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REVIEW PAPER

REAL X-RAY OPTICS - A CHALLENGE FOR CRYSTAL GROWERS

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The methods for X-rays focusing are reviewed. The importance, for the X-ray optics, of the crystals with lattice parameter gradient, or Delta crystals, is discussed and the state-of-art in the field of these crystals is shortly presented.

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1. Introduction

After the detection of the X-rays by W.C. Röntgen (1895) their physical character remained unclear for a longer time. Röntgen found qualities of the new rays which became very important for medicine and material tests, but since his experiments did not reveal any indications of refraction or diffraction he could not think of an X-ray optics comparable to light optics.

It took almost two decades until Max von Laue and his co-workers W. Friedrich and P. Knipping, encouraged by ideas of P.P. Ewald, found as the result of a diffraction experiment the famous "Laue diagram" (1912) which indicated on the one hand the wave character of the X-rays and on the other hand the periodic structure of the then used ZnS crystal. This was the birth hour of an ever increasing number of X-ray diffraction experiments and of all kinds of structure determinations and analyses covering the wide range from perfect single crystals down to amorphous materials - all of them working with fixed Bragg angles and thus being unable to change divergences of incident X-rays.

2. X-ray monochromators

Using the diffraction qualities of single crystals, several types of monochromators - flat and bent ones - have been developed in the last decades, but again they only maintained the divergence of the incident radiation.

Part of this monochromator development became in the last time the application of multilayers. Their fabrication is well known from semiconductor industry: defined amounts of two materials with strongly different densities are sputtered alternately onto a basic material. Due to their periodicity these layer systems are able to produce X-ray interferences similar to real crystals and may therefore be considered as two-dimensional or lamella crystals. The periodicity factor lies somewhere between 15 and 100 Å, so they are not suited for very short X-ray wavelengths. As will be shown later multilayers can be tailored so as to serve as divergence-changing elements.

Such elements, applicable in special cases, have been developed by applying X-ray total reflection, as did Wolters when he tried to realize an X-ray microscope. The "Wolters telescopes" are very successful in focusing incident radiation from stellar X-ray sources to the CCD sensors of X-ray satellites like ROSAT, but on an extremely high technical expense.

X-ray total reflection, however, has also found applications in our earthly laboratories for some years, based on grazing incidence in hollow glass capillaries which act as waveguides. The critical angle for total reflection Θ_{cr}, the Fresnel angle, is very low (glass: $\Theta_{cr}\left[mrad\right] \approx 32 / E\left[keV\right]$, thus $\Theta_{cr} = 1\,mrad = 3.4'$ for 32 keV radiation). This is a restriction to longer wavelengths, but at the same time offers the opportunity to partly suppress shorter wavelengths.

In the simplest form -just one capillary (Fig. 1) - a small part of an incident divergent beam can be made parallel (down to the value of the Fresnel angle), with some gain in intensity at the place of the sample [1].

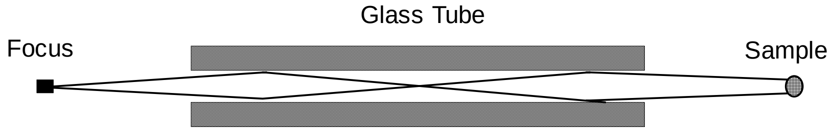

Fig. 1 A glass-tube collecting a divergent X-ray beam by total reflection.

Nowadays glass capillaries with inner diameters of about 10 μm can be fabricated, some 1000 of them are then combined to flexible fibbers, and with say 1000 of such fibbers "multifiber polycapillary optics" have been realized, e.g. collimating lenses, collecting incident radiation at a large angle and focusing it to a small line or point with energy gains up to 10^3 and more (Fig. 2). So a low-power sealed-tube can replace a rotating-anode device for some applications [2].

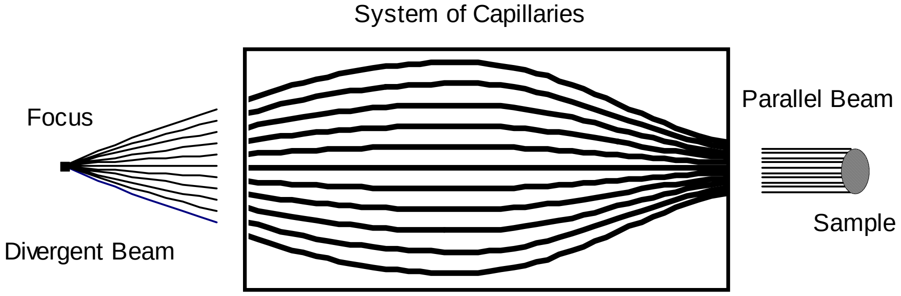

Fig. 2 A multicapillary system used as a collecting lens for X-rays.

Straight polycapillaries with square channels can even be used for synchrotron beam splitting. All in all the application of capillaries opens the way to some kind of X-ray optics.

Very recently total reflection has been applied in a "Micromirror" with an ellipsoidal profile, accurate to within 1 μm, which focuses X-rays from an 18 W generator line focus to a small point with divergence 1 mrad, delivering there the same intensity as would be available from a rotating anode generator [3] (Fig. 3).

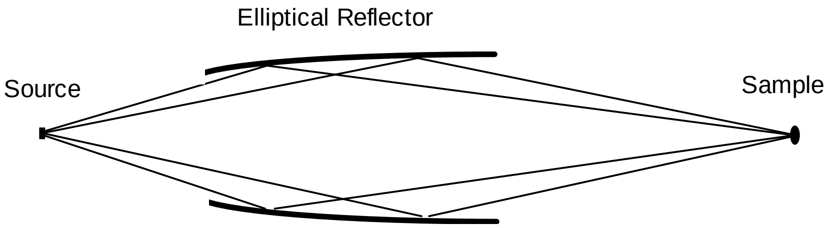

Fig. 3 Micromirror based on total reflection.

Apart from total reflection, also the refraction of X-rays can be used for optical systems. The refraction index of X-rays is extremely close to unity and has the opposite sign compared to that of visible light. Therefore a single element as shown in Fig. 4 acts as a collecting lens with very low efficiency. So one has to combine a series of these elements to get a significant change in divergence. But then the relatively large absorption has to be overcome - possible only by applying synchrotron radiation. Using 42 aluminium lenses in series the radiation source of the ESRF (Grenoble) has been reduced to a 1 × 14 m^2 small line; an X-ray microscope with a resolution of 300 nm has been realized, and the next step will be the application of beryllium as lens material, due to its smaller absorption coefficient [4].

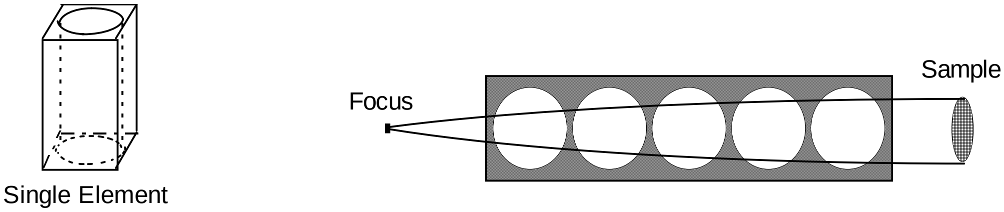

Fig. 4 Optical condensing system based on refraction by the transfer of X-rays from higher density materials (e.g. aluminium) to air.

3. New methods for X-rays focusing

A huge step in direction towards true X-ray optics was the discussion of new methods for focusing X-rays where "the Bragg angle for a given wavelength of photons is varied by varying intra-crystalline plane spacing as a function of position in the crystal" [5]. Two possible methods are mentioned: varying the temperature of a crystal as a function of position by applying a thermal gradient, and varying the elemental composition of a crystal made of two or more materials. With the second method considerably larger changes in the d spacing can be achieved. This is illustrated in Smither's paper by the following figures: An addition of 6% Sn to a Ni crystal increases the d value by 1.5%, equivalent to changing the Ni temperature by 1094^{0}C. And a still larger d gradient is to be expected in a Si-Ge mixed-element crystal: d changes from 5.434 Å (pure Si) to 5.657 Å (pure Ge), corresponding to a T of about 10^4 ^{0}C. Thus the second method is much more suited to realize a "gradient crystal", but needs highly sophisticated methods for crystal growing.

In a further paper [6] Smither discusses the application of his "variable metric crystal" to create X-ray parallel beams with narrow bandwidths (useful in work with two-crystal monochromators and synchrotron radiation) or, if the crystal is appropriately bent, to use it as focusing or defocusing lens.

Meanwhile and independent of Smither's work the present authors had applied (1987) for a patent (DE 37 02 804 C 2, 1994) and published some related work [7], where they described the

principles of what they called a "Delta crystal", which is identical to Smither's gradient crystal. Additionally the application of "lamella crystals" was mentioned there. This will be discussed below.
In Figs. 5a,b some applications of crystals with a lattice parameter gradient are shown: radiation incident at large angles from a line focus can be concentrated to parallel bundles of any width, or parallel bundles can be focused to small lines, if one crystal is used (Fig. 5a); radiation diverging from a point source can be converted into a parallel beam, or parallel incident radiation can be focused to a small point, if a second crystal rotated by 90^0 is used (Fig. 5b).

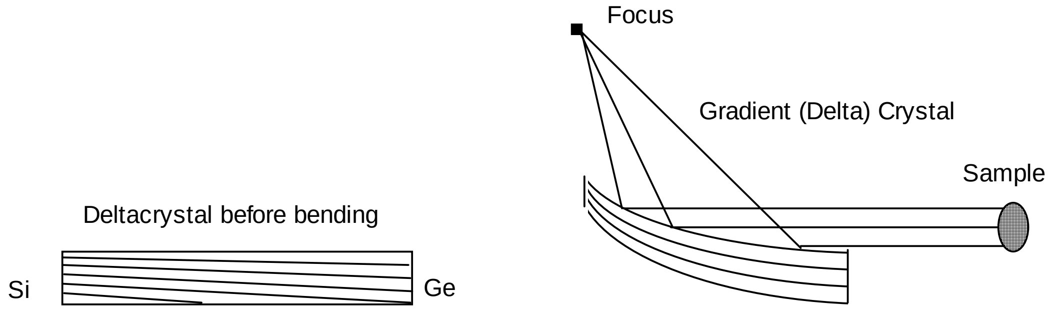

Fig. 5a Gradient crystal before and after bending, as a tool to convert a divergent X-ray beam.

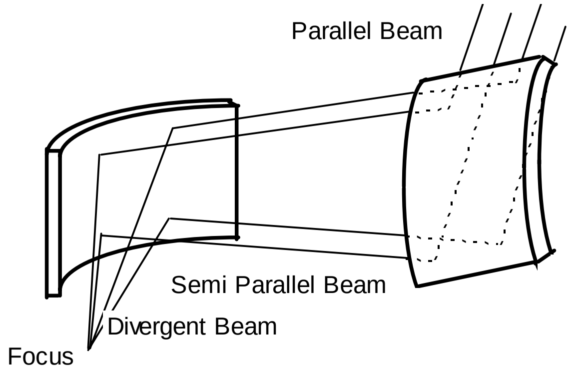

Fig. 5b Two gradient crystals convert X-ray beams in two dimensions.

Several attempts have been made to grow this kind of crystal [8]. The success, however, was at first restricted to relatively small crystals with small gradients. Abrossimov and Rossolenko, however, succeeded in growing large $Si_{1-x}Ge_x$ crystals ($0.02 \le x \le 0.07$, Czochralski technique, volume 35mm x 125 mm) which can optimize synchrotron radiation instrumentation [9]. Due to their relatively broad diffraction pattern, but high reflectivity at high energy radiation (100 *keV*) they are well suited for use as monochromators in measurements of samples with lower qualities [10].

In spite of the work of Smither, the present authors and others, the application of gradient (delta) crystals seems still to be restricted to a few examples, mainly due to the problems with the growing (and bending!) of these crystals. The development of multilayer optics, however, is much more promising at present. The periodic structure enables multilayers to diffract X-rays in a fashion analogous to a crystal, cp. Fig. 6.

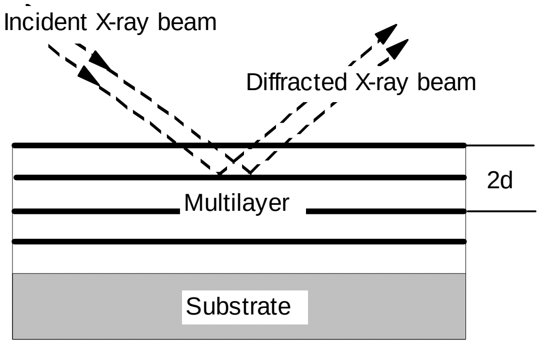

Fig. 6 Multilayer diffraction at angle $_B$ *according to the Bragg law n = 2d sin* $_B$.

So they act like lamella crystals, but offering advantages: they can be engineered to achieve best performance for specific wavelengths, they can be bent to act as collimating or focussing optics. All this becomes possible because layer pairs of tailored thickness can be realized, even with a variation of the period with the depth, resulting in so-called “super mirrors“ (or depth-graded multilayers; the name “mirror“ is somewhat misleading), allowing one to increase the angular or energy range of reflection [11].

The application of a variation of the period along the surface, i.e. of a laterally graded multilayer, has been reported in a paper “Parallel-beam coupling into channel-cut monochromators using curved multilayers“ by Schuster and Göbel [12]. The aim was an improved adaptation of divergent incident radiation to a high-resolution so-called “Bartels monochromator“ [cp.13]. The authors used a W/Si multilayer with 50 layer pairs, fabricated by sequential sputtering on a thin Si substrate, which then was bent to the shape of a parabola. In order the Bragg equation there to be fulfilled at each point, the multilayer period had to be varied between 4 and 5 nm in accordance with an appropriate formula (there is no linear dependence!). As a result CuK radiation incident with an opening angle of 0.4^0 was converted to a beam with less than 0.03^0 divergence (the acceptance angle of the Ge(022) monochromator was about 0.003^0), the measured gain in intensity being 6, but far below an estimated theoretical value. As the authors assume, this might be due to deviations of the multilayer shape from the ideal parabolic curvature. But inaccuracies in the multilayer itself might be an additional reason for this discrepancy.

As suitable periods of synthetic multilayers 1.5 - 10 nm were mentioned, corresponding to Bragg angles between 1.4^0 and 0.2^0: the application of multilayers seems to be restricted to long wavelength X-rays.

The 100 years since the detection of X-rays have brought a wide spectrum of applications for this kind of rays. Apart from the huge importance in medicine, they were used for several methods of material testing including structure analysis of crystals, semiconductors and amorphous substances. Several varieties of monochromators came into use. All of them, however, were based on von Laue's interpretation as electromagnetic waves and following the Bragg equation for the diffraction angle.

But about half a century no real attempts were made to use X-rays for realizing optics, comparable to that of light. For several applications it would be desirable to have instruments employing shorter wavelengths as light. As an example, the resolution of VLSI (very large-scale integrated) circuits, which is restricted by the wavelength of light, could be reduced theoretically by a factor of thousand.

4. Conclusions

For various reasons including the resolution in VLSI circuits and intensity amplification, the development of real X-ray optics is necessary. Within the methods described in this review the gradient (Delta) crystal seems to be one of the most suitable instruments to approach this goal.

Unfortunately the technique of growing and bending of those crystals did not made a significant progress in the last time. And it is the huge challenge for crystal growers to overcome these problems.

References

[1] Arkadiev V.A., Bzhaumikhov A.A., Gorny H.-E., Langhoff N., Schmalz J., SPIE Vol. **2859**, 220(1996).

[2] Gao N., Ponomarev I.Yu., Xiao Q.F., Gibson W.M., Carpenter D.A., Appl. Phys. Lett. **69**(11), 1529(1996); Li P.-W., Bi R.-Ch., J. Appl. Cryst. **31,** 806(1998).

[3] Arndt U.W., Duncumb P., Long J.V.P., Pina, L., Inneman A., J. Appl. Cryst. **31,** 733(1998).

[4] Lengeler et al., Appl. Phys. Lett. (to be published).

[5] Smither R.K., Rev. Sci. Instrum. **53**(2), 131(1982).

[6] Smither R.K., Fernandez P.B., Rev. Sci. Instrum. **63**(2), 1755(1992).

[7] Bradaczek H., Hildebrandt G., Uebach W., Abstracts XII ECM Moscow, Vol.**1**, 304(1989).
Bradaczek H., Hildebrandt G., Uebach W., Acta Cryst. A**49**, C374(1993).

[8] Penzel St., Kleesen H., Neumann W., Cryst. Res. Technol. **32**(8), 1137(1997).
Moshkin S.V., Boldyreva O.M., Ivanova T.I., Kuz'mina M.A., Shakhverdova I.P., Petrashen P.V., Kyutt R.N., Bradaczek H., J. Crystal Growth **172**, 226(1997).

[9] Erko A., Veldkamp M., Gudat W., Abrosimov N.V., Rossolenko S.N., Shekhtman V., Khasanov S., Alex V., Groth S., Schröder W., Vidal B., Yakshin A., J. Synchrotron Rad. **5**, 239(1998).

[10] Keitel S., Retsch C.C., Niemöller T., Schneider J.R., Abrosimov N.V., Rossolenko S.N., Riemann H., Nucl. Instr. and Meth. in Phys. Res. A414, 427(1998).
Keitel S., Malgrange C., Niemöller T., Schneider J.R., submitted to Acta Cryst. A.

[11] Joensen K.D., Hoghoj P., Christensen F., Gorenstein P., Susini J., Ziegler E., Freund A., Wood J., Proc. SPIE, Vol. **2011**, (1993).

[12] Schuster M., Göbel H., J. Phys. D: Appl. Phys. **28**, A270(1995).

[13] DuMond J.W.M., Phys. Rev. **52**, 872(1937).