# Non-Linear Pulse Propagation in Neon: The Side View

## 1) Scientific background of this proposal

It is a long-standing vision to extend experimental concepts of non-linear optics into the x-ray regime [Mukamel], where short-lived local quasi-atomic core excitations give access to interactions on atomic length and time scales. Pulse properties at modern FEL sources indeed allow investigation of nonlinear processes such as stimulated Raman scattering, amplified spontaneous emission, pulse compression, and self-induced transparency during pulse propagation [Sun,Weninger,Li2020].

Via direct observation of the transmitted beam, it has been demonstrated that non-linear X-ray pulse propagation through a medium is manifested e.g. in X-ray lasing [Rohringer] and amplification of stimulated Raman scattering [Weninger]. Advanced theoretical modelling is essential to simulate the behavior, and calculations have been based on solving the Maxwell wave equation (MWE) coupled to the time-dependent Schrödinger equation (TDSE) [Li2020]. Recently we were awarded beamtime at the SQS instrument for putting predictions of such an approach to an experimental test. In this beamtime, which has been delayed due to the pandemic, the result of the propagation through a medium will be captured using a soft X-ray spectrometer measuring in the forward direction. While much can be learned in such an approach, direct spatially resolved information about phenomena occurring along the pulse propagation cannot be achieved by probing in the forward direction only. Quasi-isotropic X-ray fluorescence is sensitively dependent on the excited medium and variations in the pulse properties. It directly monitors the population of the core hole states, Rabi oscillations are manifested in fluorescence side bands [Cavaletto, Li2020], and it can be anticipated that scattering due to radiation-driven bound-electron oscillations can be observed.

The new 1D imaging spectrometer [1D] offers the unique opportunity to measure high-resolution X-ray emission spectra with high spatial resolution perpendicularly to the incident pulse path. The investigation of pulse propagation effects was one the rationales for its construction.

## 2) Motivation for this proposal: expected results and their impact in relevant scientific area(s)

As a prototypical test case we propose a scheme that has been analyzed in some detail we use excitations at K edge of neon [Li2020]. SASE pulses are tuned to $1s^{-1}3p$ resonance of neutral Ne, and coupling to to $2p^{-1}3p$ states are considered, as well as transitions between core and valence hole states of the ion. Predictions within this five-level scheme for realistic parameters are shown in Fig. 1. The transmitted intensity (upper panel row) shows absorption of the SASE pulse at low intensities (left), due to the $1s^{-1}3p$ resonance and the ionization continuum. At higher intensities stimulated emission becomes prominent and Stokes lines corresponding to electronic excitations appear (middle and right). The X-ray fluorescence, which monitors the core hole population (middle panel row), is dominated by two peaks corresponding to $1s^{-1}3p \rightarrow 2p^{-1}3p$ and $1s^{-1} \rightarrow 2p^{-1}$ transitions. Their intensity ratio varies steeply when the beam enters the medium, as the incident SASE pulse is attenuated differently for the neutral resonance and for continuum excitations. As non-linear propagation effects develop and the pulse properties change also the probability for core hole population becomes increasingly complex. The variations match the 15-micron spatial resolution of the imaging spectrometer. The spectrometer will allow for an energy resolution that is better than the natural lifetime widths, so that any dynamics variation in peak width can be measured, and the intensity should be sufficient to resolve also side bands due to Rabi oscillations. The recently developed scheme where each SASE pulse [Li2021] is characterized using time-of-flight electron spectroscopy and ghost imaging, also gives the opportunity to measure resonant inelastic X-ray scattering spectra with a resolution better than the lifetime width. Apart from dynamics effects, fine structure is expected due to multiplet coupling in the valence excited states. The lower panel row of Fig. 1 highlights the effect of radiation-induced oscillations of the bound electrons. While this response will primarily influence the radiation in the forward direction, it may indirectly affect the observable radiation in the perpendicular direction.

We expect that this new channel of information constitutes an important step towards the understanding of, and the ability to control, the interaction between intense X-rays and matter on a fundamental level, and we foresee that it will have implications for the development of non-linear X-ray methods for studying molecular systems of chemical relevance.

**References:**

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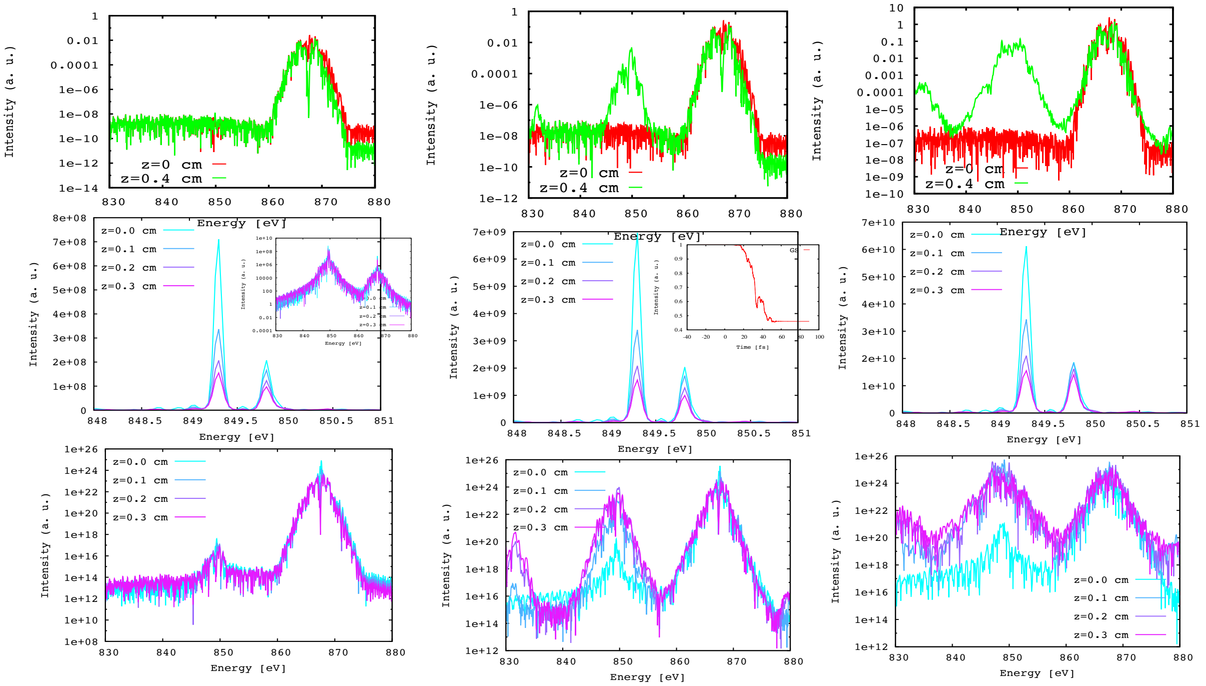

Fig. 1. Predicted observables when 20-fs SASE pulses are tuned to 867.5 eV, and peak intensity of 1.4e17 (left panel column) 1.4e18 (middle panel column), and 1.4e19 (right panel column) W/cm^2, propagate in Ne at atmospheric pressure. Transmitted intensity (upper panel row), isotropic X-ray fluorescence (middle panel row), and scattering due to the field-induced electron oscillation of the bound electrons (lower panel row) are shown.

## 3) Experimental plan

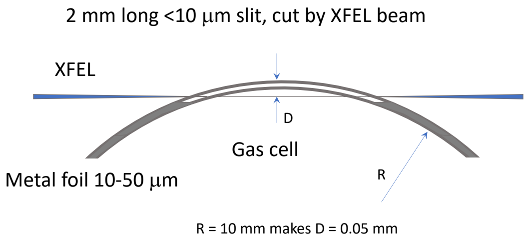

*Fig. 2. Schematics of the new gas-cell concept.*

The emission radiated in the perpendicular direction will be measured in the new 1D-imaging spectrometer. Steep pressure gradients are needed to accomplish that non-linear pulse propagation in the medium occur in sight of the spectrometer, and that self-absorption of the secondary radiation is minimized. To this end we will optimize gas-cells according to two concepts. One is a "conventional" cell where the SASE beam drills entrance and exit holes, while the fluorescence is measured through an ultrathin membrane window. For emission below the Ne edge the self-absorption is not severe, and the membrane window can be placed at some distance from the plasma to avoid rupture. The advantage with this solution is that one cell can be used to study the beam longer propagation distances than what is seen by the spectrometer, by the positioning the cell along the beam. A novel type of window-less gas-cell, dedicated for the imaging spectrometer, will also be commissioned and tested and used also for other projects. The cell will have a thin convex metal surface facing the spectrometer (Fig. 2). Using a high-precision manipulator this surface will be moved into the XFEL beam, which will cut a slit with a length adapted to the spectrometer acceptance (2 mm), and a width tailored to the cross section of the interaction region (a few micron). With this cell we estimate that pressure sufficient for investigating propagation can be reached. The limitations and opportunities of this approach will be tested.

The main experiment, which will test the predictions outlined above is straightforward. The SASE beam will be focussed in the gas-cell, while spatially resolved fluorescence spectra will be measured as a function of distance in the cell. We estimate that the count rate is sufficient for comfortable data collection, even at 10 Hz repetition rate. If this is the case, we will correlate the measured spectra with the transmitted spectra and characterize the incident pulses using the time-of-flight spectrometers [Li2021]. At the lower intensities this will enable high-resolution resonant-inelastic X-ray scattering spectra, which have so far not been measured at the Ne K edge. Should the count rate be lower than expected, we will use the full power of EuXFEL and use the maximum repetition rate to get average spectra with good statistics to test the predictions described above. The intensity dependence of the signals will be mapped in the relevant range, and also the dependence on the pulse duration.

As a second part of the beamtime, we will explore the recently established two-color option at the SQS instrument, and use the second color to seed the stimulated emission close to 850 eV, adding a dimension to the intensity mapping. The predicted difference in pulse propagation speed for on and off resonance frequencies, will result in fluorescence variations along the beam path which will be imaged in the spectrometer.

We expect that information emerging from the February beamtime, where the transmitted radiation will be measured using the “Viking” spectrometer, a conventional instrument that has been successfully used several times for similar purposes, recently at the SCS beamline. The results from this beamtime will be will guide us further in prioritizing investigation of specific parts of the multidimensional parameter space. If mechanical constraint allow, measurements of the transmitted pulse will be made simultaneously using the “Viking” spectrometer also in this project. Note that a dedicated soft X-ray spectrometer measuring in the forward will later be permanently installed at the SQS instrument, facilitating this type of experiments.

**a) Justification for the use of an X-ray free-electron laser facility and motivation for the selected instrument**

This type of experiment was one of the main motivations to set up the imaging spectrometer at the SQS instrument. Presently, this experiment cannot be done elsewhere.

**b) Justification for the number of shifts requested**

The preceding beamtime, focussing on the transmitted beam, will be helpful when we choosing parameters. It is, however, difficult to estimate the time it takes to optimize experimental conditions. The spectrometer resolution in energy and space of the spectrometer will be benchmarked, and the dependence on the incident pulse properties, such as pulse energy, duration and target density measured. From simple linear considerations we estimate that the count rate will be limited by the detector capacity to detect 100 counts/pulse. Although this would allow for fast data collection, the investigation of the spectral dependence on the parameters can only commence during one week of beamtime.

Day One: Shift 1-2: The gas-cell concepts will be tested. The conventional cell will be used for the measurements if the membrane window is resilient against rupture and contamination due to the plasma. If not, the new gas-cell concept will be implemented. The 1D spectrometer will fine adjusted to the relevant energy range

Day Two: Shift 3-4: The correlation with the time-of-flight spectrometers will be carried out, and the limits for recording conventional RIXS spectra with SASE pulses at low intensities will be explored.

Day Three: Shift 5-6: The dependence of the spectra on pulse energy, pulse duration and target density will be mapped.

Day Four: Shift 7-8: Opportunities with two-color mode will be explored

Day Five: Shift 9-10: Continued data collection.