Reflection of waveguided X-rays in two-dimensional nanostructures

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The internal reflection of an excited X-ray waveguide mode in a synthetic nanostructure, defined by electron-beam lithography, has been measured. In this device, the X-ray beam is first coupled into a conventional vertical thin-film waveguide structure and then reflected laterally at the quasi-one-dimensional edge of the waveguiding layer. The reflectivity of the quasi-one-dimensional interface has been recorded under simultaneous excitation of the (vertical) waveguide mode. The experiment constitutes an important step towards the production of a coherent nanometre-sized X-ray point source by two-dimensionally defined waveguide structures.

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1. Introduction

X-ray diffraction, spectroscopy and imaging with beam sizes in the sub-micrometre range are rapidly evolving fields of research. Progress is fuelled not only by the availability of new highly brilliant sources, but also by novel X-ray optics. In recent years, thin-film X-ray waveguide structures have been designed and proven to deliver coherent X-ray beams efficiently at the exit of the device, with vertical cross sections in the nanometre range, but with lateral extensions still defined by conventional macroscopic slits (line beam) (Feng *et al.*, 1993; Zwanenburg *et al.*, 1999). The divergence and the coherence of the beam has been used to image one-dimensional test structures by lens-less projection phase-contrast microscopy (Lagomarsino *et al.*, 1997). Furthermore, X-ray diffraction with a spatial resolution of ~ 100 nm in one dimension has been demonstrated (Di Fonzo *et al.*, 2000). We have recently shown that even beam widths of less than 100 Å can be achieved by the use of X-ray waveguides in the so-called resonant beam coupling devices (Pfeiffer *et al.*, 2001).

However, for many nano-microscopy and nano-spectroscopy applications, a two-dimensionally confined point beam instead of a one-dimensional line beam is needed. Clearly, a setup of two conventional planar waveguide structures in series cannot lead to a two-dimensionally confined X-ray beam. This is particularly due to the fact that the beam diverges in the direction of the surface normal of the planar waveguide after exiting the guiding layer. We are therefore following a new approach, in which the beam is channelled in two-dimensions by a laterally and vertically patterned nanostructure defined by state-of-the-art electron-beam lithography techniques, thus generalizing the concept of resonant beam coupling to two dimensions.

2. Properties of the device

As the first step towards the realisation of such a device, we have fabricated a thin-film guiding structure consisting of a

resonant X-ray beam coupling section combined with a well defined lateral boundary within the plane of the guiding layer (GL). Furthermore, we demonstrated the possibility of channelling an internally guided X-ray beam through total reflection at the lateral quasi-one-dimensional internal interface. The boundary consisted, in this case, of a strip of germanium of height h (1350 Å), width w (~ 800 Å), length l (2 mm) and index of refraction n ,¹ as shown in the electron micrograph of Fig. 1. On the right-hand side of the figure, the device corresponds to the conventional structure of a hard X-ray waveguide. In this case it consists of a low-density guiding layer made of spin-coated polymethylmethacrylate (PMMA) resist, of thickness $d = 1350$ Å, on top of a Si substrate coated with Cr of thickness 1600 Å. To confine the waveguide modes, the patterned resist was coated with 50 Å of Cr under normal incidence, and then subjected to two evaporation steps to deposit Ge (of thickness 800 Å) under an oblique angle of 85° to cover the side wall.² A substantial quantity of Ge deposited on top of the core was removed by reactive ion etching to provide a defined cap layer. The waveguide was not damaged by this process, since the etching process was stopped before reaching the guiding layer itself.

The guided modes are resonantly excited by shining a parallel beam onto the waveguide under grazing incidence at a set of discrete angles α_{in} . The internal distribution of the field intensities $|E(z)|^2$ is then given by the boundary conditions at the interfaces. In the case of the incoming beam being parallel to the germanium-coated edge of the guiding layer, $\varphi_i = 0$, we find the conventional waveguide effects, e.g. cusps in the plateau of total external reflection (see Fig. 2). In a simplistic argument, photons become trapped under resonance conditions in the guiding layer, propagating over an active coupling

¹ The refraction index for X-rays is given as $n = 1 - \delta - i\beta$, where δ is proportional to the electron density of the corresponding material.

² A LION LV-1 (Leica Microsystems Jena) system at 2.5 keV electron energy was used.

Figure 1

Transmission electron microscopy image of the waveguide structure. Right: 50 Å Cr/1300 Å PMMA (guiding layer GL)/1600 Å Cr/Si substrate. Left: 1600 Å Cr/Si substrate. Middle: Ge strip (width $w \simeq 800$ Å, height $h = 1350$ Å, length $l = 2$ mm).

length (in our case ~ 500 – 1000 μm) parallel to the surface, and are therefore more likely to be absorbed (Tien & Ulrich, 1970). The enhanced absorption loss then leads to a pronounced decrease in the reflectivity. At the end of the structure, photons trapped in the guiding layer exit the thin-film structure, leading to a coherent beam with a vertical cross section (z direction) corresponding to the thickness of the guiding layer and a divergence given by the full width at half-maximum (FWHM) of the Fourier transform of the excited standing wavefield. This can occur if the footprint of the beam is separated from the edge of the sample by not much more than the active coupling length.

The divergence and shape of the exiting guided mode, the angular acceptance and the absolute number of supported modes, can all be controlled by the structural and geometric parameters, and varied within a certain range (Pfeiffer *et al.*, 2000; Zheludeva *et al.*, 1995). Finally, the farfield pattern $I(\alpha_f)$ can be calculated from the electric field inside the waveguide structure by

$$I(\alpha_f) = I_0 \left| \int_{-\infty}^{+\infty} E_y(z) \exp[ik_0 \sin(\alpha_f)z] dz \right|^2. \quad (1)$$

The exiting intensity of the excited waveguided modes can conveniently be illustrated by measuring the intensity integrated over $\pm 0.03^\circ$ around the horizon ($\alpha_f = 0$) as a function of the angle of incidence α_i ; see Fig. 2. Note that the two curves in Fig. 2 corresponding to the reflected and the waveguided intensities, respectively, were both recorded at the condition $\varphi_i = \varphi_f = 0$ (see Fig. 1 for the definition of the angles). Inclining now the angle φ_i (around the surface normal) to a certain value while adjusting α_i to the condition of mode excitation (e.g. $\alpha_i = \alpha_{i,TE0} = 0.071^\circ$), the mode resonantly excited in the

Figure 2

Simulated (line) and measured (open circles) reflectivity, and waveguided mode (full circles connected by line) corresponding to Fig. 1 (20 keV X-rays).

guiding layer GL internally propagates along the direction of φ_i and finally hits the PMMA/Ge boundary interface. Provided that φ_i is smaller than the critical angle for total reflection of X-rays impinging on the PMMA/Ge interface ($\alpha_{c,PMMA \rightarrow Ge} = 0.10^\circ$), we expect a strong reflected signal at $\varphi_f = \varphi_i$.³ Indeed, we were able to observe the quasi one-dimensional reflectivity over a large range of angles $\varphi_f = \varphi_i$ (see below).

3. Experimental results

The experiment was carried out at the bending-magnet beamline ROBL/BM20 of ESRF, with an Si[111] double-monochromator adjusted to an X-ray energy of 20 keV. We used a beam size at the sample of 0.1 mm (vertical) $\times$ 0.1 mm (horizontal) (although only a small part of it was actually accepted by the sample) and a fast scintillation detector mounted on the detector arm at a distance of 680 mm.

Before discussing the reflectivity curve, however, we first turn to the farfield intensity distribution of the internally reflected modes. To verify if the shape of the field of a resonantly excited mode is conserved after total internal reflection, we have measured the farfield pattern $I(\alpha_f)$ for two angles of incidence, $\alpha_{i,TE0} = 0.071^\circ$ and $\alpha_{i,TE1} = 0.076^\circ$, corresponding to the zero- and first-order excitation of resonant modes, while φ_i and φ_f were adjusted to provide total internal reflection ($\varphi_i = \varphi_f = 0.072^\circ$); see Fig. 3. One can clearly see the symmetric nearly Gaussian shape of the TE0 mode (Fig. 3 top) with a maximum centred at the horizon of the sample at $\alpha_f = 0$, while the maxima of the TE1 farfield pattern occur symmetrically at $\alpha_f = \pm 0.037^\circ$. Even more surprising is the good agreement of the measured data with the calculated farfield intensity distribution of the excited TE0/TE1 modes. Note that the internal reflection $\Delta\varphi = \varphi_i + \varphi_f = 0.144^\circ$ was not included in the calculation. Obviously, the reflection simply deflects the beam

³ This angle can be calculated by $\alpha_{c,PMMA \rightarrow Ge} = (\alpha_{c,Ge}^2 - \alpha_{c,PMMA}^2)^{1/2} = 0.10^\circ$ according to the values obtained for $\alpha_{c,PMMA}$ (0.066°) and $\alpha_{c,Ge}$ (0.121°) from fitting the measured reflectivity data shown in Fig. 2.

Figure 3

Simulated (line) and measured (open circles) farfield intensity of the internally reflected ($\varphi_i = \varphi_f = 0.072^\circ$) waveguided mode [top: TE0 ($\alpha_i = 0.071^\circ$); bottom: TE1 ($\alpha_i = 0.076^\circ$)] for 20 keV X-rays.

and the associated farfield pattern into the corresponding direction, without any observable deformation of the intensity distribution (other than the change in direction). This result clearly underlines the possibility of producing devices using reflections in two dimensions, even with the limited precision of current lithographic processes. Importantly, we note the advantage that in this geometry the otherwise disturbing primary beam which passes the sample at $\alpha_f = -\alpha_i$ and $\varphi_f = -\varphi_i$ is now well separated from the exiting mode by a tilt angle of $\Delta\varphi = 0.144^\circ$ out of the plane of reflection. However, the specular reflected beam (SB) is still present because of the unavoidable leakage of the mode through the cap layer, and occurs in Fig. 3 for angles $\alpha_f = \alpha_i$.

Having looked at the farfield intensity distribution $I(\alpha_f)$ for fixed values of α_i , φ_i and φ_f , we can now turn to the intensity $I(\varphi_i)$ as measured in a lateral reflectivity scan ($\varphi_f = \varphi_i$) under the condition of mode excitation (fixed α_i , e.g. $\alpha_i = \alpha_{i,TE0} = 0.071^\circ$); see Fig. 4. The measured reflectivity curve can be compared with simulations of 20 keV X-rays impinging from inside the PMMA onto a PMMA/Ge interface. From the critical angle $\alpha_{c,PMMA \rightarrow Ge}$, the density of the Ge strip is determined to be $\sim 9\%$ smaller than the literature value for bulk Ge. Furthermore, the simulation indicates a root-mean-square (r.m.s.) roughness of $\sigma = 22 \text{ \AA}$. Concerning the absolute flux throughput obtained, we have measured an absolute number of photons of $\sim 800 \text{ counts s}^{-1}$ at the bending magnet BM20 (ESRF) for the excited TE0 mode and a tilt angle $\Delta\varphi$ of 0.144° . Although the value for that measurement is very low, we are convinced that it can be increased by at least a factor of 10, or even 100, by further optimizing the design and the fabrication of the structures. In particular, the fact that the device only accepts an incident-beam cross section of $\sim 2 \times 3 \text{ \mu m}$, which is the projection of the active coupling length in the direction of the incident beam, can be overcome by using appropriate prefocussing optics. Finally, a four to five orders of

Figure 4

Simulated (line) and measured (open circles) reflectivity $I(\varphi_i)$ ($\varphi_f = \varphi_i$) in lateral geometry of the internally guided X-ray waveguided mode [$\alpha_i = 0.071^\circ$ (TE0), α_f integrated, 20 keV X-rays].

magnitude increase in flux throughput can be anticipated in an experiment using both an improved device and an undulator beamline.

4. Summary

In summary, we have demonstrated that resonantly excited X-ray modes inside an X-ray waveguide device can be reflected off a lateral edge defined by electron-beam lithography and consecutive oblique evaporation of a high-Z material, in this case germanium. Obviously, the experiment presented here can be used to characterize such a synthetic quasi-one-dimensional interface. More importantly, the results pave the way for the design of future X-ray optical nanostructures. To date, X-ray multilayers represent a powerful tool in X-ray analysis. In the future, two-dimensionally patterned nanostructures may be used to tailor X-ray optical properties. The observation of total internal reflection resulting from the density contrast between the polymer and the evaporated material at the edge of the guiding layer demonstrates the feasibility of both lateral and vertical guidance of an X-ray beam by multiple reflections. This result is an important step towards the production of a two-dimensionally confined nanometre-sized X-ray beam. The fabrication of two parallel edges may provide the basis to excite a second resonance in the lateral plane while maintaining the resonance conditions of the vertical waveguide structure. Alternatively, curved quasi-one-dimensional interfaces, as can presently be defined by the same electron-beam lithography method (using an appropriate mask aligner), may be employed to focus the beam geometrically in the lateral direction.

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