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XUV-Pump/XUV-Probe Strong-field Transient Absorption on Neon at FLASH

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We present first XUV-pump/XUV-probe transient absorption spectroscopy experiments conducted at the free-electron laser FLASH. Exploiting the partial temporal coherence of the stochastic light fields, this scheme provides access to transient changes in the XUV spectra which are related to dynamics of electronic bound states on a timescale below the average pulse duration. Those transient changes manifest themselves in time-dependent changes in the spectral structure of the FEL-induced dipole response. Experimentally, we split the FEL beam into approximately equal parts with intensities of ~10^{13} W/cm² and average pulse durations of about 50 to 100 fs using the split-and-delay unit at beamline BL2 [**1**]. Both pulses, denoted by pump and probe are simultaneously detected after transmission through the neon target, and are separately resolved (offset in space) in our grating-based photon spectrometer ($E/\Delta E$ ~10^3).
Here, we study the time-dependent XUV spectral response of the neon atom and its doubly-charged ion (Ne^{2+}) at photon energies close to 50 eV. The pulse-delay (τ) dependent absorbance of the probe pulse is shown in Fig. 1(a) and exhibits resonance lines due to ^3P–^3D 2p–3d spin-orbit multiplet transitions of Ne^{2+} populated in the presence of the FEL pulse via sequential two-photon absorption. A prominent $\Delta\tau = 2.2\pm0.4$ fs "coherence feature" is imprinted on the spectral lines at $\tau = 0$, which can be explained by an enhanced coupling of these states due to overlapping temporal intensity peaks of the almost identical pump and probe replica pulses. The essential feature of reduced absorbance is reproduced by employing a non-perturbative few-level model and stochastic fields to drive the transitions (cf. Fig. 1b).
In the near future, a key application will be the precise characterization of asymmetric Fano line shapes in order to study the impact of intense FEL radiation on electron correlation and Fano interference [**2**].

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[**1**] M. Wöstmann, *et al.*, *The XUV split-and-delay unit at beamline BL2 at FLASH*, J. Phys. B **46**, 164005 (2013).
[**2**] U. Fano, *Effects of configuration interaction on intensities and phase shifts*, Phys. Rev. **124**, 1866 (1961).
[**3**] T. Pfeifer, *et al.*, *Partial-coherence method to model experimental free-electron laser pulse statistics*, Opt. Lett. **35**, 3441 (2010).

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