A Simple Short-range Point-focusing Spatial Filter for Time-resolved X-ray Fluorescence

C. Höhr[1], E. Peterson, E. Landahl, D. A. Walko, R. W. Dunford, E. P. Kanter, and L. Young

Argonne National Laboratory, Argonne, Illinois 60439, USA

Abstract. A simple, compact, point-focusing spatial filter for x-ray fluorescence is presented. This construction maintains the large solid angle and directionality of existing designs but is more easily machined. Combined with a selective absorber, it can be used as an x-ray low-pass filter; this is one common approach taken in inelastic x-ray scattering studies and fluorescence spectroscopy. When combined with a scintillation detector, this device forms a large solid angle energy-selective x-ray detector with ns-scale time resolution that is useful for timing studies.

Keywords: time-resolved x-ray fluorescence, point focusing spatial filter, Argonne Advanced Photon Source
PACS: 61.10.Eq, 61.10.Ht

INTRODUCTION

Ultra-fast photo-induced changes in the local structure surrounding a central atom are typically studied with laser-pump/x-ray probe spectroscopies such as EXAFS (extended x-ray absorption fine structure) and XANES (x-ray absorption near-edge structure) [1]. In such studies, x-ray induced fluorescence detection is typically used due to its elemental specificity. X-ray fluorescence is particularly suitable to study molecules in solution at low concentrations ($\sim$ mM). Due to the large ratio of solvent to solute molecules, the desired signal is weak and there is substantial background from scattering and the possibility of further contamination from auxiliary x-ray lines. Thus, for these studies, a multi-element, energy-dispersive Ge detector is typically used [1] to select the x-ray energy of interest, e.g. the Cu $K\alpha$ line. However, such detectors have slow time response ($\sim 2\ \mu$s) and are cumbersome and expensive. Therefore, it would be of general interest to have a large solid angle, energy-selective, fast x-ray detector; herein, we report on such a device.

DESIGN

The detector required must be spatially and spectrally resolved to distinguish fluorescence from scattering. At a synchrotron x-ray source, where the laser repetition rate may be much less than the x-ray bunch rate, the detector must be fast enough to reject signals from neighboring (laser-off) x-ray bunches. While an energy-dispersive Si-drift detector can be used to detect emitted x rays, it requires microsecond time spacing between x-ray bunches. We wish to enable work in the Argonne Advanced Photon Source's (APS) 24-bunch mode, where the x-ray pulses are separated by only 154 ns. This mode has the advantage of more available beamtime and a shorter x-ray pulse duration [2]. Faster detectors, in general, have worse energy resolution and often a smaller solid angle. Here, we use a NaI detector with a time resolution of 50 ns and an aperture of 50 mm; the combination of a low-pass absorption filter and a point-focusing spatial filter (PFF) achieves sufficient reduction of unwanted background for studying the near-edge features of Kr. We chose a 41 μm thick Se foil as an absorption filter, corresponding to three absorption lengths for x rays 50 eV above the Se K-edge. We selected a so-called Z$-$2 filter since Br is not practical as a filter material. In order to block the re-fluorescence of the filter, a common method is to combine it with a PFF. The original idea of a PFF was given by Stern and Heald [3] with later improvements by Behne *et al.* [4] and found its application in the widely used Lythle detector [5]. A monolithic PFF is described by Seidler and Feng [6]. The spatial filter permits the fluorescence signal

[1] present address: Triumf, Vancouver, British Columbia, Canada

CP879, *Synchrotron Radiation Instrumentation: Ninth International Conference*,
edited by Jae-Young Choi and Seungyu Rah

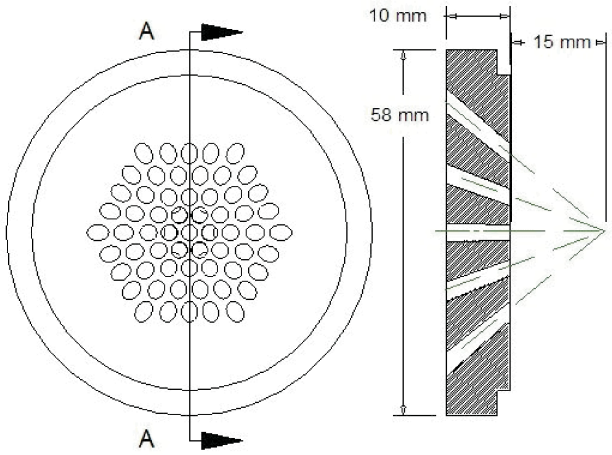

FIGURE 1. Bottom and cross-sectional view of the first design of the point-focusing filter (PFF) made from Al. Each hole has a solid angle of ~ 0.01 sr and all 61 holes together have a total solid angle of ~ 0.61 sr.

photons to travel directly from the target to the detector but blocks most of the Se-filter fluorescence, which its emitted isotropically. The net relative efficiency R of the Se filter and PFF for detecting photons below the Se K-edge relative to those from above depends on the length L of a PFF channel from its entrance to exit aperture and on the area A of one exit aperture:

$$R \propto \frac{L^2}{A} \,. \tag{1}$$

For this assumption, direct transmission from the sample is ignored, assuming the filter is thick enough. Also, scattering from the channel walls is neglected. For more details see [4] and the references within.

For a first test and to demonstrate the feasibility, we chose to design a PPF made from a solid Al disc, 10 mm thick and 58 mm in diameter. The 61 holes were drilled using tapered bits available in the local machine shop. This results in a focal length of 15 mm (see Fig. 1). The use of one tapered drill bit instead of several drill bits of different sizes for a step-tapered hole [6] allows a shorter machining time and therefore less cost. It also makes better use of the body material and consequently reaches a bigger solid angle. Each hole has a solid angle of ~ 0.01 sr and all 61 holes together have a total solid angle of ~ 0.61 sr.

TEST AND RESULTS

We tested the PFF at the 7ID beamline of the APS. Monochromatic 14 keV x rays were focused to a FWHM of ~ 10 μm into a Kr gas sample from an effusive gas jet. In addition, a ~ 100 μm FWHM focused laser beam (2.5 mJ energy per pulse, 40 fs pulse width, 800 nm central wavelength, 1 kHz repetition rate) was overlapped in space and time. Kr atoms were ionized by the laser and subsequently probed by the x rays as a function of time delay between pump and probe, space, x-ray energy and laser polarization (for experimental details and discussion of the results, see [7, 8]). In Fig. 2 the background suppression is demonstrated, measured with a Si-drift detector for energy resolution. Spectra (a) - (d) show the fluorescence for different filter combinations. The yields of the different peaks normalized to the corresponding Kr $K\alpha$ peak are listed in Table 1. For the unfiltered spectrum (a) the Kr $K\alpha$ fluorescence, along with background from stainless steel (SS) and Kr $K\beta$ fluorescence which contains elastic scattering background, is seen. As expected, with just the low-pass Se-filter between target and detector, the energy spectrum (b) shows both additional Se fluorescence and an immense reduction for SS and Kr $K\beta$ (which is inseparable from the elastic scattering background). From Fig. 2 (c) it is clear that the PFF alone already eliminates background scattering from the stainless steel of the sample chamber due to its different point of origin. For the same reason, the elastic scattering background mixed with the Kr $K\beta$ peak is significantly reduced. The Kr $K\beta$/Kr $K\alpha$ ratio is now 0.146 and agrees with the literature value [9] of $K\beta/K\alpha = 0.151 \pm 0.005$. Finally, the Se filter and PFF combined (Fig. 2 (d)) purge the spectrum from SS, Se and Kr $K\beta$ fluorescence. In addition, comparing count rates, the advantage of a 50

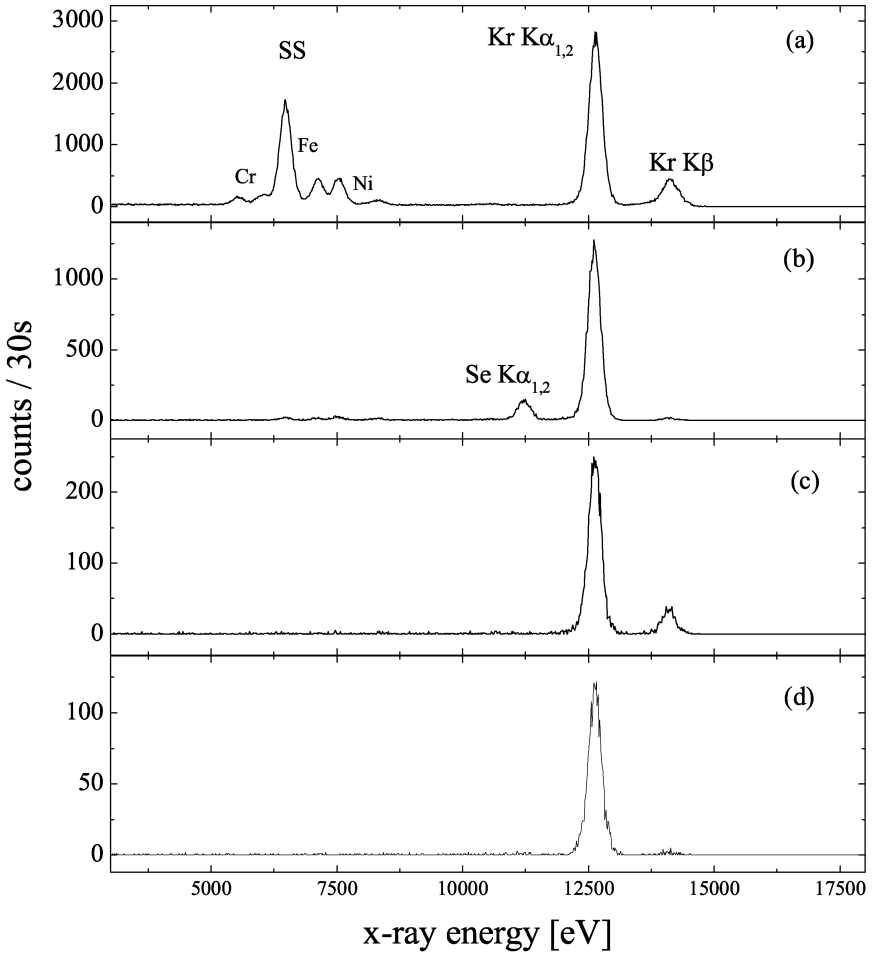

FIGURE 2. X-ray fluorescence spectra of Kr measured with a Si-drift detector and different filter combinations. The raw spectrum (a) shows the Kr $K\alpha$ and $K\beta$ peaks as well as some background at lower energies originating from stainless steel (SS) and under the Kr $K\beta$ peak from elastic scattering. Spectrum (b) shows a suppression of the Kr $K\beta$ and elastic background x rays due to the Se-absorption filter and also a new peak caused by the fluorescence of the Se filter itself. In (c), measuring with the PFF only, the SS is strongly suppressed due to its different point of origin as well as the elastic background underneath Kr $K\beta$. Finally in spectrum (d), with both the Se filter and the PFF in place, a nearly pure Kr $K\alpha$ fluorescence is recorded. Yields normalized to the Kr $K\alpha$ in the corresponding spectrum can be found in Table 1.

mm NaI detector in combination with the PFF over the Si-drift detector is not just the needed time resolution but also a six-fold increase in count rate due to the larger solid angle.

After this successful first test, a second, modified PFF was designed and machined with Mo as the body material, as seen in Fig. 3. Using Mo for the PFF channel walls will result in a more than one order of magnitude smaller contribution from Se fluorescence scattering off the walls than the directly propagated contribution from the Se

TABLE 1. Yields of the peaks in Fig. 2 (a) - (d) normalized to the corresponding Kr $K\alpha$ peak.

	(a) raw	**(b)** Se filter	**(c)** PFF	**(d)** PFF + Se filter
Fe $K\alpha$	0.57 ± 0.01	0.026 ± 0.002	0.002 ± 0.001	-0.002 ± 0.001
Se $K\alpha$	—	0.122 ± 0.002	—	0.005 ± 0.002
Kr $K\beta$	0.21 ± 0.01	0.018 ± 0.002	0.146 ± 0.003	0.017 ± 0.003

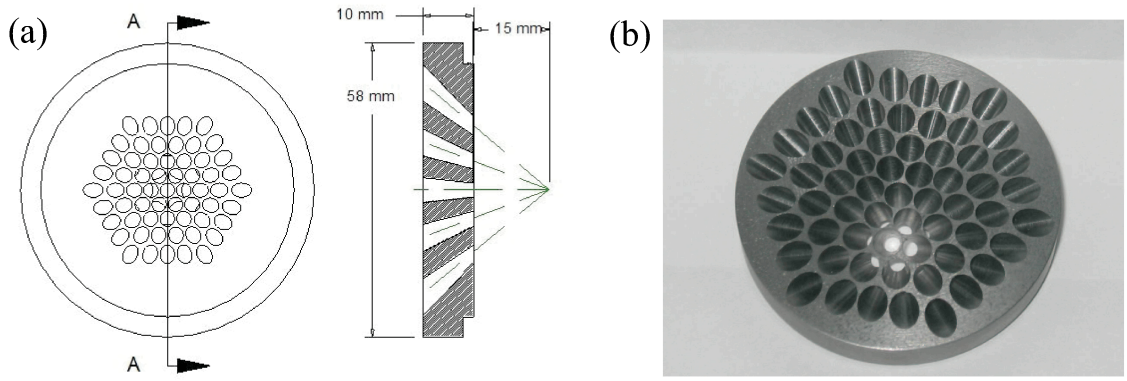

FIGURE 3. Construction drawing (a) and photo (b) of the advanced design made out of Mo with a solid angle of 0.03 sr per hole, with a total solid angle of 1.83 sr for all 61 holes.

filter [4]. The holes are more tapered (solid angle of 0.03 sr) which results in an overall solid angle of 1.83 sr and therefore an expected further improvement of the count rate by a factor of three. Although this new design has a three times bigger channel exit aperture A and, hence from Eq. 1, a three-fold decrease in efficiency R, we still expect a reduction in background sufficient for our needs. A successful test of this new PFF will allow the most common fill pattern of the APS to be used for time-resolved x-ray fluorescence studies.

ACKNOWLEDGMENTS

We are very thankful to Gerry Seidler, Ed Stern, and Steve Heald for fruitful discussions and the loan of an absorption filter. This work was supported by the Chemical Sciences, Geosciences, and Biosciences Division and the Advanced Photon Source by the Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy, under Contract No. W-31-109-Eng-38.

REFERENCES

1. L. Chen, *Angew. Chem. Int. Ed.*, **43**, 2886 (2004).
2. E. Landahl *et al.*, in preparation.
3. E. A. Stern and S. M. Heald, *Rev. Sci. Instrum.*, **50**, 1579 (1979).
4. E. A. Behne, Yejun Feng, and G. T. Seidler, *Rev. Sci. Instrum.*, **72**, 3908 (2001).
5. F. W. Lythle *et al.*, *Nucl. Instrum. Methods*, **226**, 542 (1984).
6. G. T. Seidler and Yejun Feng, *Nucl. Instrum. Methods Phys. Res. A*, **469**, 127 (2001).
7. L. Young *et al.*, *Phys. Rev. Lett.*, accepted.
8. C. Höhr *et al.*, submitted.
9. R. B. Firestone, *Table of Isotopes, CD-ROM Edition*, edited by V. S. Shirley, Wiley Interscience, 1996.