm{Re}(E_m - E_{m'})$ corresponding to periods on the (multi-)femtosecond scale (Fig. 2). Since $|\widehat{\beta}_\mathbf{K}|^2$ is proportional to two one-photon two-electron transition probabilities, it is three to four orders of magnitude smaller than $|\gamma_\mathbf{K}|^2$. Consequently, $\mathrm{Re}(\gamma_\mathbf{K}^*\widehat{\beta}_\mathbf{K})$ is enhanced by orders of magnitude compared to the true pump-probe signal $|\widehat{\beta}_\mathbf{K}|^2$.

[1] Such as single ionization or ejection of one electron in each pulse.

[2] We obtain the autoionizing DES as isolated states by *exterior complex scaling*, cf. [12].

The modulation amplitude is $A_M(\tau) = 4|\int_M \gamma^*_{\mathbf{K}}\widehat{\beta}_{\mathbf{K}}\mathrm{d}\mathbf{K}|$, where M is the region of final-state electron momenta integrated over. $A_M(\tau)$ is the experimentally accessible signal and agrees remarkably well with the (inaccessible) direct contribution from the DES pump-probe path, $P^\beta_M = \int_M |\widehat{\beta}_{\mathbf{K}}|^2 \mathrm{d}\mathbf{K}$ (Fig. 3).

It is now of crucial importance to identify observables which the signal $A_M \propto P^\beta_M$ can give access to. Single-photon double ionization of DES is mediated by final state correlation, i.e., to lowest order perturbation theory by the well-known two-step-one (TS1) process where one electron absorbs the photon and ejects the second electron by a collisional interaction [13]. The probability for this process is

$$P^\beta_M(\tau) \propto \int_M \mathrm{d}\mathbf{K} \langle \psi_\beta | \hat{\mu} r_{12}^{-1} | \mathbf{K}_{(0)} \rangle \langle \mathbf{K}_{(0)} | r_{12}^{-1} \hat{\mu} | \psi_\beta \rangle \approx \langle \psi_\beta | \hat{\mu}^2 r_{12}^{-2} | \psi_\beta \rangle , \qquad (1)$$

where $\mathbf{K}_{(0)}$ represents the uncorrelated final two-electron continuum state, $\hat{\mu} = p_{z,1} + p_{z,2}$ is the dipole transition operator and $|\psi_\beta\rangle$ is the DES part of the intermediate wave packet. In the second step, the closure approximation $\int_M |\mathbf{K}_0\rangle\langle\mathbf{K}_0| \mathrm{d}\mathbf{K} \approx \mathbb{1}$ has been used. Eq. 1 agrees surprisingly well with A_M (Fig. 3). Fig. 3 clearly represents signatures of the time-resolved correlation dynamics appearing in the non-coincident single-electron spectrum.

In summary, we have shown how correlated dynamics in doubly excited states of helium can be accessed by two-photon interferometry with identical attosecond pulses, without requiring coincident detection [8]. Absorption of two photons from the probe pulse provides a *reference wave* that the signal of interest interferes with and that greatly enhances the observable signal. We acknowledge support by the FWF-Austria, grants No. SFB016 and P21141-N16 (S.N. & J.B.) and by the NSF through a grant to ITAMP (J.F.). C.T. and L.A.C. acknowledge support from LANL, which is operated by LANS, LLC for the NNSA of the U.S. DOE under Contract No. DE-AC52-06NA25396. The computations used the Vienna Scientific Cluster, Institutional Computing resources at Los Alamos National Laboratory, and NSF TeraGrid resources provided by NICS and TACC under grant TG-PHY090031.

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