# Time resolved x-ray microscopy

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SCIENTIFIC MOTIVATION

In the frame of R&D activity on future SPARX X-FEL source, the development of x-ray optics to successfully manipulate FEL radiation cannot be neglected. The unique characteristics of pulsed radiation produced by FEL have to be preserved both in beam transport and in conditioning optics for experimental purposes. This work gets into the larger field of x-ray optics development and testing in which the X-ray physics group of the Institute of Photonics and Nanotechnologies (IFN) headed by S. Lagomarsino, is involved for many years. The main new issue in working with FEL radiation (produced in a seeded scheme) with respect to ordinary synchrotron radiation is coherence and pulse duration in the femtosecond scale. Thus the first requirement for optics is the preservation of time structure of pulses and temporal (i.e. longitudinal) coherence of radiation. In other words, dispersion properties of different optics used should be understood in detail.

Another important characteristic of FEL radiation is the high degree of spatial coherence which allows, in principle, coherent microimaging and diffraction, but which in turn requires optical elements which preserve the coherence properties.

In view of time resolved x-ray microscopy with nanometer spatial resolution and femtosecond time resolution, an interesting optical element is the x-ray waveguide (WG) [1,2]. In fact, it has been demonstrated that WG is a suitable optics for x-ray microscopy because it can produce beams of nanometer size, fully coherent in transverse direction [3, 4].

Guiding optics, such as WGs and optical fibers are widely used for many types of radiation from microwave to visible optics and, in the last ten years, in hard x-ray range, too. The dispersion features of WGs and fibers were extensively studied in the past to fabricate suitable devices for optical communication. The utilization of WGs could be extended from XUV to hard x-ray spectral regions, however paying attention to their dispersion properties that, in principle, could affect dramatically the propagation of short pulses. A WG is an optical resonator in which incoming radiation can propagate only as discrete set of modes. The condition on refraction index $n_{guid}$ of guiding layer with respect of refraction index $n_{cl}$ of cladding layers for resonance to take place is always: $n_{guid} > n_{cl}$. Two different kinds of coupling were used up to now with hard x-rays: "resonant beam coupling" and "front coupling" schemes (fig 1).

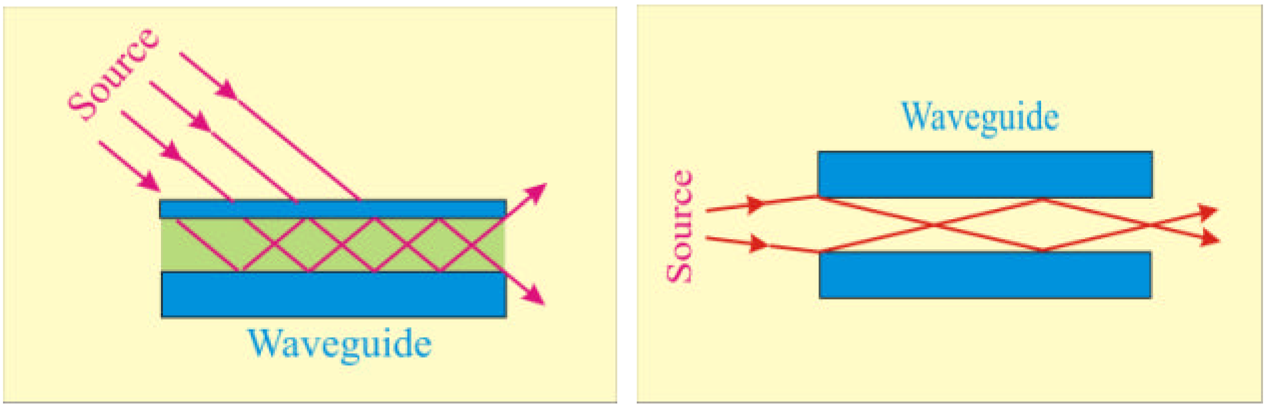


**Fig. 1: Resonant beam coupling scheme (left) and Front coupling scheme (right).**

In the former [1] the cladding and guiding layers are deposited on a flat substrate, and the upper cladding layer is so thin to allow incident radiation, in condition of total external reflection, to evanescently penetrate in the structure and couple with some guided mode depending on angle of incident. Specific calculation of transmission properties of such a scheme are reported in literature [5].

The latter scheme (front coupling) involves focalization of the x-ray beam just at one side of entrance of WG [2]. In this way good efficiency of coupling could be reached but the propagating field is in general a superposition of different modes. A slight modification of this scheme involves the formation of a standing wave field in front of the entrance of the device [6] in such a way that the mode selection becomes, in principle, possible (Fig. 2). In this configuration the incoming beam is totally reflected just at the entrance of WG, thus preparing the standing wave field. Propagation of the $m^{th}$ mode occurs when the angle of incidence matches the resonance angle $\theta_m$ for that mode.

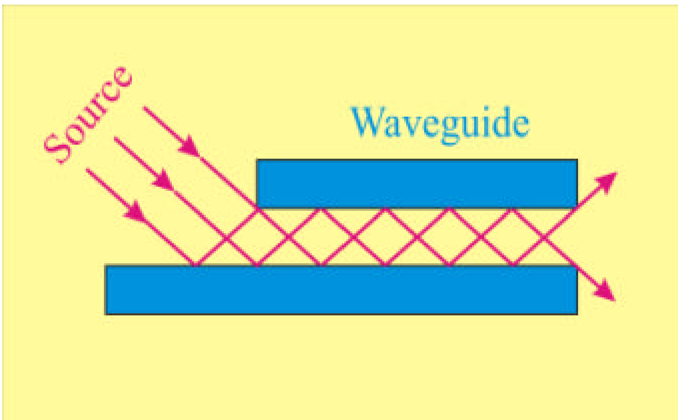


**Fig. 2: Front coupling scheme with mode selection.**

Other advantages in this second scheme of coupling are directly related with the possible use of WG with FEL radiation. First of all front coupling WG can be fabricated with vacuum as core layer and this makes this device capable of working also in the soft x-ray region, where absorption would hamper the use of solid guiding layers. In Fig. 3 we

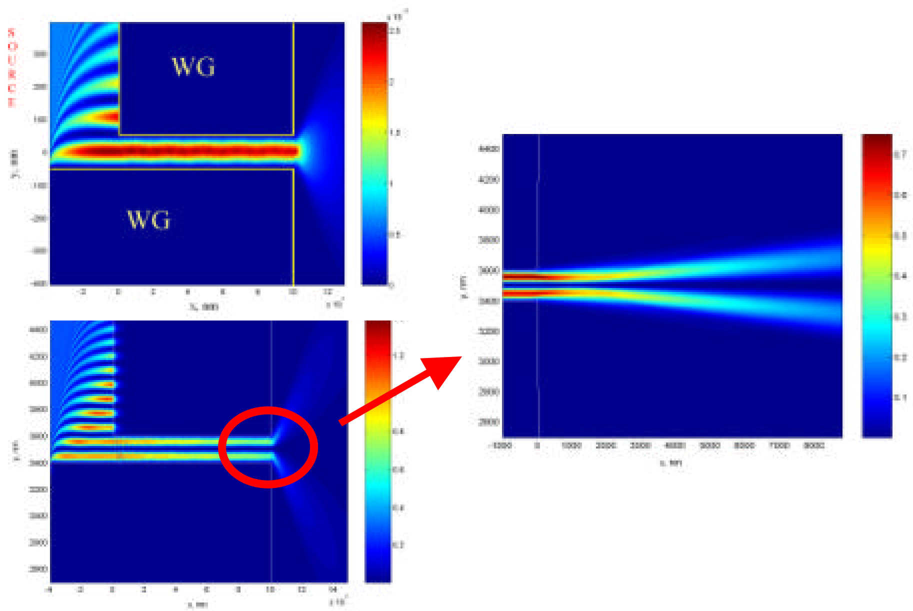


**Fig. 3: Example of propagating modes for 1st order (upper left) and second order (lower left). In this latter case beam splitting is accomplished. A magnified image of the exit of WG in the case of second mode is also shown on the right.**

report the calculated intensity distribution for a typical situation of front coupling with mode selection. The simulation was performed by means of a finite-difference approach, numerically solving Helmoltz equation in parabolic approximation [7]. In the left part we report the distribution for first resonance order. Second order, with beam splitter effect, is reported in left part.

WAVEGUIDE AS OPTICAL ELEMENT FOR TIME-RESOLVED X-RAY MICROSCOPY

Since propagation of guided modes is inherently an interference effect, the radiation at the WG exit is coherent. This allowed, for example, phase contrast microscopy with high spatial resolution [1]. The WG spatial and angular acceptance, $A$ and $Df$ respectively, is related to the phase space coherent volume, which is given approximately by: $A*Df\sim 0.44\,l$ [5]. The WG transmission, in case of front coupling and vacuum core layer, is connected with the small penetration of the electromagnetic wave, which has nodes at the two interfaces between guiding and cladding layers, into the cladding layers. The attenuation length can be expressed analytically by:

Fig. 4 shows the attenuation length (i.e. the length of WG for which intensity is fallen down of a factor *e*) for a planar waveguide with a gap of 400 nm as a function of wavelength in the range 1-10 nm, for different choice of cladding layers material. Both in the water window (2.3 nm < λ < 4.4 nm) and in the carbon window (4.4 nm < λ < 5 nm) waveguides with appreciable lengths (> 1 mm) have good transmission properties. These two wavelength ranges are important for x-ray microscopy of biological tissues, because good contrast can be obtained even “in vivo” between different cellular elements without the need of contrast enhancement by means of additional colouring substances as in the case of visible light.

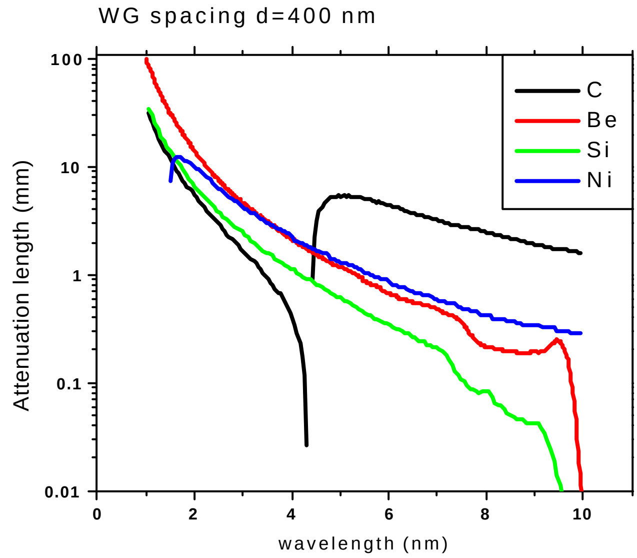


**Fig. 4: Attenuation length for various cladding material. The spacing of WG considered is d=400nm**

All this considerations are valid insofar as dispersion of device is low enough to permit reliable manipulation of pulses. For this purpose we carried out detailed calculations to evaluate dispersion parameter $D = -\frac{2\pi c}{\lambda^2} GVD$ related to group velocity dispersion $GVD = \left.\frac{\partial^2 k_z}{\partial \omega^2}\right|_{\omega_0} \equiv k_z''(\omega_0)$. These formulas refer to the propagation of a pulse with central angular frequency $\omega_0$ in the direction $z$. In the case of a two-dimensional WG (i.e. a WG that confines radiation in two directions) we numerically evaluated dispersion for different material composing cladding starting from tabulated values of index of refraction $n$=1-δ-iβ [9]. Our calculations show that dispersion is actually low due to the small variation of index of refraction with wavelength in the x-ray region far from absorption edges. As an example we can see in Fig. 5 the calculated dispersion parameter for Berillium and Carbon. These materials were chosen for practical

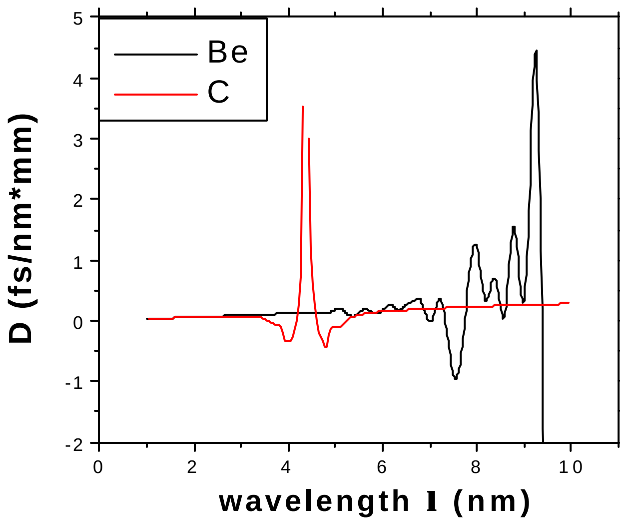


**Fig. 5: Dispersion parameter calculated for Be and C cladding WG with a gap of 400x400 nm$^2$.**

application in wavelength range between water and carbon windows.

However, it is interesting to note that close to absorption edges, significant anomalous dispersion takes place. Accurate quantitative evaluation of this effect is not easy, because the tabulated values of the real and imaginary parts of the refraction index close to absorption edges are not very much reliable, but nevertheless we have clear indications that with adequate WG parameters (length, gap value, type of cladding material), and at given wavelengths, the WG can be used as an efficient anomalous dispersive optical element. With a suitable incident chirped beam, schemes can be then adopted to significantly shorten the pulse time length.

The unique property of WG to produce two coherent beams at its exit in the second resonance mode, suggests its use as a beam splitter. The two beams are separated by the angle $2\lambda/d$, and this allows to let them going along different path lengths before to reach the sample, thus realizing a retardation line. Two possible schemes are presented in Fig. 6, the first involving just one mirror and the second two mirrors. It is worth to note that the two beams are inherently coherent, because they are produced in the same interference process, and with a correct choice of the WG parameters are also essentially dispersion-free. Thus all x-rays pump-probe experiments, without the need of external sources, can be realized with this scheme. This circumvents the complicated problem of synchronization in the femtosecond scale. The two beams from the waveguide can be used, for example, to dynamically study radiation damage processes. One or both beams can also be refocalized on the sample to enhance photon density, through appropriate use of elliptically bent mirrors [10].

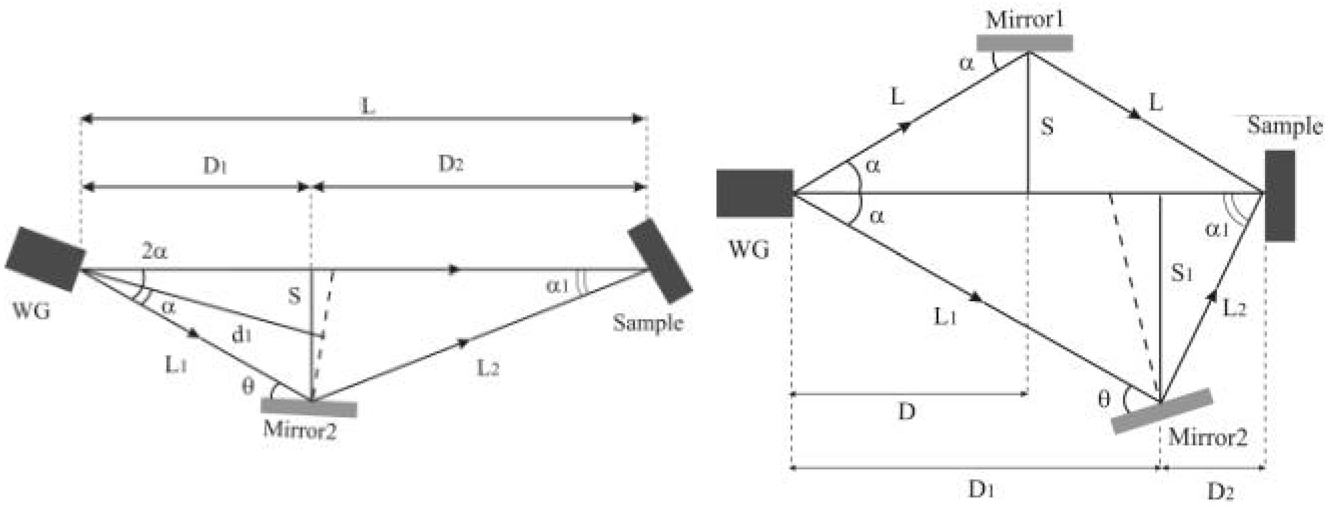


**Fig. 6: Experimental schemes involving WG as a beam splitter for realization of delay lines.**

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