

Partial coherence of SASE pulses enhances time-resolution in pump-probe experiments

K. Meyer, C. Ott, P. Raith, A. Kaldun, Y. Jiang¹, A. Senftleben, M. Kurka, K. Schnorr, A. Rudenko², O. Herrwerth³, L. Foucar², K.U. Kühnel², M. Kübel³, M.F. Kling³, A. Yamada⁴, K. Motomura⁴, K. Ueda⁴, S. Duesterer⁵, R. Treusch⁵, J. Ullrich⁶, C.D. Schröter, T. Pfeifer, and R. Moshhammer

Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany

¹Shanghai Advanced Research Institute, Chinese Academy of Sciences, Pudong, Shanghai 201210, China

²Max-Planck Advanced Study Group at CFEL, 22607 Hamburg, Germany

³Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany

⁶IMRAM, Tohoku University, Sendai, Japan

⁵HASYLAB/DESY, Notkestr. 85, 22603 Hamburg, Germany

⁶Physikalisch-Technische Bundesanstalt (PTB), 38116 Braunschweig, Germany

The extreme coherence of laser light has led to fascinating developments in laser science ranging from Bose-Einstein condensates to the generation of ultra-short pulses and high-resolution spectroscopy. In contrast, free-electron laser sources based in self-amplified spontaneous emission (SASE) deliver rather unstable light pulses with temporal intensity profiles that vary from shot to shot. The chaotic pulse structure has to be considered in time-resolved experiments. Here we show that the temporal resolution can be actually enhanced by more than a factor of ten using such noisy light fields.

Our concept of noise-enhanced pump-probe spectroscopy is demonstrated for the example of SASE-type free-electron laser pulses and molecular wave-packet dynamics in a D_2^+ molecular ion, as observed in an XUV-pump – XUV-probe experiment recently performed at FLASH [1]. Although the average FEL pulse duration was 30 fs, an induced molecular dynamics with a period of only 22 ± 4 fs [1] was observed.

In our simulation [3], a set of FEL pulses is generated using the partial-coherence method [2]. We develop a model to calculate the ion yield distribution of the Coulomb-exploding doubly-ionized deuterium molecules as a function of their kinetic-energy release (KER) and the time delay τ between pump and probe pulse, starting from simple rate equations. The obtained KER-distribution is shown in Fig. 1 revealing a period of 23 ± 1 fs for the oscillating nuclear wave packet (vibrational motion of D_2^+), in excellent agreement with the experiment.

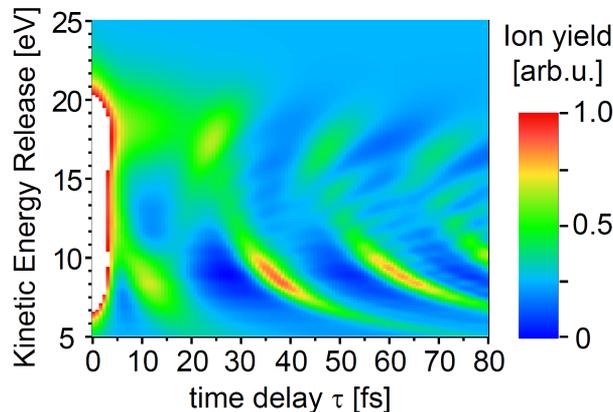


Figure 1: Calculated kinetic-energy-release-distribution as function of time delay τ for FEL pulses with average pulse length of 30 fs and 5 fs coherence length. The spectrum is an average of 2000 FEL pulses. The constant background is subtracted.

Our finding also has important consequences not only for FEL science. The results carry broad and useful implications for nonlinear time-resolved optical spectroscopy, including attosecond spectroscopy. For more details, see [3].

Autocorrelation measurements for pulse-length determination

The noisy structure of single FEL pulses also reveals itself in non-linear autocorrelation measurements performed to determine the average pulse length [4]. Using again a pump-probe scheme with split XUV pulses at 28 eV photon energy from FLASH, the yield of multiply ionized argon atoms was recorded as a function of the time delay τ . If the process is non-linear, a time-dependent signal is expected from which the pulse duration can be extracted.

In figure 2, yields of Ar^{3+} and Ar^{4+} from the same experimental run are displayed as a function of τ . One can see two apparent features: a broad pedestal and a sharp spike. Using our partial-coherence method [2] we could show that the former is related to the average pulse duration and the latter to the coherence time, which is the Fourier inverse of the spectral width. The dashed curves in the figures are autocorrelation spectra for simulated FEL pulses. For both ion yields, the same input parameters were chosen: An average pulse duration of 80 fs and a spectral width of 1.5 eV. The only difference is the assumed order of non-linearity: two for Ar^{3+} and three for Ar^{4+} , which in both cases equals the number of photons necessary in the final ionization step. The autocorrelation measurements with accompanied simulation illustrate the existence of two characteristic time scales accessible in pump-probe studies with SASE photon pulses.

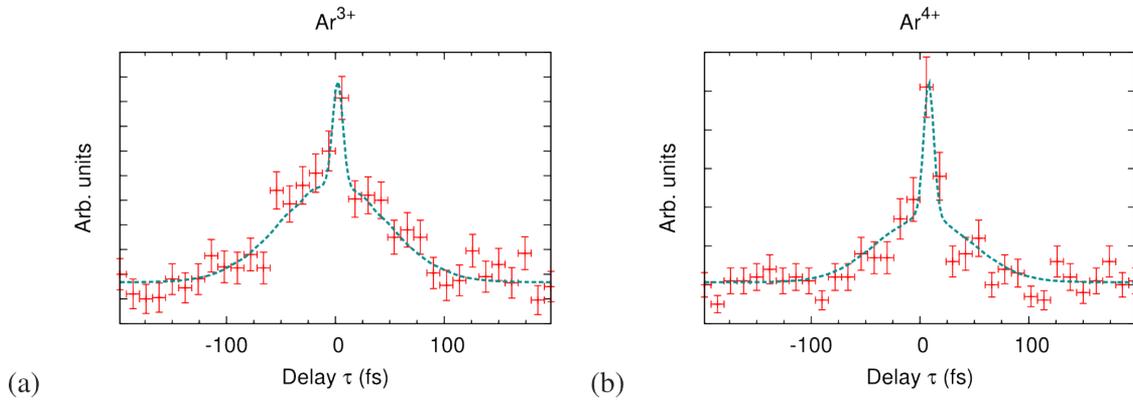


Figure 2: Autocorrelation signals obtained at a photon energy of 28 eV with an electron beam bunch charge of 0.15 nC: (a) Triple- and (b) quadruple-ionization of argon. Experimental data points are in red and traces obtained from pulses generated by our partial-coherence method are shown as dashed lines.

References

- [1] Y.H. Jiang *et al.*, Phys. Rev. A **81**, 051402(R) (2010).
- [2] T. Pfeifer, Y. Jiang, S. Düsterer, R. Moshhammer, and J. Ullrich, Opt. Lett. **35**, 3441 (2010).
- [3] K. Meyer *et al.*, Phys. Rev. Lett. **108**, 098302 (2012).
- [4] A. Senftleben *et al.*, Proc. ICOMP12/ATTO3 (in print)