International Workshop on the Science with and the Instrumentation for Small Quantum Systems at the European XFEL

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Report of Working Group I on Gas Phase Instrumentation

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General

The "First International Workshop on Science with and Instrumentation for Small Quantum Systems at the European XFEL" was held in Aarhus (Denmark) from October 29th to 31st, 2008. After several plenary talks giving an introduction into the field, the details were discussed in two working groups with the aim to determine the requirements for future experiments at the European XFEL and to define the instrumentation for the SQS (Small Quantum Systems) endstation. Working Group I, coordinated by M. Meyer (Orsay) and Th. Möller (Berlin), was dedicated to techniques and instrumentations for experiments on neutral gas phase targets.

The discussion started with nine short presentations (15 min talk + 5 min discussion), which were covering the broad range of scientific interest of the community ranging from experiments on atoms and small molecules to studies of large bio-molecules, clusters and nano-particles. Principal aim of these talks was to summarize the state-of-the-art for the different experimental techniques (electron, ion and fluorescence spectroscopy) and methods (pump-probe, two-color, high field, coincidences, imaging etc.) as well as to provide the basis for defining the future needs for particular studies at the SQS Instrument at the European XFEL.

The following topics were treated in detail:

- Experiment on nano-particles (E. Rühl, Berlin)
- X-Ray femtochemistry on small molecular systems (T. Laarmann, Hamburg)
- Studies on clusters (T. Ditmire, Austin, Texas)
- X-ray fluorescence (D. Rolles, Hamburg)
- Time-resolved pump-probe experiments (J. Costello, Dublin)
- Non-linear processes in the X-ray regime (J. Marangos, London)
- Molecular alignment and orientation (H. Stapelfeldt, Aarhus)
- Velocity map imaging (M. Vrakking, Amsterdam)
- Reaction microscope/ COLTRIMS (R. Moshammer, Heidelberg)

From these presentations and, in particular, from the concluding discussion (90 min), it became clear that the general characteristics of the XFEL and the SQS Instrument will open up an extremely exciting new research field for studies in the gas phase, mainly related to the interaction with strongly bound core electrons and providing thereby a unique access to many experimental parameters, which are not in reach with other X-ray sources. But in order to take full advantage of the performances of the new machine and in order to assure efficient and successful experiments, it became also clear that some general as well as some more specific requirements for the SQS Instrument should be fulfilled, which are listed and described in the following.

1. Layout of the SQS Instrument

a) General

One fixed and very versatile endstation for experiments on neutral gas phase particles should be defined for the SQS Instrument, which shares the available space behind the SASE-3 undulator with the SCS Instrument. This endstation will host various standard equipments (outlined below) to be used by most of the users. The precise outline of the endstation will be discussed in further meetings and should be defined in close contact with the interested community.

In addition, the SQS Instrument should provide *one open beam port* for special instrumentation. Many experiments in the gas phase rely nowadays on a very sophisticated experimental geometry and detector arrangement. Non-standard experimental techniques aiming to obtain a more and more detailed, sometimes even complete picture of the complex interaction processes represent one of the qualities of gas phase instrumentation. The open beam port will allow experiments at the XFEL with these highly specialized experimental set-ups, which are developed and characterized in the home laboratory before installation at the SQS endstation.

b) Fixed SQS Endstation

In order to avoid long alignment procedure between the different experiments, i.e. in order to optimize the use of available beamtime for each group, a fixed and versatile endstation should be installed for the SQS Instrument. This station should mainly serve for more conventional experiments using high-performance analyzers for "standard" electron, ion and fluorescence spectroscopy. At minimum the experimental chamber should be equipped with

- a high-resolution time-of-flight spectrometer,
- a VMI (velocity map imaging) analyzer,
- a Thomson parabola,
- a COLTRIMS set-up
- an advanced 2D-photon pixel detector.

The chamber should be built modular and should provide the option to place detectors 'in series'. All units should be easily removable or exchangeable, without changing the general alignment of the interaction volume with the XFEL radiation. An easy access for different gas inlet systems (effusive jet, molecular beam, cluster source, electro-spray etc.) has to be integrated in the design of the chamber. Various viewports and entrance windows for optical laser light should be foreseen. Concerning the vacuum requirements, the experimental chamber should provide a vacuum of better than 10^{-9} mbar in order to enable the use of the different techniques listed above.

Existing chambers as well as the new experimental chamber under construction by the Advanced Study Group at the CFEL can serve as model systems, since they are used at different places (LCLS, FLASH etc.) in different experimental arrangements.

2. Specific Laser Systems

One major point of discussion was related to the requirements of intense optical/NIR laser systems, which are essential for various experiments and which will be used in combination with the XFEL in a *synchronized pulsed mode*. This additional equipment will enable studies on laser-prepared targets, such as e.g. two-color pump-probe experiments, time-resolved studies of fragmentation processes and experiments on laser-aligned or laser-oriented molecules, as well as the analysis of final reaction products by laser spectroscopic techniques. In order to fulfill the particular needs for the different experiments the installation of (at least) two laser systems with complementary characteristics is required.

a) Pump-Probe Laser

High pulse energies are needed for most of the application as well as variable temporal widths ranging from picoseconds (for spectroscopic studies) to the femtoseconds (for time-resolved measurements). The corresponding laser system is quite similar to the one already discussed in the Technical Design Report (TDR) for the XFEL. The main characteristics are

- temporal width of less than 50 fs,
- pulse energy of up to about 5 mJ,
- fundamental wavelength at 800nm, plus possibility to generate the 2nd and 3rd harmonics,
- option for wavelength tunability in the UV to NIR wavelength regime,
- repetition rate of 10 Hz is needed to synchronize the laser pulses to the macro-structure of the XFEL pulse distribution. In addition, a high repetition rate (>100KHz) is highly desirable in order to adapt its pulse structure as good as possible to the micro-bunch structure of the XFEL.

Furthermore, the need of a high harmonic source, which can be based on these laser systems, close to the experimental station was emphasized. Such fs-XUV photon source can serve to replace the XFEL for testing of the equipment.

b) Molecular Alignment Laser

The possibility of molecular alignment and/or orientation by an external optical/NIR laser is of large interest for some of the envisaged experiments. Two approaches to obtain molecular alignment were discussed, namely the (i) adiabatic and (ii) non-adiabatic (impulsive) alignment, which are based on different laser performances.

(i) 1-10 ns, >10 mJ, >100 kHz,

(ii) 10-100 fs.

For both lasers the repetition rate should match as good as possible the micro-bunch structure of the XFEL. Since the interaction between the laser and the molecules doesn't rely on a resonant excitation process at a precise wavelength, a fixed wavelength (probably 800nm) is needed for this type of laser.

c) X-Ray Pump-Probe

In addition, one-color pump-probe experiments using two parts of the XFEL beam was discussed as an interesting and exciting option, but the scientific advantages and possibilities as well as the experimental realization need further discussion.

3. Characteristics of the Photon Beam

a) Energy Range

The most interesting range of photon energies requested by the community covers the region between *250 and 2500 eV.* Most of the ionization thresholds and resonant excitations of the strongly bound core electrons of chemically interesting atoms, like C, N, O, and S, as well as of some rare gases, like Ne and Ar, and the transition metal L-edges are covered by these energies. Part of the discussion focused on the question whether the energy range below 800 eV could be accessed by operating the FEL at reduced electron energy. This energy region is of particular interest, since it covers the threshold of the 1s electrons of C (about 290eV), N (about 400 eV) and O (about 560 eV), which are the principal atomic constituents for many large molecules of chemical and biological interest.

b) Time Structure

In order to adapt at best the characteristics of the X-ray beam to the experimental needs for gas phase experiments, a *repetition rate as high as possible* would be highly desirable. For all experiments based on a coincident detection scheme, the high repetition rate is essential and any developments allowing to exceed the currently envisaged 30.000 pulses/second would be strongly supported by the community.

c) Polarization

Many experiments take advantage for the well-defined polarization of the XFEL radiation, sometimes in combination with the also well-defined and variable polarization of the external laser. The use of linear polarization of the XFEL is envisaged for most of the experiments, but many studies would directly benefit from the availability of circularly polarized X-ray radiation. Therefore the option to use variable polarization, i.e. changing of the relative orientation of the linear polarization vector and switching of the helicity of circularly polarized light, is also highly desirable.

d) Beam size

For most of the experiments the standard focus size of about 10 microns is sufficient, but for some applications an additional focusing optics reaching the *sub-micron focus size* (>1 micron) should be also available at the endstation. Only by squeezing the intense FEL light to these small spatial dimension will enable most of the non-linear effects to be discovered, i.e. one of the unique characteristics of the XFEL to be fully explored.

4. Diagnostics

The diagnostics of as many parameters as possible for both the *XFEL and the optical/NIR laser* is essential for almost all studies. Only the knowledge of all parameters characterizing the light pulses can assure the full analysis and correct interpretation of new phenomena.

As a minimal requirement, information about
- pulse energy,
- spectral distribution,
- arrival time

should be provided for each individual pulse within the micro-bunch structure. For two-color pump-probe experiments the relative delay between the optical and the X-ray pulse has to be monitored with a temporal resolution of better than 50 fs.

5. Follow-ups

At the end of the workshop all participants agreed that the discussions in the working group was a necessary and very useful step towards the definition of the SQS Instrument at the XFEL, but also that further exchange is needed. As a next step, in course with the XFEL Users Meeting in January 2009, three topics could be further discussed and specified:

- Laser installations for alignment and pump-probe,
- Target preparation: gas jets, cluster sources, electrospray etc.,
- Definition of the particle detectors, specification of the pixel detector and of the chamber for the fixed endstation.