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# Coherence effects in gas-phase nonlinear spectroscopy with SASE-FELs

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Nonlinear spectroscopy approaches have revolutionized entire science areas from physics&chemistry to biology and medicine. Based on the interaction with several mutually coherent fields, these methods now routinely operate throughout a major part of the electromagnetic spectrum from radio to UV with applications from nuclear-magnetic-resonance(NMR) imaging to the study of chemical dynamics in small and large molecules. They rely on the availability of intense light for (at least) interacting twice (two-photon-type) with a system of interest. Intense high-frequency light of FELs thus fulfills this important requirement, fueling the vision of site-specific pumping and probing of electron dynamics in molecules. Another frontier requirement towards unfolding the full potential of such x-ray nonlinear spectroscopy is the exploitation of coherent (or even quantum-)correlated interactions with high-frequency light fields. Impressive progress on this frontier has been achieved in recent years with seeded-FEL schemes, in particular FERMI@ELETTRA.

Here, we will discuss some recent experiments in which SASE FELs, in contrast to common expectation, are used to drive coherent interactions with gas-phase samples.

A first example is the strong-field dressing of Helium atoms in a Fraunhofer-type absorption experiment, where a modification of the absorption line shape was observed as a function of intensity [1]. This demonstrates XUV-driven phase control of an atomic resonance on a time scale shorter than the (17 fs) excited-state lifetime, manifesting in the interference of the driving FEL(sub-)pulses and the emitted light from the coherently excited atoms.

As a second result, a time-resolved transient-absorption experiment in Neon atoms based on a split-and-delay approach revealed a 2-fs coherence spike near time delay zero [2]. This was enabled by mutually coherent pump and probe pulses for each SASE shot, employing the spatial coherence in the FEL beam, enabled by an existing split-and-delay unit [3]. In addition, intensity-dependent Stark shifts on XUV transitions in $Ne^{2+}$ atoms could be observed, indicating strong coupling at high frequencies.

Spectral interference structures were observed in the >1 eV probe spectrum near 50 eV transmitted through the Neon target [4]. Depending directly on the time delay between pump and probe pulses, this result is a proof-of-principle for realizing coherent pump-Stokes excitation scenarios directly with SASE FELs.

We propose a cooperative project within the scope of this workshop to drive the evolution of advanced single-shot based data-analysis methods to extract the key (phase) information from such measurements.

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[2] Ding, Rebholz, Aufleger, Hartmann, Meyer, Stooß, Magunia, Wachs, Birk, Mi, Borisova, da Costa Castanheira, Rupprecht, Loh, Attar, Gaumnitz, Roling, Butz, Zacharias, Düsterer, Treusch, Cavaletto, Ott, Pfeifer, PRL 123, 103001 (2019).
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[4] Ding, Rebholz, Aufleger, Hartmann, Stooß, Magunia, Birk, Borisova, da Costa Castanheira, Rupprecht, Mi, Gaumnitz, Loh, Roling, Butz, Zacharias, Düsterer, Treusch, Ott, Pfeifer, Faraday Discuss. DOI: 10.1039/D0FD00107D (2020).

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