RIXS at the Hidden Resonances of Highly Ionized Xenon: State-Resolved REXMI

1) Scientific background of this proposal

Core-level ionization is typically followed by Auger cascades that give rise to multiply ionized states. In some cases, like for single-photon Xe 3d ionization the electronic decays have been mapped in detail [Jonauskas], and transition pathways up to Xe^{6+} have been observed. With the pulse properties available at the SQS instrument (6 mJ, 1 μm^2 focus, 25 fs) multiphoton excitation is prominent, leading to charge states up to Xe^{39+} [Boll]. The ionization dynamics can be modelled using Monte Carlo simulations [Son] and it is demonstrated that resonance-enhanced X-ray multiphoton ionization (REXMI) [Rudek] is crucial to describe the charge state distribution. Moreover, the SQS pulses can be tuned to hidden resonances in multiply ionized Xe to emphasize population of specific charge states.

The novel the 1D imaging soft x-ray spectrometer [1D] to be installed at the SQS instrument is ideal to gain electronic- state-specific information on REXMI processes, and it provides a unique opportunity to investigate exotic electronic states in highly charges ions. The key idea here to spectrally resolve the fluorescence emitted after the x-ray-induced creation of high charge states as a function of photon energy and intensity, to gain insight into these fundamental nonlinear processes.

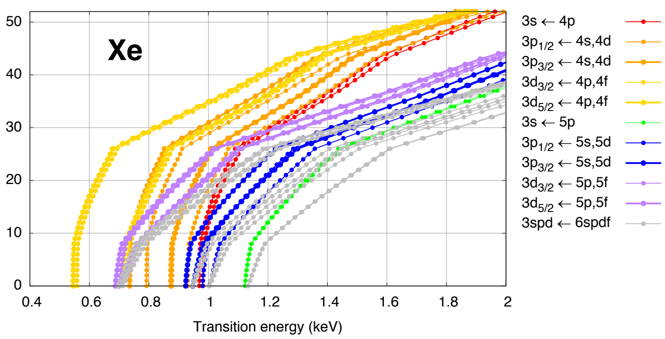

Fig. 1. A prediction of emission energies for radiative transitions to the M shell in Xe.

Technically, general measurement schemes will be established, a new gas-cell will be characterized, throughput, as well as energy and spatial resolution achievable in photon-in-photon-out experiments at the SQS instrument will be determined, and methods for correlating SASE pulses with X-ray emission spectra (XES) will be verified.

REXMI models will be tested in a state-selective manner, and RIXS processes in highly charged ions will be monitored. Both these directions are unique, and takes us in to unchartered territory.

2) Motivation for this proposal: expected results and their impact in relevant scientific area(s) (references relevant to the scientific case, not necessarily involving proposers, can be listed in this section)

XES is sensitive not only to the charge state of the ions but also to specific electronic states. In the soft X-ray range emission from highly charged ions has earlier been observed in experiments with ion traps [Kozlov] and, in collision experiments with ion beams [Bliman], and where highly charges ions impinge on a surface [Jaboski, Schweska]. The emission energies for transitions in Xe^{n+} are predicted in Fig 1. In contrast to charged particles photons are unaffected by high fields and plasma potentials, so that the electronic transitions can be monitored without distortion, and essentially background free. With an expected energy resolution in the 100-200 meV range some natural spectral linewidths will be resolved. For intensities in the 10^{18} W/cm^2 regime, dynamical Stark shifts are expected on the order of at least 500 meV for high charge states (3s/3p->4p/s,d), and even larger for the more efficiently core-screened 3d shell. Intensity scanning of the x-ray pulses thus allows determination of not only the energy but also the coupling-matrix element values predicted by multi-electron relativistic electronic-structure theory.

The observable states typically have multiple open shells, strong electron correlation and prominent relativistic effects. It can be noted that already Xe 4p vacancies in Xe^+ [Svensson] shows a breakdown of the orbital picture due to the strong interaction between $4p_{1/2}^{-1}$ and the $4d^{-2}$ continua. The 3d absorption in neutral Xe is enhanced by 3d→εf continuum resonances just above the ionization limits [Sonntag], and it is expected that bound 3d→4f excitations are found below ionization limits in positively charged Xe ions.

For non-resonant excitation a complex emission pattern is expected in the energy range covered by the spectrometer, corresponding to filling of M shell vacancies (Fig. 1). The tunability of the SASE pulses allows for Resonant Inelastic X-ray Scattering (RIXS), where specific core hole states are emphasized. This simplifies the assignment of the spectral lines in a radical way, lets us address details in REXMI dynamics, and gives access to exotic ionic states that have earlier not been investigated.

REXMI results [Boll] make it plausible that we can investigate on $3d^{-1}4f^{+1}$ excitations up to very high charge states, Xe^{24+}. A major decay path would lead to the highly correlated $4p^{-1}nl^{+1}$-derived final state excitations around 150 eV energy loss. This allows for a detailed investigation of how coupling and correlation in these states vary with charge states.

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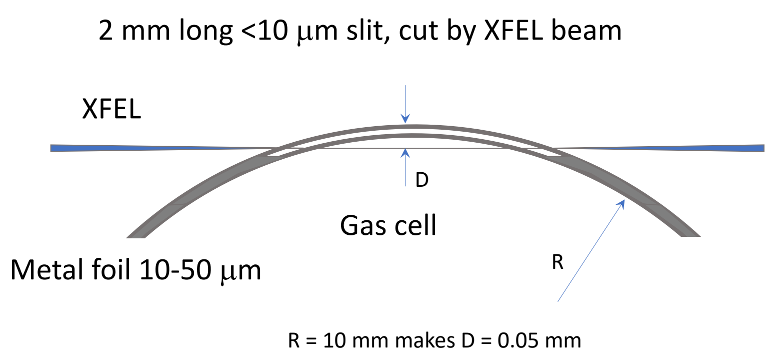

Fig. 2. Schematics of the new gas-cell concept.

3) Experimental plan

Steep pressure gradients are mandatory to accomplish that the crucial processes occur in sight of the spectrometer, and that self-absorption of the secondary radiation is minimized. Simultaneously, a high target density must be maintained. For this purpose, a novel dedicated gas-cell will be commissioned and tested. The cell will have a thin convex 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 will maintain sufficient target density for the Xe experiment, and we estimate that pressures sufficient for investigation of propagation effects in further experiments can be reached. The limitations and opportunities of this approach will be tested.

We will start by measuring overview XES spectra of (initially) neutral Xe atoms at non-resonant excitation (1.8-2.0 keV) as a function of pulse intensity. We expect to observe a multitude of spectral lines due to radiative decay of M shell vacancies [Boll] in the 500-1200 eV range. Along with the state-selective characterization of ionization pathways as a function of intensity it will also benchmark the spectrometer performance. The imaging capability will be tested by the intensity variations close to the focus. Moreover, the pressure dependence will be investigated, focussing on how attenuation due to absorption and on-set of non-linear processes vary along the beam.

Next, the SASE frequency will be tuned to specific REXMI resonances [Boll], to investigate primarily $3d^{-1}4f^{+1} \rightarrow 4p^{-1}nl^{+1}$ RIXS in various charge states. Note that the yield for radiative decay increases as Auger and Coster-Kronig channels are attenuated as the ions are stripped of outer electrons, and we can expect strong RIXS resonances also where REXMI features are absent.

For most of these experiment time integrated signals give abundant information, and the high repetition rate of the European XFEL will be an important asset. Shot-by-shot spectra cannot be measured since the detector does not detect more than 100 counts/s. To achieve high energy resolution, we will use a scheme where the SASE pulses are characterized in an upstream electron spectrometer, and correlated to the spectra measured in the X-ray spectrometer [Li]. This scheme will be used for high-resolution RIXS not only for the present experiment, but also in general at the SASE3.

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

The case is ideal for testing the capabilities of the new x-ray photon spectrometer at SQS. Unique electronic-state-selective information about REXMI dynamics and multiply-x-ray-excited exotic states in atomic ions can only be observed at FELs. Presently, this experiment cannot be done elsewhere.

b) Justification for the number of shifts requested

As a new multidimensional ballpark is entered, it is difficult to estimate the time it takes to optimize experimental conditions. The resolution in energy and space of the spectrometer will be benchmarked for the new setup geometry, and the dependence on the incident pulse properties, such as pulse energy, duration, and central SASE frequency, as well as on Xe 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: Setting up the gas-cell and investigating the accuracy to which the FEL beam can cut the slit in the metal sheet. SASE pulses will be tuned far above threshold (1.8-2.0 keV) and XES spectra will be measured as a function of pulse intensity. At low intensity the spectra will be dominated by 4p→3d emission around 520-530 eV [Boll], and upon increasing intensity, additional lines will appear in the range up to 1.1 keV. Time will be spent to fine-focus the instrument over this large energy range.

Day Two: Shift 3-4: Guided by calculated transition energies and cross sections the SASE central frequency will be tuned to specific resonances. First focus will be on the 3d→ef shape resonance of the neutral atom, and exploration of the 4f orbital collapse in ions of low charge. Correlation with the pulse-characterizing upstream electron spectrometer will be implemented.

Day Three: Shift 5-6: Only a part of the multidimensional landscape can be investigated in a first beamtime. The first focus will be the variation with charge state of Ground-State→ $3d^{-1}nf^{+1}$→$4p^{-1}nl^{+1}$ scattering.

Day Four: Shift 7-8: New type of resonances become within reach as outer shells are emptied, and we will explore also 3d→5p and 3d→4p resonances in higher charged states, as well as 3s→4p and 3p→4d resonances.

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