PRELIMINARY SPECIFICATIONS FOR AN ERL-BASED FEMTOPULSE SOURCE FOR ULTRAFAST X-RAY SCIENCE

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Abstract

A recent workshop held at CEA, has led to discussing the characteristics of a possible high brilliance femtopulse hard X-ray Linac facility that would complement hard X storage rings in the domain of ultrafast X-ray science and would satisfy timely the European needs in a new and rapidly developing area. The source could be based on a 6 GeV recirculator, in the MHz repetition rate range with energy recovery, and use superconducting RF technologies of TESLA type as developed at CEA together with SRF photogun source technics. The facility would feature an ”undulator farm” in the keV range.

1 INTRODUCTION

A Workshop held monthly at CEA/Grenoble during the last semester of 2001, upon the initiative of one of the co-authors [1], has led to the present summary report. Initially, this ”XPF” Group (XPF stands for X-ray Pico-Femto second) was intended to draw the attention of Grenoble scientists about the new prospects offered by emerging SR technologies for *Ultrafast X-ray Science*. In fact, it turns into the establishment of a preliminary scientific case and to some requirements for a user facility that would complement the ESRF in the PF regime. The scientific applications are all derived from the experience gained at the ESRF beam lines by various scientists, both users and beam line responsibles. The subjects, discussed hereafter, encompassed specific aspects in biology, chemistry, nanomagnetism and holography, and some hints to the R&D in view of a machine that could satisfy these needs.

2 ATOMIC MOTIONS IN BIOLOGY

One of the most exciting prospects of a femtosecond X-ray source is that the short pulse can probe atomic motions in chemical and biochemical reactions on the chemical time scale. This time scale is defined by the time it takes to form and break chemical bonds, normally 10 to 100 fs. Spectroscopies can usually determine when a molecule breaks up or change, but there is no information about where the molecule went. By contrast, the interference of an ultra short X-ray pulse with a moving molecule gives rise to a spatial variation in the scattered intensity that can be inverted into changes in bond lengths and angles. Ultra fast experiments are always done using the pump and probe scheme where the time resolution is obtained from the delay between the optical pulse and the X-ray pulse. The optical pulse synchronizes the ensemble (time zero) and raises the energy above a reaction barrier. The delayed X-ray pulse takes a snapshot of the spatial configuration. In myoglobin, for example, one can track, using the Laue method, motion of gas molecules (CO, NO and O_2) in and out of the protein - a motion essential to the transport of oxygen from the lungs to muscles [2]. Another example of an ultra fast process is the structural relaxation following electron excitation [3].

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3 MOLECULAR SYSTEMS

There is an enormous potential of chemical processes for which time-resolved structural information would be highly desirable. Since the amplitude of spontaneous fluctuations of a molecular system is, in general, small, most structural changes must be initiated. There are many ways to achieve this, all of which can be induced with pulsed lasers on an ultrafast time scale. Typical examples for these laser-induced structural changes are : photo-initiated chemical reactions from simple chemical systems (dissociation, isomerization, atom transfer) to complex biological systems (hemoglobin, myoglobin, photoreaction center, retinal), laser-induced electron transfer, within and between molecules, at interfaces and the subsequent molecular reorganization, impulsive generation of phonons and vibrations in liquids and solids, temperature jumps in protein folding etc. The time resolution required to resolve the elementary steps in nearly all of these processes is ≤ 1 ps. The example of the HgI2 photodissociation as a ”simple” test case for such studies is described in [4]. It has revealed a certain number of experimental requirements for structural studies of transient species. The time resolution must be sufficient : the dissociation of the molecule of mercury iodide is completed in less than one ps, most of the processes are achieved in less than 10 ps. The signal to noise ratio must be high, implying both sufficient X-ray flux and the minimization of ”useless” signals, such as the contribution of air, fluorescence of the sample , etc. In the case of HgI2, by intensive signal analysis, an effect of the exciting laser on the diffraction signal was revealed, but the signal-to-noise ratio was insufficient for a detailed analysis of the effect. The overall stability is extremely important, since differences of structural signals will usually be measured (instabilities in the X-ray flux, drifts from the CCD detector).

Requirements for bio-motion & chemical reactions
Although it would be nice to have 10 fs X-ray pulses, the low charge per bunch precludes it. It is more realistic at this stage to aim at $100 - 200$ fs pulses, with similar accuracy for arrival time on the sample. The bunch frequency should be in the 1-10 kHz range, and with dark periods isolating a single bunch. A central energy of 15 keV is required. A 2x5 m long undulator [5] at 6 GeV would provide enough flux for experiments such as 3D-mapping of a small protein or the transient structure of a small molecule in solution. It further appears that 6 GeV is a *real optimum* in view of heatload problems.

4 NANOMAGNETISM

The physics of magnetization switching in nanosystems is the subject of very intense studies. These are motivated by the needs of high density magneto-recording industry where fast magnetization reversal is essential to insure a high data transfer rate. Computer hard disks presently work at $300 - 400$ Mbit/sec, corresponding to a reversal time of the magnetic bits of the order of some nanoseconds, and sub-ns reversal speeds are at reach. Using the time structure of X-rays in the single bunch mode at the ESRF, time-resolved X-ray magnetic circular dichroism (XMCD) measurements have been performed in pump-probe mode with few ns magnetic field pulses. For a Co/Cu(8nm)/FeNi spin valve, it has been shown that in the static and the dynamic regimes a different coupling can exist between the two ferromagnetic layers [6]. The scattering counterpart of XMCD, X-ray magnetic scattering (XMRS) is powerful probe of magnetic nanodomains [7] and 3D magnetic moment distribution in heterostructures. Coherent scattering can be performed using a pencil beam (20 μm), with hopes to reconstruct the magnetic disorder. Such magnetic holography can be developed into time-resolved mode, similarly to XMCD.

Requirements for nanomagnetism
Energy : $500 - 900$ eV ($L_{2,3}$ edges of transition metals), with extension to 1.5 keV for M-edges in rare earths.

X-ray bunch length : < 1 psec ; looking at fundamental processes requires to go well below the ps.

Repetition rate : up to 2 MHz, sufficient to guarantee a current of the order of a few mA's.

5 ATOMIC X-RAY HOLOGRAPHY

This emergent method is based on the same principles as traditional holography with light : a coherent wave (called the reference wave) illuminates the object and the detector surface. The intensity modulation caused by the interference between the reference wave and the wave scattered by the object (called the object wave) is recorded. This interference pattern contains both the phase and the magnitude information of the object wave. Therefore the original wavefront can be reconstructed, giving the 3D spatial arrangement of the objects. Direct extension of optical holography to the imaging of atoms was prohibited by limitations on the detector resolution and on the size of the radiation source. The real space resolution has reached the diffraction limit, and the imaging of light atoms has also been demonstrated [8].

Requirements for time-resolved holography
A X-ray hologram picture, at a given energy, requires at least $\sim 10^{10}$ photons. Since the practical efficiency is $\sim 10^{-5}$, this results in 10^{15} photons/picture in the incident beam. We will now assume an incident "pink" beam from the undulator (rbw 1%), that is without a monochromator. In that case, ESRF would deliver the 10^{15} photons in 0.1 sec, and 10 seconds allows for good statistics at a few X-ray energies. Using an ERL source with 100 fs bunches at 1 kHz repetition rate, an average flux of 10^{11} in the undulator 1% rbw is expected, thus 10^{15} photons in 1000 sec. This turns again, for good statistics at a few energies, to about 1 day. Furthermore, at a repetition rate of 1 MHz (1 ps bunches) the acquisition time would decrease to the minute range.

6 SUMMARY

Tab. 1 summarizes the source characteristics in order so as to meet the physics requirements above.

Table 1: Summary of SR requirements.

	pulse length	rep. rate	X-ray energy (keV)
Biology	$100-200$ fs	$1-10$ kHz $^{(*)}$	opt. ∼15 (4– 40)
Chemistry	$\ll 1$ ps	1 kHz	opt. $\sim 15-20$
Magnetism	< 1 ps or $\ll$ 1ps	$<$ few MHz	$0.7-0.9$ (up to 1.5)
Holography	100 fs–1 ps	up to few MHz	$10-30$

$^{(*)}$3 pulses spaced by more than 150 ns are of interest

7 SKETCH OF A POSSIBLE FACILITY

Based on the experience gained with storage rings and on a review of 4th generation light sources discussed worldwide it turns out that the Energy Recovery Linac solution (ERL) appears as a suitable candidate. The main objective concerning the discussions on the machine characteristics was to review the principles and the beam characteristics, to highlight the main difficulties and to give some guidelines for unavoidable R&D to be performed prior to proposing a hard X-ray User facility.

Starting from user requirements on number of photons per pulse (in the range of 10^7 ph/0.1%Bw/pulse) it turns out that a beam energy of 6 GeV would be optimum assuming an in-vacuum undulator of 6 mm gap or somewhat

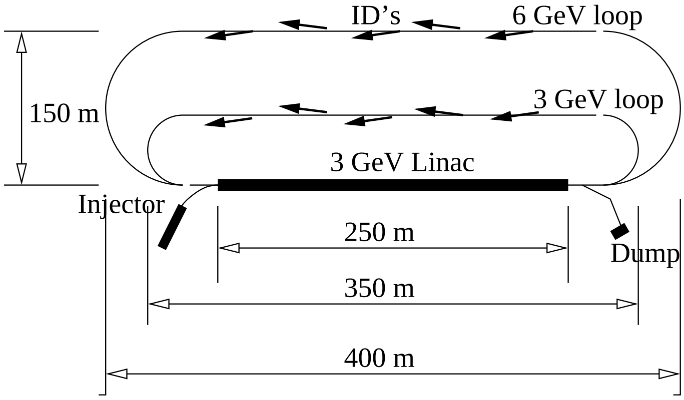

Figure 1: General structure of the facility.

below, with transverse emittance 0.2 nm in both planes and charge 1 nC per bunch. Depending on the type of experiment, the bunch length should vary from 0.1 ps to 1 ps after bunch compression, and the repetition rate from 1 kHz to several MHz (Tab. 1). In the MHz regime, no solution exists today for an electron source to deliver the 1 nC charge per bunch and a small emittance. A compromise between these performances would then be necessary. Several laboratories are engaged in an R&D effort for such an electron source (CW RF gun or DC gun) delivering a charge of less than 100 pC [9]. In the kHz regime, a well optimised pulsed RF gun could deliver the needed characteristics. The bunch compression necessary to obtain sub-picosecond bunches should be obtained at the final energy level without affecting the beam qualities. This represents an important challenge and needs strong R&D effort and possibly tests on the intermediate phases of the project.

Size and cost minimization of a 6 GeV continuous-wave (CW) superconducting electron accelerator, with up to a few mA beam, lead to a recirculation scheme. The optimum number of passes in the Linac is a matter of hardware cost [10, 11], yet two passes looks acceptable if one refers to other studies [12] whereas a higher number would have several drawbacks such as optics complexity for permitting coexistence of six or more accelerated and decelerated beams with very different rigidity ranges in the Linac, SR related emittance deterioration, beam instability intensity thresholds. In order to reach such high beam power level (several tens of MW), while limiting the installed electrical power to the 10 to 20 MW level, energy recovery (ER) is mandatory. It also permits to dump the beam at very low energy. However, such a scheme in a recirculating Linac complicates very much the overall design and the advantages should be carefully examined.

The proposed conceptual scheme for the 3 GeV superconducting Linac makes use of the developments made for the TESLA collider [13]. It could be composed of TTF-like accelerating modules containing 8 cavities operating at 1.3 GHz. Assuming an accelerating gradient of 20 MV/m, 18 modules (i.e., 144 cavities) are needed. With a Q_0 of 10^{10}, the dissipated power at 2 K is 40 W per cavity. These

Table 2: Machine parameters

Energy	GeV	3 & 6
Injection energy	MeV	10
dump energy	MeV	<10
Linac :		
Energy	GeV	3
RF frequency	GHz	1.3
Length	m	250
Mean accelerating gradient	MeV/m	12
Filling factor	%	60
Number of cryogenic modules		18
Number of cavities		144
Cryogenic module :		
energy gain per module	MeV	166.4
number of cavities/module		8
Length	m	12
9-cell cavity :		
accelerating gradient	MeV/m	20
Q_0		10^{10}
Operating temperature	K	2
Electrical Length	m	1.04
Beam :		
Max intensity	mA	< 10
Bunch charge	nC	1
Rep. frequency	MHz	< 10
Transv. emittance	πm.rad	10^{-6}

gradient and Q_0 have been routinely obtained in pulsed operation and low current [14]. It remains to be proved that they can be reached in CW mode with very high currents. The main developments will be on cryogenics, higher order mode (HOM) couplers, and stability. At the maximum current (1 nC, 10 mA) and with 4 passes in the Linac, the power which is lost by the beam in HOM is about 400 W per cavity. This power should be very efficiently extracted from cold temperature by couplers and absorbers of a new type, providing at the same time the necessary damping of these modes for preserving the beam quality. Even with a 95% extraction efficiency, 20 W of HOM power would be dissipated at cold, giving a total load of about 60 W per cavity at 2 K. This is well above the maximum power which can be extracted by the present design of the cryogenic system of the TESLA cavities. It may appear from further studies that the operating gradient should be set at a lower value, which may also be closer to an economical optimum. For an efficient use of the ER scheme, the cavities should be coupled to the RF sources with as high as possible external Q. The limitation here is sensitivity to microphonics. A sophisticated feedback system must be developed associated with active tuning system. A set of possible machine parameters is given in Tab. 2. Order of magnitude of installed electrical power for different modes is given in Tab. 3 assuming an external Q of 10^7 and a 90% extraction efficiency of HOM.

A development in four phases can be envisaged (see

Table 3: Installed power (RF+cryogeny)

Beam energy (GeV)	Installed power (MW)		
	Zero current	$< I >= 10$ mA	
	without ER	without ER	with ER
1	2.8	24.2	5.4
2	5.5	48.4	10.6
3	8.4	73	15.8
6	8.4	140	23.7

Fig. 1 for the geometry) :
- *phase 0 :* 10 MeV injector + 1 cryomodule + one small recirculation loop ; beam energy 176.4 MeV ;
- *phase 1 :* 10 MeV injector + 1 GeV Linac + 10 MeV beam dump ; beam energy 1 GeV ; installed power sized to 5.4 MW (for a reference upper value of 10 mA in the Linac). Two stages possible : *phase 1.1* with no recirculation loop, *phase 1.2* with the first, full 3 GeV return operated with the partial 1 GeV Linac ;
- *phase 2 :* the Linac is extended to occupy a full 250 m length, Linac energy upgraded from 1 up to 3 GeV ; beam energy 3 GeV. High intensity is made available with ER whereas 6 GeV low intensity is available with recirculation through the first return loop ; installed power progressively sized for 6 GeV in two passes with ER, i.e. 24 MW ;
- *phase 3 :* the second, 6 GeV return loop is built, Linac energy 3 GeV, beam energies 3 GeV, and 6 GeV available after 2nd recirculation ; beam power can in principle be raised to 6 GeV, 10 mA (i.e. 40 mA in the Linac).

The scheduling of the construction in a sequence of phases allows solving the important problems listed above, before building the high-energy machine. Of particular importance is the " phase 0 " with energy around 100-150 MeV and a recirculation loop. The same scenario has been proposed in other laboratories, in particular Cornell [12].

8 CONCLUSION

The scientific needs identified here concern bio-motions, chemical reactions, nanomagnetism and atomic holography. Even though the requirements in the different areas, as collected in Tab. 1, vary, there are a number of common features : all are pump-probe experiments, the pump being primarily a laser (hence the issue of synchronization) all require 1 nC per bunch, and would be happy with more (charge vs. bunch-length trade-off) all need a high degree of overall stability (including flux drift) a FEL approach was discarded, primarily for sample integrity.

A key parameter for the machine is the energy : it appears that 6 GeV is the optimum energy (and not any lower value). The proposed accelerator is an energy recovery Linac (ERL) of 3 GeV with double pass (Fig. 1) and with general characteristics as given in Tab. 1. The flexibility required to realize the various time structures imposes the CW mode. The construction steps allow to first deliver soft X-rays (and some harder X-rays as high undulator harmonics). Photon optics, under definition, indicates that present technology can handle the ERL undulator flux. A related aspect is the X-ray bunch compression to produce even shorter pulses.

Acknowledgements

We would like to thank here every member of the XPF Group, and collaborators. Apart from the co-authors : D. Block, E. Dooryhee, A. K. Freund, M. Jablonka, G. Naylor, Y. Petroff, S. Pizzini, G. Renaud, J.-L. Revol, A. Snigirev, O. Tchoubar, M. Tegze, H-P. Trommsdorf and J. Vogel.

We thank in particular J.-L. Revol (ESRF) for his reading and commenting on the manuscript.

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