Instrument for single-shot X-ray emission-spectroscopy experiments

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ABSTRACT

We present the design and the characteristics of a portable and compact photon spectrometer to be installed in free-electron-laser (FEL) beamlines for photon in – photon out experiments, in particular single-shot X-ray emission spectroscopy. The instrument is operated in the 30 – 800 eV energy range with two channels and is designed to be initially used in the LDM (Low-Density Matter) and TIMEX (TIme-resolved studies of Matter under EXtreme conditions) beamlines of the FERMI@ELETTRA FEL, covering the whole spectral range of FERMI-1 and FERMI-2 emissions. The design of the instrument is tailored to achieve high spectral resolution in the whole interval of operation, high acceptance angle and high dynamic range. These characteristics are achieved in a compact environment to give a portable instrument that may be easily installed in different experimental chambers. The design consists of an entrance slit, a grazing-incidence diffraction grating and a detector. The number of elements within the optical path is kept to a single component, to minimize the losses due to reflectivity. The grating is spherical with variable line spacing along its surface, to provide an almost flat spectral focal plane that is perpendicular to the direction of the diffracted light. The detector is a back-illuminated CCD. The spectral resolution is better than 0.2% in the 30 – 800 eV region and the acceptance angles are 10 × 17 mrad in the 30-250 eV and 5 × 17 mrad in the at 250-800 eV.

Keywords: XUV spectrometers, free electron laser, diffraction gratings, single-shot photon diagnostic, X-ray emission spectroscopy

1. INTRODUCTION

X-ray emission spectroscopy (XES) and Resonant Inelastic X-Ray Scattering (RIXS) are nowadays standard techniques in surface and solid-state investigations at third generation synchrotron radiation sources. However, the development of 4th generation free-electron-laser sources like FLASH, FERMI@Elettra, LCLS and XFEL opens new perspectives for single-shot XES and RIXS measurements both for low-density matter and for condensed matter under extreme conditions [1-4]. The availability of 10^{12} – 10^{14} photons per pulse makes possible, in principle, the collection of high-quality spectra with a single shot or with a limited number of pulses of selected intensity. This opens the way to novel applications to materials under extreme conditions or to isolated molecules and clusters both in the soft and hard X-ray regimes.

In this work we present the design of an innovative, portable and compact photon spectrometer to be used in FEL beamlines for photon in – photon out experiments, in particular for single-shot x-ray emission spectroscopy. The

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instrument is design for the 30 – 800 eV energy range, using two separate channels, and is intended to be used at the LDM and TIMEX beamlines of FERMI@ELETTRA [5-6], covering the whole spectral range of FERMI-1 and FERMI-2. Furthermore, its upper energy limit fits well with the current spectral interval of operation of FLASH in Hamburg and also with the upcoming SASE 3 beamline at XFEL. The design of the instrument gives high spectral resolution in the whole spectral interval of operation, high acceptance angle and high dynamic range. These characteristics, achieved in a compact environment, give a portable instrument that may be easily installed in different experimental chambers.

2. OPTICAL DESIGN

The optical design is itself well established, consisting of an entrance slit, a diffraction grating working in grazing incidence and a detector. The number of elements within the optical path is kept to one component, namely the grating, to minimize the losses due to reflectivity. The grating will be spherical with variable line spacing along its surface, to provide an almost flat spectral focal plane that is approximately perpendicular to the direction of the diffracted light [6]. A schematic of the diffraction geometry is presented in Fig. 1.

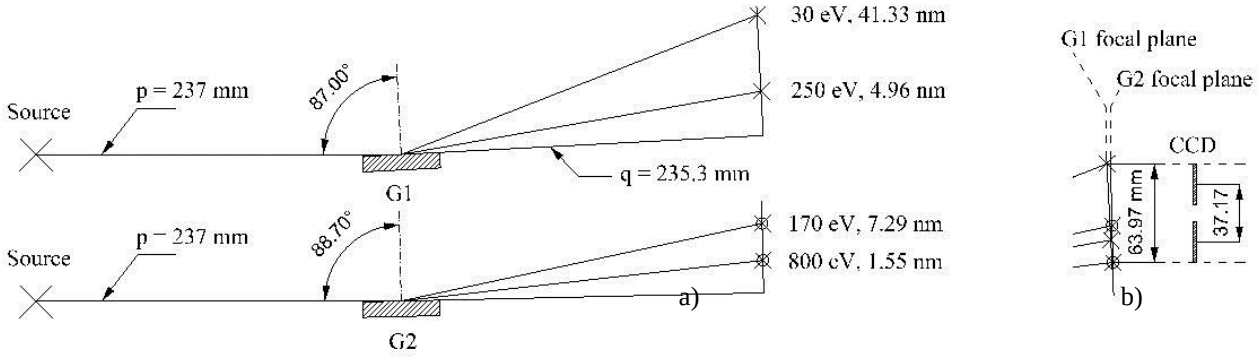

Figure 1. a) Schematic of the grating operating geometry. Source indicates the ideal position of a point like source illuminating the gratings. b) Detail of the focal plane: the two spectral curves (for the G1 and G2 gratings) are superimposed. The length of the two overlying spectral range is about 64 mm, considering the detector length of 26.8 mm, the minimum detector translation is about 37 mm.

The gratings are commercially available optical elements, able to give in a compact design a spectral resolution better than 0.2% in the 30 – 800 eV region, as shown in Fig. 2 for the two separate gratings, and acceptance angles as high as 10 mrad. They have already been proved in FEL beamlines for the monitoring of the FEL harmonics at FLASH [7, 8].

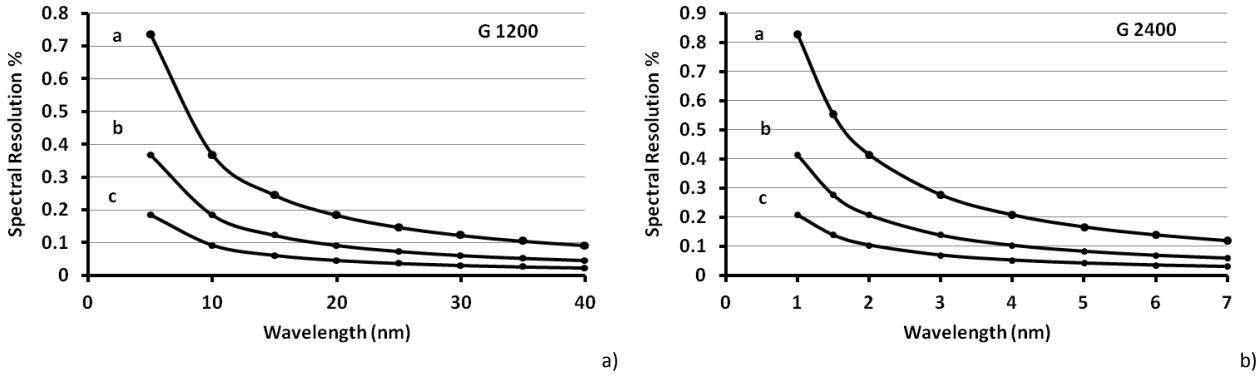

Figure 2. Theoretical spectral resolution calculated for three different slit aperture: the curves a, b, c refer to a slit aperture of respectively 200, 100 and 50 um. The panel a) is for the grating G1200, panel b) for the grating G2400.

As a detector, a back-illuminated CCD camera with high sensitivity in the extreme ultraviolet will be used. At present, cameras with small pixel size (15-20 um) and large format (>1 Kpixels) are available, to achieve high spectral resolution on a broad spectral region. Furthermore, the silicon quantum yield (i.e. the electrons generated by each interacting

photon) is in the 8 (30 eV) – 220 (800 eV) range, that is higher than the read-out noise that is normally provided in such cameras. Consequently, it can be assumed that the CCD is operated in the almost single-photon counting regime, being sensitive to the single interacting photon. The measured quantum efficiency of XUV-enhanced CCDs is in the range from 0.4 at 30 eV to 0.8 at 800 eV [9]. The dynamic range may be as high as 30000 at 30 eV and 2000 at 800 eV. Since the size of available CCDs is smaller than the length of the focal curve, the detector is mounted on a linear translator and moved along the focal curve to acquire the spectral interval of interest. The proper combination of flat-field gratings and CCD detector gives the extended spectral region of operation, the high spectral resolution, the high sensitivity and the high dynamic range.

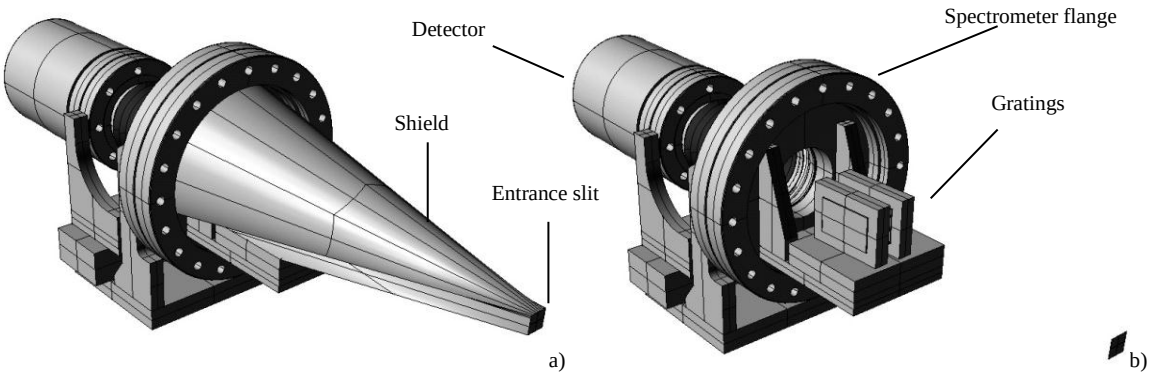

Figure 3. 3D schematic drawings of the spectrometer mounted on a flange that can be directly interfaced with the experimental chambers of LDM and TIMEX. The detector is mounted on a linear translation stage to be moved along the focal curve and is connected to the interface flange through a bellow. The internal part is completely shielded against diffused light. An external pumping system has to be provided to guarantee ultra-high-vacuum operations.

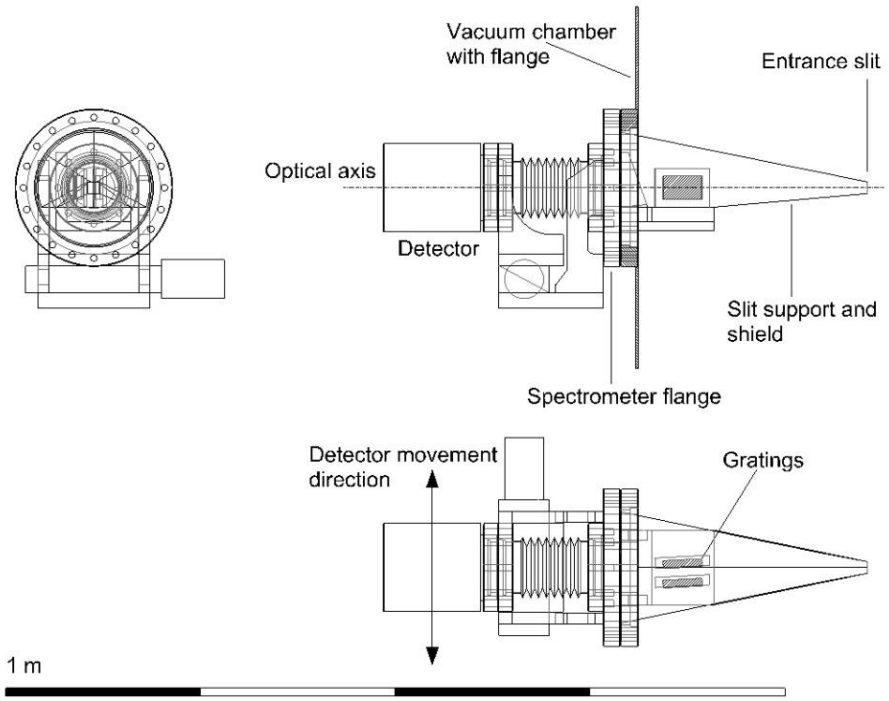

Figure 4. Schematic of the instrument. The three projections are visible and a length bar is added to give the correct scale of the instrument. The global size is about 700 mm × 250 mm × 250 mm.

The single-shot operation requires to collect a number of photons as high as possible. This is achieved by maximizing the collecting angle, putting the entrance slit close to the sample. E.g., a 100-um-wide slit placed at 20 mm from the sample gives a collection angle of 5 mrad in the spectral direction. Obviously, larger collection angles are possible, either by opening the slit either by putting the slit closer to the sample, up to the maximum aperture angle that is accepted by the grating. In the perpendicular direction, the collection angle is limited by the size of the detector and may be as high as 20 mrad. Furthermore, the instrument has to be portable in order to be installed it on different experimental chambers, namely LDM and TIMEX for the present application case, but also other FEL beamlines. These two main requirements, i.e. the portability and the needing to put the slit close to the sample, brought us to propose the realization of the prototype schematically shown in Fig. 2 and Fig. 3. The main instrumental parameters are summarized in Tab. 1.

Table 1. Instrumental parameters.

Global features		
Energy coverage	30 – 800, 2 channels	eV
Flange connection	CF160	
Input arm (entrance slit- grating distance)	237	mm
Accepted angle (G1, G2)	10 - 5	mrad
Grating G1 (G1200)		
Resolution	< 0.2% (slit 100 um, 10- 40 nm)	
Peak efficiency	18 % @ 110 eV	
Groove density	1200	mm^{-1}
Incidence angle	87	deg
Energy coverage	30 – 250	eV
Grating G2 (G2400)		
Resolution	< 0.2% (slit 100 um, 2- 7 nm)	
Groove density	2400	mm^{-1}
Peak efficiency	2 % @ 410 eV	
Incidence angle	88.65	deg
Energy coverage	170 – 800	eV
Detector		
format	1300 × 400	
pixel size	20 × 20	um
minimum detector translation to cover the full spectral band	38	mm
Full well capacity	300	Ke^-
Read out noise	3.5 (@100kHz)	e^-

3. APPLICATION EXAMPLES

3.1. Atomic, molecular and cluster physics

The use of XES and RIXS techniques is still limited in atomic and molecular physics, the main limiting factors being the low density of the sample and the small solid angle of collection for photons. Nevertheless new projects are currently developed at the new and future 3rd generation machines (MAX-IV [10], SLS [11, 12]), taking advantage of the improvement in beamline and spectrometer performances. At the same time, it is continuously increasing the complexity of systems whose electronic structure can be investigate in the gas with inner-shell photoexcitation and photoionization techniques. It is now possible to perform experiments on a wide choice of organic molecules of biological interest, metal-organic complexes and small and large clusters. These studies aim to follow the evolution of the electronic properties of matter starting from a single isolated particle towards surface and bulk material in the condensed phase, passing through the steps of atomic and molecular aggregates of increasing complexity. Given the extremely low number of target particles in typical experiments, it is expected that inner-shell studies of clusters would greatly benefit from the advances in the 4th generation synchrotron radiation sources. Indeed, an inner-shell photoionization study of rare-gas clusters was among the first experiments performed at the first European FEL facility (TESLA, Hamburg) [13]. The inner-shell spectroscopies provide both site and element specificity in the experiments. An additional advantage offered

by the XES and RIXS techniques is that the penetration depth of soft x-rays is larger than that of electrons, which allows to probe atoms also below the cluster surface. Furthermore, photon detection techniques are not sensitive to plasma charging effects that arise when an intense FEL beam ionizes the sample material.
At the first step, we intend to perform inner-valence and inner-shell studies on soft x-ray fluorescence of molecular targets of lower complexity, such as smaller organic molecules of biological interest (pyrimidines, purines), of organometallic systems of catalytical interest (metallocenes) and rare-gas cluster with high aggregation numbers (Rg_n, n>1000). The complexity of the targets can later be increased to biotic systems in tautomeric equilibrium, such as DNA bases, small peptides on one side and porphyrins or bacteriochlorins and their metal complexes on the other side. A microplasma source is also available at the LDM beamline, for experiments on clusters of metals and metalloids and their oxides. Therefore, such metal clusters will also be studied whose inner-shell binding energies occur in the energy range of the FERMI@Elettra source.
Additional perspectives in RIXS schemes are added when considering pump-probe experiments, where a first excitation step is followed by ultrafast time development of the excited states and subsequent probe of core excited state by a FEL pulse. When using an optical laser photon as a pump, the probing of core electrons by RIXS and XES will allow a direct investigation of photophysical and photochemical processes with the elemental sensitivity of core electron spectroscopies. Although expected counts rate are still quite small for such experiments, since only a small percentage of the gas at the interaction point can be transferred to an excited state, we stress the fact that in the XES experiments there will be no background from inelastically scattered electrons, as encountered in alternative photoionization experiments. Additionally, simpler dipole selection rules govern the spectrum, resulting in an easier interpretation of the experimental data than in electron spectroscopy. Alternatively, the use of both pump and probe photon beams from the FEL will open new ways of exploring the evolution of core excited states through stimulated X-ray emission processes. In such experiment the width of the incoming radiation must be large enough so that the relevant energy losses can be probed with RIXS. Theoretical works [10, 11] have discussed such "four-wave mixing" schemes and similar non-linear phenomena (like pulse compression in a medium) which can be also probed with the FERMI@Elettra light.
Finally, experimental application of the apparatus concerns time-resolved pump-probe experiments in the strong-optical-field regime. Such experiments will exploit the combination in an atomic/molecular gas jet of the FEL radiation with the ultrashort and intense optical laser pulses synchronized with the FEL. The emerging XUV radiation will then be spectrally analyzed as a function of the delay between the FEL and the IR pulses, as discussed in a deeper detail in the following section. This class of experiments can be exploited for an all-optical measurement of the time duration of the FEL pulse.
As a test case, we briefly discuss the feasibility of the LDM experiments at FERMI@ELETTRA. We assume to have 10^{13} photons/pulse with the light focused into a 20-μm spot, length of the interaction region >0.5 mm, a molecular beam set-up to bring up the test gas to 10-20 mbar of equivalent pressure, atomic photo-absorption cross-section of 1 Mb at core edges of relevant light elements of the 2nd row (C 1s, N 1s, O 1s) or higher (10 Mb) for L edges of metals of the 3rd and 4th row. We can expect at least $\approx 10^{10}$ excitation events per pulse in the case of the K-edge of light elements. With a XES quantum yield of 2% and a collection efficiency of $8 \cdot 10^{-7}$, including also the detector efficiency, we expect 10 to 100 counts per FEL shot. The count rate will be higher in the more favorable case of metal L-edges. Considering the FERMI@Elettra repetition rate of 50 Hz, we also foresee the possibility of exploiting immediately the RIXS and XES techniques in pump-probe schemes for investigation on tautomerism through two-photon excitation schemes or on photocatalytic systems, such as porphyrins, where at least 10% of the molecules in the jet have been excited by photons coming from a IR-VIS-UV fs-laser.

3.2. Condensed matter and matter in extreme conditions

In recent times, various combination of photon-in photon-out spectroscopies have been used for chemical and structural investigations of condensed matter [15]. The sensitivity of resonant and non-resonant XES to modifications of the electron and atomic structure is well-known but its use has been of course limited to static properties excluding transient or excited states.
The development/availability of an efficient detector for XES a the TIMEX beamline and later at the XFEL under construction would be of paramount importance for a deeper understanding of the ultrafast transitions occurring on solid targets (surfaces and thin films) as an effect of the interaction with subpicosecond pulses. Our plan is to collect single-shot X-ray Emission spectra below and above the absorption edges, using both resonant and off-resonant conditions, taking advantage of the intensity of the pulses and of the tunability range. The simplest configuration that can be adopted in a typical pilot experiment at the TIMEX beamline is to use directly the first harmonic of the FEL1 beam as a pump to produce excited states up to the warm dense matter regime. Simple calculations shows that extreme high pressure and

temperature regimes are obtained and kept, at solid-state densities, for a few picoseconds after the pulse. The X-ray emission will be then that characteristic of the strongly excited system and new information about the electron and atomic structure can be obtained by these challenging single-shot measurements. We expect that for high intensities (>10^{12} ph/pulse), measurements with reasonable statistics collecting a few FEL excitation events are obtained. Typical first experiments include simple ultra-thin foils and/or surfaces of light elements and compounds (Si, Al and their oxides) and on light metals (Li, Na, Mg) for which electron excitation energies are found in the energy range of FEL1. The collection of XES spectra in the 10-200 eV energy range, as an effect of the FEL1 pulse, will be used to study the transitions occurring at the targets as a function both of the intensity and of the FEL1 pump wavelength. Those experiments can be extended to Be, B, and C (and to the L-edges of Al, Si) using the FERMI-2 source.
The set of experiments we are proposing could represent the first XES measurements of those materials under transient extreme conditions up to warm dense matter regime, typical of the large planet or star interiors. They are intended to provide information on the electronic and ion structure of the systems with the typical time resolution of the pump pulse. An important advantage of those XES experiments is that they will provide spectroscopic energy-resolved information using the fixed wavelength provided by the FEL, for a given intensity of the pulse. We envisage that, tuning for example the FEL energy just below the absorption edge, we can reconstruct the single-shot XAS (X-ray Absorption Spectroscopy) spectrum using the Kramers-Heisenberg relationships, as demonstrated experimentally using 3rd generation facilities [16]. In this way, structural XAS determinations on materials under extreme transient conditions would be available using a single (or a series of) FEL shot of given wavelength.
The proposed instrument fits the TIMEX end-station at FERMI@ELETTRA, that dedicated to experiments under extreme conditions. The end-station is conceived to perform transmission, reflection and pump-probe experiments and the inclusion of photon in-photon out spectrometry is an important additional technique. The basic idea for testing the spectrometer is to collect initially soft x-ray emission spectra (XES) from solid samples as a consequence of the excitation with a FEL soft x-ray pulse. This will allow direct ultrafast measurements of the changes in the electronic structure of the investigated samples. For the initial experiments, the easiest and already available configuration is certainly using just the X-ray FEL pulse to excite the system, using the XES emission spectrum as a probe. This allows to probe the highly excited solid system within the typical timescale of the pulse width. In a second phase, we plan to use pump-probe configurations including pumping/probing the system with a laser pulse. This set-up requires time-delay techniques to probe specific time delays in the picosecond regime (1-10 ps typically), possibly related to states with ions and electrons approaching thermal equilibrium. Even for a XES initial experiment using only the FEL pulses we remark here that: i) the duration of the pulse can be varied in a range (possibly 0.1-1 ps or more); ii) strongly excited states (non-thermal melting) are reached within 0.1-0.2 ps and can be probed directly. Clearly, by tuning intensity, wavelength and pulse duration we expect to reach different excited states probed by XES.
Regarding the expected detected events on the detector, this should not be a serious issue for condensed matter. Very good XES spectra collected at synchrotrons using 10^{10}-10^{11} monochromatic photons are available in the literature, thanks to the reasonably high X-ray emission yield and electron density of condensed matter. For soft X-rays, where the fluorescence yield decreases, there are already published examples of XES spectra collected at FELs. Looking at specific interesting cases and using tabulated values for X-ray emission yields, a collection efficiency of 8×10^{-7} and 10^{12} photons/pulse, we expect ≈240 detected events/pulse for Li (E_p 55 eV), ≈160 events/pulse for Ge (E_p 30 eV), ≈200 events/pulse for Se (E_p 55 eV), ≈360 events/pulse for Si (E_p 100 eV) and ≈2000 events/pulse for C (E_p 290 eV).

3.3. Time-resolved pump-probe experiments

The apparatus can be used also for time-resolved pump-probe experiments in the strong-optical-field regime, that will be realized by combining the FEL radiation with the ultrashort and intense laser pulses at 800 nm, available and synchronized with the FEL. The two beams will be collinearly focused in an atomic (or molecular) gas jet. The peak intensity of the IR pulse will be in the range 10^{12}-10^{14} W/cm^2, the intensity of the FEL pulse will be suitably attenuated according to the experimental requirements. The emerging XUV radiation will then be spectrally analyzed as a function of the delay between the XUV and the IR pulses. Several phenomena are expected to be observed owing to the interaction of the two pulses. In particular we will concentrate on two of them: 1) dynamical Stark shift in neutral or ionized species and 2) enhancement of high-order harmonic generation by intense IR pulse mixed with FEL pulses.
An intense IR pulse induces a transient AC Stark shift in the energy levels of neutral atoms or molecules. Exploiting two delayed IR pulses, it is possible to induce such effect in ions. In a first set of experiments, the Stark shift will be probed looking to the transient absorption of the (attenuated) XUV pulse, temporally overlapped to the IR shifting pulse. This kind of experiment has been recently performed with isolated attosecond XUV pulses as a probe [17]. The proposed experiment explores nevertheless a completely different range of parameters, owing to the long pulse duration of the

FEL with respect to the IR optical cycle. It is worth noting that, in some circumstances, the IR pulse could even lead to transient transparency for absorption of XUV light tuned to suitable transitions [18]. The acquisition of the XUV spectrum as a function of the time delay between the two pulses can be exploited for an all-optical measurement of the FEL duration.

An intense IR pulse focused in a gas jet produces high order harmonics which span the VUV and XUV spectral regions. It has been theoretically proposed that the injection of an XUV seed along with the intense IR driving pulse should lead to the generation of new harmonic components [19] as well as to the enhancement of the harmonic radiation [20]. This latter effect has been also experimentally demonstrated exploiting the XUV harmonics themselves as a seed [21]. The generation/enhancement of harmonics induced by mixing intense IR pulses with the FEL pulses will be investigated as a function of the delay between the two pulses. The dependence of the enhancement on the wavelength of the FEL seed as well as on the nature of the target gas will be investigated. Again, since the effect is maximized when the two pulses are temporally overlapped, the acquisition of the harmonic spectrum as a function of the delay between the two pulses can be exploited for a measurement of the FEL pulse duration.

4. CONCLUSIONS

We have reported the design of an innovative and portable photon spectrometer to be used in FEL beamlines for photon in – photon out experiments. The usefulness in some application examples has been discussed and the mechanical feasibility reported.

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