X-ray waveguiding studies of ordering phenomena in confined fluids
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Chapter 3

A tunable x-ray waveguide

In this chapter we present a planar x-ray waveguide with a tunable air gap as the guiding medium. Discrete transverse-electric modes excited in the air gap propagate almost undisturbed. Filling the air gap with a fluid allows for studies of ordering phenomena in a confined geometry. Since the guided modes are mainly confined to the guiding layer, background scattering from the plates is very low. Starting from the propagation characteristics of the modes in the empty waveguide, requirements on the x-ray source and on the positioning accuracy of the plates are derived. The construction of the waveguide is described and measurements of the far-field angular distributions of intensity exiting the waveguide are presented which illustrate the waveguide's properties.

3.1 Introduction

We have designed a device which confines a fluid between two plates in a gap tunable down to tens of nanometers. The distance is set by a combination of piezo-driven actuators and optical interferometry. The structure of the confined fluid is studied using a x-ray beam incident on the device as shown in Fig. 3.1a. Since at x-ray wavelengths the refractive index of the fluid is higher than that of the confining plates, the device acts as a waveguide for x-rays and part of the incident beam is confined to the fluid-filled gap.

Because the field of a guided mode is evanescent within the plate material, the background of scattered intensity from the sample environment is very small. In addition, the sample material is positioned in a wave field which is coherent in the
confining direction and has a known amplitude and phase at each point. Variations in the sample's density then give rise to mode coupling effects as discussed in the previous chapter. While in optical transmission technology such intermodal scattering phenomena are generally undesired, here they are exploited for the detection of density variations within the confined sample (see chapter 6). Scattering experiments can be performed both in and out of the plane of the waveguide, see Figs. 3.1a and 3.1b.

To excite a single mode, the incident beam should have a sufficient spatial coherence. This is achieved when the source is sufficiently small and far away from the waveguide. The source should also have a high intensity because the waveguide entrance intercepts only a small part of the beam in the vertical direction. Both requirements are met at a high-brilliance undulator of a third-generation synchrotron radiation source.

For the device to function as a waveguide and sample container, it should have extremely flat and parallel plates. The gap has to be set and monitored with nanometer precision. This chapter describes how this is achieved. In section 3.2 we treat the propagation of x rays through a planar waveguide and discuss the requirements imposed on the x-ray source and the device. In section 3.3, the technical layout of the device is given. Results of waveguiding experiments are presented in section 3.4.

3.2 X-ray waveguiding

3.2.1 Mode excitation and propagation

We consider the propagation of transverse-electric (TE) modes [8] through a waveguide as depicted in Fig. 3.1. The bottom plate, having a much larger diameter than the upper one, acts as both as pre-reflector and a post-reflector. The plates are horizontal, the plane of incidence of the beam is vertical. The electric field vector is perpendicular to the plane of incidence, i.e., in the horizontal plane. The modes have a standing-wave character across the gap and are evanescent in the confining plates. The wavelength of the x rays is of the order $\lambda \approx 0.1$ nm and the reflection angles are grazing (typically less than 0.1°). Where applicable, the small angle approximation is used in trigonometric relations.
Figure 3.1 Schematic of the waveguide and the scattering geometry in (a) side view and (b) top view. The fluid (shaded) is confined in a gap of width $W$. The angles of incidence and exit are $\theta_i$ and $\theta_e$. The reflections from the bottom plate in front of the waveguide entrance and behind the exit are indicated as well. The detector is rotatable in the vertical and horizontal planes. Angles and distances are not to scale.
Figure 3.2 The reflection of a plane wave from (a) a single surface and (b) two parallel surfaces. Due to interference of the direct and reflected waves, a standing wave pattern is formed above the surface, see (a). The presence of a parallel surface at a node of the standing-wave pattern makes the wave propagate in the direction parallel to the surfaces. The internodal spacing is related to the wavelength and the angle of incidence $\theta_i$ as shown. Angles and distances are not to scale.
A tunable x-ray waveguide

First consider the reflection of a plane wave from the bottom plate only. Interference of the incident and reflected waves results in a standing-wave field above the surface as shown in Fig. 3.2a. For angles of incidence $\theta_i$ well below the critical angle for total reflection the phase change upon reflection is $\pi$ and the field profile is approximately given by

$$\phi(x) = \sin(k_0\theta_i x),$$

with $k_0 = 2\pi/\lambda$ the wave number, $\lambda$ the wavelength, and $x$ the coordinate along the direction normal to the surface. The field has a fixed node at the surface ($x = 0$) and tunable nodes above it. The node spacing ($\lambda/2\theta_i$) is varied by changing the angle of incidence at fixed photon energy.

If a second plate is positioned at one of the nodes as shown in Fig. 3.2b, a waveguide is formed and the standing-wave field becomes a particular propagating mode of the waveguide. The $m^{th}$ mode is the one having $m$ nodes between the plates (not counting the two nodes at the plate surfaces). This mode occurs at angles of incidence $\theta_i$ equal to

$$\theta_m = \frac{(m + 1)\lambda}{2W},$$

where $W$ is the distance between the plates and $m = 0, 1, 2, ...$. The corresponding field profile is given by Eq. (2.4)

$$\Psi_m(x, z) = \phi_m(x) e^{-i\beta_m z},$$

where $\Psi_m(x, z) \equiv E_y(x, z)$. The mode $\phi_m(x)$ given by

$$\phi_m(x) = \sin(k_0\theta_m x).$$

Here, the coordinate $z$ is parallel to the plates, in the direction of propagation ($z = 0$ at the waveguide entrance). The propagation constant $\beta_m = k_0 \cos \theta_m \simeq k_0(1 - \theta_m^2/2)$ for mode $m$ is the component of the wave vector in the direction of propagation.

The waveguide only supports modes at angles $\theta_m$ smaller than the critical angle for total reflection $\theta_c$ which, far from absorption edges and for an interface with air, is given by [17]

$$\theta_c \simeq \sqrt{2\delta}$$

$$= \lambda\sqrt{n_e r_e / \pi},$$
where $n_e$ is the average electron density in the plate material and $r_e$ the classical radius of the electron. Modes at larger angles are radiation modes which are not confined and are lost through absorption in the plate material. Substituting $\theta_m = \theta_c$ in Eq. (3.2), we find that the maximally allowed mode number equals $m_{\text{max}} \simeq 2W \theta_c/\lambda - 1$. Using Eq. (3.5) in the latter expression, one sees that $m_{\text{max}}$ is independent of the wavelength in the x-ray regime and only a function of $W$ and of the average electron density $n_e$. The minimum distance $W_{\text{min}}$ between the plates at which wave propagation may occur, is the distance at which just the zero$^{\text{th}}$ mode is supported ($m_{\text{max}} = 0$). Hence, $W_{\text{min}} \simeq \lambda/2\theta_c$, again independent of the wavelength. For example, SiO$_2$ plates at a distance of 650 nm support modes up to number $m_{\text{max}} \simeq 31$. Their minimum distance for single-mode propagation equals $W_{\text{min}} \simeq 21$ nm. It is interesting to compare the latter value with the one for the equivalent optical waveguide consisting of a SiO$_2$ guiding layer surrounded by air (the x-ray waveguide turned inside out). The critical angle for total reflection at $\lambda \simeq 500$ nm is $\theta_c = 50^\circ$ and one has $W_{\text{min}} \simeq 210$ nm. Hence, reducing the wavelength by a factor of 5000 does not result in a correspondingly smaller value of $W_{\text{min}}$. This, of course, has its origin in the refractive index being much closer to one at x-ray wavelengths, yielding a much smaller value for $\theta_c$ of $\sim 0.1^\circ$.

We now consider the case that the upper plate is positioned not on a node of the wave field but somewhere in between two nodes. The same situation arises if the angle of incidence deviates somewhat from a mode angle: $\theta_i \neq \theta_m$. In that case, the intensity of the incident field redistributes itself over all possible modes, guided modes and radiation modes. Let us denote the wave field at position $(x, z)$ of the waveguide as $\Psi(x, z)$. At the entrance the wave field is given by

$$\Psi(x, 0) = \sin(k_0 \theta_i x).$$

(3.6)

Taking the orthogonal modes $\{\phi_m\}$ as a basis, one has the following Fourier expansion for the wave field:

$$\Psi(x, 0) = \sum_{m=0}^{m_{\text{max}}} c_m(\theta_i) \phi_m(x),$$

(3.7)

where the summation is restricted to the guided modes only (the radiation modes are assumed to be absorbed in the cladding). The expansion coefficients are given
by

\[ c_m(\theta_i) = \frac{2}{W} \int_0^W \Psi(x, 0)\phi_m(x)dx \]

\[ = \frac{2(-1)^m \theta_m \sin(k_0\theta_iW)}{k_0W} \left( \frac{\theta_i^2 - \theta_m^2}{\theta_i^2 - \theta_m^2} \right), \] (3.8)

where use has been made of Eqs. (3.4) and (3.6). Upon propagation along \( z \), each mode \( m \) changes its phase by a factor \( \exp(-i\beta_mz) \). Within the waveguide, the field is therefore given by

\[ \Psi(x, z) = \sum_{m=0}^{m=\text{max}} c_m(\theta_i)\phi_m(x)e^{-i\theta_mz}, \] (3.9)

with the coefficients \( c_m(\theta_i) \) given by Eq. (3.8). It is readily verified that \( c_m(\theta_n) = \delta_{mn} \), with \( \delta_{mn} \) the Kronecker delta. The total power of the wave field is proportional to the intensity integrated over the cross section of the waveguide:

\[ P(\theta_i) = \int_0^W |\Psi(x, z)|^2 dx \] (3.10a)

\[ = \sum_{n,m} c_n(\theta_i)c_m(\theta_i) \int_0^W \phi_n(x)\phi_m(x)dx \]

\[ = \frac{W}{2} \sum_n c_n(\theta_i)^2, \] (3.10b)

where the proportionality constant is taken to be unity. The power in mode \( m \) is given by \( P_m(\theta_i) = (W/2) \cdot c_m(\theta_i)^2 \) and the fraction \( F_m(\theta_i) \) of incident power transferred to mode \( m \) at angle \( \theta_i \) equals

\[ F_m(\theta_i) = \frac{P_m(\theta_i)}{P(\theta_i)} \]

\[ = \frac{c_m(\theta_i)^2}{\sum_n c_n(\theta_i)^2}. \] (3.11)

For angles equal to a mode angle: \( F_m(\theta_n) = \delta_{mn} \). The angular distribution of \( F_m(\theta_i) \), which is centered around \( \theta_i = \theta_m \), has a full width at half maximum (FWHM) equal to the angular mode spacing \( \lambda/2W \).

The above expressions for the mode angles and the wave field are valid under the assumption that the field at the plate surfaces is exactly zero. In reality, the field penetrates the plates over a limited depth interval. The evanescent wave field has a \( 1/e \) decay depth \( \xi_m \approx k_0^{-1}(\theta_i^2 - \theta_m^2)^{-1/2} \) which varies typically from 6 -7
nm for the lower modes to 13 nm for modes located at angles \( \sim 0.02^\circ \) below \( \theta_c \). For a gap width of, e.g., 650 nm, the contribution of the evanescent wave field is negligibly small for most modes, which justifies our assumption. Only for plate distances down to \( W_{\text{min}} \), or for mode angles close to \( \theta_c \), does the confining material carry a substantial fraction of the wave amplitude. In these cases, more elaborate expressions for the wave field have to be used (see chapter 4).

The modes emerging from the waveguide are identified by measuring the angular distribution of intensity at a large distance behind the waveguide exit. The far-field amplitude diffracted from the exit plane at angle \( \theta_c \) is given by adding Eqs. (2.48) and (2.49):

\[
A(\theta_i, \theta_e) = \left( \frac{i}{R\lambda} \right)^{1/2} e^{-i\kappa R} \int_0^W \Psi(x, L) \left( e^{-i\kappa \theta_0 x} - e^{i\kappa \theta_e x} \right) dx, \quad (3.12)
\]

where \( L \) is the length of the waveguide, \( \Psi(x, L) \) the wave amplitude across the exit plane and \( R \) the distance from the waveguide exit to the detector. The variable \( \theta_i \) in Eq. (3.12), refers to the implicit \( \theta_i \)-dependence of \( \Psi(x, L) \). The two phase factors in the integral account for the path length differences between waves emitted from different positions within the exit plane, the first one relating to the directly emitted waves and the second one relating to the waves post reflected from the bottom surface (the minus sign takes the phase change over \( \pi \) into account, which means that the reflection is assumed to be perfect). We substitute in Eq. (3.12) the expression for \( \Psi(x, L) \) as given by Eq. (3.9) and perform the integration. This leads to the following expression for the intensity per unit angle in the far field:

\[
I(\theta_i, \theta_e) = |A(\theta_i, \theta_e)|^2 = \frac{W^2}{R\lambda} \left| \sum_{m=0}^{m_{\text{max}}} c_m(\theta_i) c_m(\theta_e) e^{-i\beta_m L} \right|^2, \quad (3.13a)
\]

where both \( c_m(\theta_i) \) and \( c_m(\theta_e) \) are given by Eq. (3.8), with \( \theta_i \) and \( \theta_e \) as arguments. It is readily verified that \( I(\theta_m, \theta_n) = \delta_{mn} W^2 / R\lambda \). As expected, a single mode \( m \), excited at the entrance at an angle of incidence \( \theta_i = \theta_m \), will emerge from the exit at the same angle \( \theta_e = \theta_m \). Experimental evidence for a properly functioning waveguide is provided by measuring a two-dimensional contour plot of the intensity \( I(\theta_i, \theta_e) \) as a function of both in- and outgoing angles \( \theta_i \) and \( \theta_e \). The plot will show a sequence of maxima along the diagonal, which correspond to excitation of the consecutive modes.
So far we considered the propagation of modes through an empty waveguide. Filling the device with a homogeneous fluid will bring the refractive index of the guiding medium closer to that of the confining planes. This in turn lowers the critical angle for total reflection $\theta_c$ at the interface. Let the refractive indices of the confining and guiding media be $n_1 = 1 - \delta_1$ and $n_2 = 1 - \delta_2$, with $\delta_1 > \delta_2$. Equation (3.5) has to be modified into

$$\theta_c = \sqrt{\frac{2(\delta_2 - \delta_1)}{1 - \delta_2}} \approx \sqrt{2(\delta_2 - \delta_1)}.$$  

The presence of, e.g., dimethylformamide (DMF) between SiO$_2$ plates lowers $\theta_c$ by 25%. Consequently, the total number of guided modes and the minimum gap width for guided wave propagation are lowered and raised by 25% and 20%, respectively. Other properties of the waveguide hardly change, apart from absorption in the fluid. If, however, density variations are present in the liquid, then mode coupling takes place and the contour plot of $I(\theta_i, \theta_e)$ displays off-diagonal peaks. This is the subject of chapter 6.

### 3.2.2 Requirements on the x-ray source

We discuss the source characteristics and the specifications for the parallel-plate geometry required for proper waveguiding. Numerical examples are given assuming an x-ray wavelength of $\lambda = 0.0930$ nm (photon energy 13.3 keV) and a gap width of $W = 650$ nm, which are typical values in our experiments.

The photon beam should have sufficiently high energy that it can easily pass through a fluid-filled waveguide of a few mm length. On the other hand, as the beam size is much larger than the gap width, one wants to avoid transmission of the beam through the sides of the confining plates, causing an unwanted background of transmitted intensity (for more details, see chapter 4). A photon energy of $\sim 13.3$ keV is a reasonable choice. At this energy the $1/e$ decay depth in SiO$_2$ is 0.6 mm and in, e.g., DMF 9.1 mm. Within a waveguide of 5 mm length about 58% of the beam is transmitted by DMF and 0.02% by the SiO$_2$ plates.

The x-ray beam is generated by the undulator source of beamline ID10A [24] at the European Synchrotron Radiation Facility (Grenoble, France) and passed through a monochromator with narrow bandwidth before it enters the waveguide. The incident beam is partially incoherent in the transverse directions due to the
spatial extent of source and in the longitudinal direction due to the non-zero bandwidth of the monochromator. The degrees of coherence in these directions affect the characteristics of mode propagation through the waveguide in different ways. It is customary to express the degree of coherence as a length scale over which the wave fronts can be considered to be effectively a monochromatic plane wave. We define the coherence lengths along the vertical ($v$) and horizontal ($h$) directions as

$$\xi_v = \frac{\lambda D}{\sigma_v} ; \quad \xi_h = \frac{\lambda D}{\sigma_h} ,$$

(3.15)

with $D$ the distance between the source and the waveguide entrance and $\sigma_v$ and $\sigma_h$ the vertical and horizontal source sizes (FWHM). At the ID10A beamline, $D = 45$ m, $\sigma_v = 23.6$ $\mu$m and $\sigma_h = 928$ $\mu$m. Hence, $\xi_v = 177$ $\mu$m and $\xi_h = 4.5$ $\mu$m. As the waveguide accepts in the vertical direction only a thin slice of the beam having a width $2W$ (the factor two arising from the pre-reflection), the condition $\xi_v \gg 2W$ is always fulfilled and the beam can be considered to be fully coherent along this direction. In the horizontal direction, which lies in the plane of the waveguide, the waveguide accepts all of the beam. Therefore, the beam is incoherent along this direction. Full coherency in both directions is only attained if a pinhole is placed in front of the waveguide, which limits the beam size to less than $\xi_h$. The longitudinal coherence length $\xi_l$ is given by

$$\xi_l = \frac{\lambda}{\Delta \lambda} \cdot \lambda,$$

(3.16)

with $\Delta \lambda/\lambda$ the monochromator bandwidth. The (111) reflections of the Si and diamond crystals which were used as monochromators in our experiments have bandwidths of $\Delta \lambda/\lambda = 1.4 \times 10^{-4}$ and $5 \times 10^{-5}$, yielding coherence lengths of $\xi_l = 0.66$ $\mu$m and $1.86$ $\mu$m, respectively. Coherency along the direction of propagation is achieved if $\xi_l$ is much larger than the difference in total path length travelled by the wavefronts belonging to two different modes. For a given mode $m$, the wavefronts travel over a distance equal to $L/\cos \theta_m$, with $L$ the length of the waveguide. The maximum path length difference (PLD) is that between the zero$^n$th-order mode and the highest-order mode with its mode angle close to the critical angle for total reflection: $PLD_{\text{max}} \simeq L(1/\cos \theta_c - 1/\cos \theta_0) \approx L(\theta_c^2 - \theta_0^2)/2$. For a waveguide of length $L = 5$ mm and a typical critical angle of $\theta_c \simeq 0.12^\circ$, we find $PLD_{\text{max}} \simeq 10$ nm. We conclude that the longitudinal coherency condition $\xi_l \gg PLD_{\text{max}}$ is easily met, even for modes far apart. This is due to the small angles at which the x rays
bounce against the confining surfaces, making path length differences extremely small.

3.2.3 Requirements on the waveguide

The range of incidence angles presented to the waveguide is approximately equal to the ratio \( \sigma_v/D = 0.5 \mu \text{rad} \). This range is much smaller than a typical angular mode spacing \( \Delta \theta_m = \lambda/2W \approx 70 \mu \text{rad} \). Therefore, a single mode \( m \) is excited, provided the incidence angle \( \theta_i \) is exactly tuned to the corresponding mode angle \( \theta_m \). In the absence of a vertically focusing element between source and waveguide, which is the case in our experiments, the condition \( \sigma_v/D \ll \Delta \theta_m \) is equivalent to the above condition \( \xi_v \gg 2W \).

The angle of incidence \( \theta_i \) is to be set with an accuracy sufficiently high that a single mode is excited at the waveguide entrance. Suppose \( \theta_i \) deviates slightly from the mode angle \( \theta_m \) by \( \varepsilon \). Expanding the fraction \( F_m(\theta_i) \) of power transferred to mode \( m \), as given by Eqs. (3.8) and (3.11), one obtains

\[
F_m(\varepsilon) \approx 1 - \left( \frac{1}{4\theta_m^2} + \frac{k_0^2W^2}{3} \right) \cdot \varepsilon^2
\]

\[
= 1 - \left( \frac{1}{4(m+1)^2\pi^2} + \frac{1}{3} \right) \cdot k_0^2W^2 \cdot \varepsilon^2
\]

\[
\approx 1 - \frac{1}{3} k_0^2W^2 \cdot \varepsilon^2,
\]

(3.17)

The second term is the fraction of power lost to modes other than mode \( m \). For the latter fraction to be less than \( p \), the angle of incidence should deviate from the mode angle \( \theta_m \) by less than

\[
|\varepsilon| \lesssim \frac{\sqrt{3p}}{k_0W}.
\]

(3.18)

For a power loss of at most 2\%, i.e. \( p = 0.02 \), the angle \( \theta_i \) has to be set with a precision \( |\varepsilon| < 5.6 \mu \text{rad} = 0.3 \text{ millidegree} \). Alternatively, if \( \theta_i \) is set to a mode angle \( \theta_m \) but the real gap width deviates from the corresponding value \( W \) by an amount \( \Delta W \), then

\[
\frac{\Delta W}{W} \leq \frac{\sqrt{3p}}{(m+1)\pi}.
\]

(3.19)

The allowed deviation \( \Delta W \) decreases proportionally with the node spacing of the standing wave field across the gap. In order to let mode \( m = 0 \) pass with less than
2% loss we have to set the gap width with a relative precision of $\Delta W/W \leq 0.078$. For $m = 30$, a precision $\Delta W/W \leq 2.5 \times 10^{-3}$ is required. For a gap width $W = 650$ nm, these numbers correspond to $\Delta W$ values of 51 and 1.6 nm, respectively. Not only should the incidence angle $\theta_i$ and plate distance $W$ be set within narrow limits, they should be kept stable within these limits over time. Whereas stability is readily achieved for the angular setting, the plate distance may drift due to temperature variation. Continuous monitoring and adjustment of the gap width are required.

The upper plate may be tilted with respect to the lower plate along the direction of propagation. In a tapered waveguide the field profile across the gap will be different from that of an unperturbed mode (see also chapter 5). However, for sufficiently small tilt angles the perturbation will be small. Let the widths at the entrance and exit be $W$ and $W' = W + \theta t L$, with $\theta_t$ the tilt angle which is assumed to be positive. The mode angle for mode $m$ changes continuously from the value $\theta_m = (m + 1)\lambda/2W$ to $\theta'_m = (m + 1)\lambda/2W'$. If the mode angle change $\theta_m - \theta'_m$ is much less than the initial mode angle spacing $\lambda/2W$, than one expects the field profile belonging to mode $m$ to remain essentially undisturbed. Substituting the expressions for $\theta_m$ and $\theta'_m$ and $W'$ into the condition $\theta_m - \theta'_m \ll \lambda/2W$ we obtain

$$|\theta_t| \ll \frac{W}{(m + 1)L},$$

where the modulus of $\theta_t$ accounts for possible negative tilt angles. The condition on the tilt angle, Eq. (3.20), is most stringent for the highest mode number. Assuming $m = m_{\text{max}} \approx 30$, $L = 5$ mm, $W = 650$ nm, we find that the tilt angle $\theta_t$ has to be much smaller than 4 $\mu$rad. Let us take $\theta_t = 0.5 \mu$rad. Over a length $L$ of 5 mm, the corresponding change in gap width is 2.5 nm. Therefore, one should be able to set the plates parallel with nanometer control.

The propagation of modes may also be affected by the roughness of the plate surfaces. Due to scattering from surface imperfections, intensity is coupled to other modes. The fields of higher-order modes scatter more strongly, because their first and last antinodes are closer to the plate surfaces. The distribution of scattered intensity over the modes is dependent on the type of roughness and on the length scale over which the roughness is correlated [25]. Here we take the empirical approach of measuring the mode propagation characteristics for variously prepared waveguide surfaces with known roughness as measured by atomic force
3.2.4 Detection of far-field angular intensity distribution

The detector measuring the far-field angular distributions of intensity exiting the waveguide should be capable of resolving the maxima associated with two neighboring modes as well as the subsidiary diffraction maxima arising from the finite size of the gap, see Eqs. (3.8) and (3.13b). A reasonable angular acceptance is \(1/10^4\) of the angular mode spacing, e.g. \(\sim 10 \mu\text{rad}\) for a mode spacing of \(\lambda/2W \approx 100 \mu\text{rad}\). Resolving modes for gap sizes larger than \(\sim 2000\) nm becomes increasingly more difficult. Moreover, for the excitation of a single mode at the entrance of such a large gap, the setting of the angle of incidence is no longer precise enough.

We have performed experiments using a NaI scintillation detector and a position sensitive CCD camera. Adjustable slits in front of the scintillation detector define the horizontal and vertical angular acceptance. When the detector is placed at a distance of 2 m from the exit of the waveguide, the opening angle is typically \(\sim 10 \mu\text{rad}\). The far-field angular intensity distributions are measured by scanning the detector along the \(\theta_e\) axis. The CCD camera (Sensicam [26], air cooled, 12 bit dynamic range) contains a chip with 1280x1024 pixels of 6.7 \(\mu\text{m}\) with an area of 8.6\(\times\)6.9 mm\(^2\). At 2 m from the exit of the waveguide, the chip covers an angular range of 0.24° horizontally and 0.19° vertically with a resolution of \(\sim 3 \mu\text{rad}\). When the camera is mounted onto a translation stage, it is possible to cover a larger range of exit angles. The use of the CCD camera instead of the scintillation detector speeds up experiments by a factor 50.

3.3 Apparatus

3.3.1 Design

A waveguiding device has been built which meets the requirements derived in the previous section, see Fig. 3.3. It fits onto a double tilting stage with 170 \(\times\) 170 mm\(^2\) mounting area (HUBER 5203.2 [27]), enabling the horizontally positioned waveguiding device to be tilted over small angles along the azimuths parallel and
perpendicular to the beam direction. The former rotation serves to change the angle of incidence $\theta$, in steps of 0.5 millidegree, the latter rotation is used to set the waveguide plates parallel to the horizontal plane. The tilting stage is part of a horizontal diffractometer set-up described elsewhere [24].

The waveguide plates are polished fused-silica disks with $< \lambda/20$ surface accuracy (General Optics [28] and Melles-Griot [29]). The disks were coated with a semi-transparent metal film, enabling measurement of the plate distance by optical interferometry, see section 3.3.2. The top and bottom plates are 6 mm thick and have diameters of 5 and 25 mm, respectively. The diameter of the bottom plate is large enough for excitation of the lowest mode TE$_0$ at the waveguide entrance and for the post-reflection behind the waveguide exit, which requires a length of $W/\tan(\theta_0) \approx 9$ mm at either side of the waveguide. The bottom plate is clamped onto a three-point mount and its surface is in the center of rotation of the double tilting stage.

For the coarse approach of the upper plate to the bottom plate, a tripod of piezo-driven inchworm© motors (Burleigh IW-700 [30]) is used. The upper plate is pulled by springs against the motor heads. Three radial V-shaped grooves in the plate, in which the motor heads rest, keep the plate centered irrespective of temperature variations. The strength of the three pulling springs is chosen such that the force on the motorheads does not exceed the maximum allowed axial load. Simultaneous movement of the motors lowers the upper plate. By moving the motors independently we are able to eliminate a possible tilt angle between the top and bottom plate. The inchworm motors have a large travel range (6 mm) and can make steps as small as 4 nm. A lateral distance of approximately 100 mm between the motors yields a theoretical angular resolution of 0.04 $\mu$rad. The stepping action of an inchworm motor, however, is not exactly reproducible. Small adjustments to the plate distance are therefore made by an additional piezo-driven translator (Physik Instrumente S-310.10 [31]) which is incorporated into the upper mounting plate (see Fig. 3.3). The device is controlled by a digital DC-power supply providing a translation range of 1 $\mu$m with a theoretical resolution of 0.016 nm, which is well below the required positioning accuracy. The necessary repeatability in the plate distance and the tilt angle is achieved by monitoring these by optical interferometry and, if needed, by manually adjusting the piezo-motors at intervals of a few seconds.
Figure 3.3 Side view of waveguide set-up in cross section. Main parts: (A) tripod of inchworm motors, (B) piezo-driven vertical translator, (C) support rods, (D) clamp for plate, (E) top and bottom plates, (F) $90^\circ$-deflecting mirror for white-light beam. The beam path for optical interferometry is indicated as well. The horizontal bar indicates a distance of 50 mm.
3.3.2 Measurement of plate distance and tilt angle

The interferometric method of distance measurement is based on the analysis of Fringes of Equal Chromatic Order (FECO) which occur under illumination of an interferometer with white light [32]. The light, produced by a 100 W lamp (Müller Elektronik-Optik [33]), is transported through a flexible lightguide (Müller Elektronik-Optik [33]) to a condenser lens which collimates the beam. An infrared filter in the beam path reduces the heat load. A 90°-deflecting mirror directs the beam onto the parallel-plate system which forms the optical interferometer, see Fig. 3.4. At each point across the plates, transmission occurs whenever half a wavelength of the light matches the distance between the semi-transparent metal films deposited onto the plates. In the transmitted light, the interferometer surface is imaged 2:1 onto the entrance of a spectrometer (Yobin-Yvon HR460 [34]) by means of an imaging fiber bundle consisting of thousands of 10 μm fibers forming a square area of 4 × 4 mm² [34]. The vertical entrance slit of the spectrometer selects a small stripe, typically 10 μm wide and 4 mm high, from the image. The incident light is dispersed in wavelength by the spectrometer grating and imaged onto the chip of a digital CCD camera (Photometrics [35]) as fringes, see Fig. 3.5.

The set-up is sensitive to tilt angles along the direction selected with the entrance slit. When the fringes appear tilted as in Fig. 3.5a the inchworm motors are used to remove the tilt (Fig. 3.5c). To detect and correct a possible tilt angle along the direction perpendicular to the previous one, the image on the entrance slit is rotated 90° by twisting the fiber bundle around its axis at the spectrometer side. The tilt is measured and eliminated in both directions iteratively.

The distance $W$ between the parallel plates is given by

$$W = \frac{m\lambda_m}{2n},$$  \hspace{1cm} (3.21)

where $\lambda_m$ is the wavelength in vacuum corresponding to the fringe of integer order $m$ and $n$ the refractive index of the medium between the plates (e.g., a fluid). Equation (3.21) has two unknowns: the distance $W$, to be determined, and the fringe order $m$. The latter is found by writing Eq. (3.21) for two neighboring fringe orders $m - 1$ and $m$ and subtracting them:

$$m = \frac{\lambda_{m-1}}{\lambda_{m-1} - \lambda_m},$$  \hspace{1cm} (3.22)
Figure 3.4 Schematic of the optical interferometer based on the principle of fringes of equal chromatic order. The interferometer consists of parallel SiO$_2$ plates on which are sequentially deposited a semi-transparent metal film and a SiO$_2$ spacer layer. The parts function as indicated in the text.

It follows that

$$W = \frac{\lambda_{m-1}\lambda_m}{2n \cdot (\lambda_{m-1} - \lambda_m)}.$$  \hspace{1cm} (3.23)

Hence, the distance $W$ is most easily deduced from the measured values for the wavelengths of two neighboring fringes. The larger the distance between the fringes, the smaller is $W$ (Fig. 3.5d). The lower limit of the distances in our set-up is mainly determined by the spectral sensitivity of the CCD camera which becomes zero for wavelengths smaller than $\lambda = 400$ nm. Assuming an air gap with $n = 1$, the smallest measurable distance is $\lambda/2 = 200$ nm according to Eq. (3.21). This limit can be overcome by the use of spacer layers on top of the metallic films, as in the surface force apparatus [36]. The spacer layer is transparent for optical wavelengths at normal incidence and acts as a reflecting medium for the x rays at glancing incidence. However, the relation between the gap width $W$ and the wavelength $\lambda$ now becomes more complex than in Eq. (3.21). Let us assume that the spacer layer at either side of the gap has thickness $d$ and refractive index $n'$. 
Figure 3.5 Example of fringe patterns taken on a parallel-plate system with aluminium coatings and SiO$_2$ spacer layers of 640 nm thickness: (a) for a system with a tilt angle $\theta \approx 80 \text{ mrad}$ and an average plate distance of $W \approx 7.9 \text{ m}$, (c) for the plates at a distance $W = 6.8 \text{ m}$ with the tilt angle eliminated, and (d) for the plates at closer distance $W = 390 \text{ m}$. Panel (b) shows the wavelengths corresponding to the fringes in (a). The small excursion in the fringes in panel (d) indicates the presence of a dust particle in the gap or an irregularity in one of the plate surfaces.
The relation is given by \[36\]

\[
\tan \left( \frac{2\pi n W}{\lambda} \right) = \frac{(1 - r^2) \sin(2\pi n d/\lambda)}{2r - (1 + r^2) \cos(2\pi n d/\lambda)},
\]

with \(r \equiv \frac{(n' - n)}{(n' + n)}\). Substituting in Eq. (3.24) the measured values for the wavelength \(\lambda_m\), we obtain the value of \(W\). A suitable spacer material is SiO\(_2\), because it acts as a protective layer for the metal coating, preventing it from being oxidized in the air and from being chemically attacked by the fluid. On our silica substrates we deposited, by thermal evaporation, first an aluminium layer of \(\sim 30\) nm thickness and then a SiO\(_2\) layer of 640 nm thickness. It is for this system, with air gap, that we obtained the optical fringe patterns of Fig. 3.5. From the fringe distances shown in panels (c) and (d), we deduced a gap width \(W\) of 6.7 \(\mu\)m and 390 nm, respectively. The sharpness of the fringes is determined by the reflectivity of the metal [32], which for aluminum is 0.92 at \(\lambda \simeq 500\) nm. We have also prepared silica substrates with only a metal layer on top (no spacer layer). In this case, chromium was chosen and layers of 30 nm thickness were deposited by thermal evaporation. Chromium has excellent adhesion to silica. The resulting root-mean-square surface roughness, as measured by atomic force microscopy over an area of \(11 \times 11\) \(\mu\)m, was \(\sim 0.5\) nm for the chromium surfaces and \(\sim 1\) nm for the surfaces of the SiO\(_2\) spacer layers. For both surfaces the x-ray waveguiding properties were found to be very similar at least up to the eighth mode.

The fringe patterns were measured using a 150 l/mm spectrometer grating. With the CCD chip having an area of \(11 \times 11\) mm\(^2\), the field of view in the wavelength-dispersing direction is 159 nm wide. For an entrance slit opening smaller than the 22 \(\mu\)m pixels of the CCD-camera, a wavelength resolution of 0.32 nm is achieved (pixel size \(\times\) grating dispersion). This corresponds with an uncertainty in the plate distance of \(\sim 2\) nm in the system for which the patterns of Fig. 3.5 were taken. For the lower-order modes this is much smaller than the maximum allowed deviation according to the criterion Eq. (3.19). Over a fringe length of 4 mm, a distance change of 2 nm corresponds with a tilt angle of 1 \(\mu\)rad. Hence, the criterion for undisturbed mode propagation, as given by Eq. (3.20), is easily met.

During the experiment the plate distance may drift slowly because of temperature variations. In a fluid-filled waveguide, slowly changing capillary forces may also be at work. Usage of the motorized tilting stage on which the wave-
guide is mounted, causes vibrations which, however, decay quickly and leave the system again in the initial state. It is therefore necessary to monitor the optical fringes continuously and to keep a fringe at its preset position, mostly by using the piezo-driven translator.

The optically determined plate distance is systematically slightly larger than the value deduced from the measured angular mode spacing in the x-ray waveguiding experiments (e.g., 20 nm larger for a gap of ~ 400 nm). The reason for this apparent discrepancy is an additional phase change of the light upon reflection from the metallic films, which is not accounted for in Eqs. (3.21) and (3.24).

When the waveguide is filled with a fluid, the fringes shift to higher wavelengths. From the magnitude of the shift the optical refractive index of the fluid can be determined using Eq. (3.21). Deformations of the fringes reveal inhomogeneities in the refractive index, i.e., in the density or composition of the fluid film.

### 3.4 X-ray waveguiding experiments

We have tested the performance of the waveguide set-up in a series of x-ray scattering experiments. Here, we present the results of one such experiment on an empty waveguide with SiO\textsubscript{2} surfaces and having a length \( L = 5 \) mm. A photon energy of 13.33 keV (\( \lambda = 0.09301 \) nm) was selected. A beam of 0.1 mm diameter was defined by a pinhole in front of the waveguide set-up. The beam intensity passing through a gap of 650 nm was typically \( 4.3 \times 10^8 \) photons/s. TE modes with increasingly higher mode number were excited at the entrance by changing the angle of incidence \( \theta_i \) in small steps. At each \( \theta_i \) value the intensity \( I(\theta_i, \theta_e) \) diffracted from the waveguide exit was recorded as a function of the exit angle \( \theta_e \) with the use of a position-sensitive detector consisting of a fluorescent screen followed by a CCD camera (Sensicam, cooled, 12 bit, 6.7 \( \mu \)m pixel size [26]). In this detector, positioned at 2390 mm distance from the waveguide exit, each pixel defines an acceptance angle of 2.8 \( \mu \)rad in the vertical plane. The CCD camera covers a \( \theta_e \)-range of 0.16°. In the perpendicular direction, the CCD detector images the distribution of intensity scattered in the horizontal plane. In the absence of a scattering medium inside the waveguide it is just the horizontal beam profile that is imaged.

The intensity distribution \( I(\theta_i, \theta_e) \) was measured at a gap width \( W = 650 \) nm,
Figure 3.6 Logarithmic contour plots of the intensity diffracted from the exit of the waveguide as a function of $\theta_i$ and $\theta_e$. The x-ray wavelength $\lambda = 0.0930$ nm and the waveguide length $L = 4.85$ mm. (a) Measured intensity distribution for a waveguide with plates at a distance $W = 650$ nm. (b) Intensity distribution calculated with Eq. (3.13b) for the same plate distance.

for $\theta_i$ and $\theta_e$ values ranging from zero to 0.037°. The results are shown in Fig. 3.6a in the form of a logarithmic grey-scale plot. The plot was made by integrating the image, taken at each incidence angle $\theta_i$, horizontally over the width of the beam. The resulting distribution in $\theta_e$ represents a single vertical slice in the contour plot. Repeating this procedure for a sequence of $\theta_i$ values with increments of 1 millidegree leads to the plot shown, in which grey levels were also generated by interpolation between points. At each angle $\theta_i$, the data collection time for the CCD was 0.5 s. It took in total 100 s to record the data of Fig. 3.6a. Within the angular range shown, eight discrete TE modes are clearly seen along the diagonal. The observed angular mode spacing of 0.0041° is consistent with a gap width of 650 nm. The subsidiary maxima and minima at either side of the main maxima are Fraunhofer diffraction features arising from the finite width $W$ over which the field amplitude is non-zero.

A comparison was made with calculations of $I(\theta_i, \theta_e)$ using Eq. (3.13b). The only parameters entering the calculations are $\lambda, W$ and $L$, for which the experimental values are taken. The results of the calculations are also displayed as a
contour plot, see Fig. 3.6b. The measured and calculated patterns look strikingly similar, apart from a small constant background making the minima in the experimental data less pronounced. The similarity demonstrates that the propagation of modes through our waveguide is close to ideal. In particular the interference between modes, seen as a modulation of the intensity minima along the diagonal, is reproduced by the calculations. These modulations arise from the phase factor \( \exp(-i\beta_m L) \) in Eq. (3.13b), which is mode-dependent. For illustration, we show in Fig. 3.7 two vertical slices from the contour plots (with background subtracted), one at an incidence angle equal to a mode angle, \( \theta_i = \theta_5 \), and one in between two mode angles, \( \theta_i \approx (\theta_5 + \theta_6)/2 \). In the last case, the standing wave field at the entrance of the waveguide does not match the field of any of the modes. Hence, the intensity is redistributed over all the modes (guided modes and radiation modes) but mainly the nearest-neighbor modes (TE\(_5\) and TE\(_6\)). The minima in the experimental diffraction pattern are slightly less pronounced than the calculated minima (dashed curve). A good fit is obtained after convoluting the calculated distribution \( I(\theta_i, \theta_e) \) with a Gaussian distribution in \( \theta_i \) having a FWHM of 1.27 millidegree (solid curve). The calculated and measured intensity distributions differ slightly in the tails. For an improved fit at these angles, the effects of the evanescent wave and of surface roughness would have to be taken into account, see chapter 4.

The x-ray waveguide with tunable air gap has been developed by us with the purpose of using it for structural investigations of confined fluids. If the fluid is homogeneous, then the mode propagation characteristics should be the same as for the empty waveguide, apart from a reduction of the total number of guided modes (see section 3.2.1). We verified this by inserting a tiny drop of DMF between the plates. Indeed, we observed essentially the same \( I(\theta_i, \theta_e) \) distribution as in Fig. 3.6. Apparently, the fluid's meniscus at the entrance and exit planes of the waveguide make a sufficiently large angle with the propagation direction that refraction effects are negligible. For inhomogeneous fluids, e.g. a colloidal suspension of SiO\(_2\) particles in DMF, we have observed very different \( I(\theta_i, \theta_e) \) distributions, having some very strong off-diagonal peaks. These can be attributed to ordering effects in the colloid as is discussed in chapter 6. For example, consider a colloidal solution in the waveguide with the colloidal particles arranged in layers parallel to the plates. The wavefronts corresponding to a single propagating mode
Figure 3.7 Diffraction patterns from the exit of the waveguide for different angles of incidence: (a) $\theta_i \approx \theta_5$ and (b) $\theta_i \approx (\theta_5 + \theta_6)/2$. The dashed curves are patterns calculated with the use of Eq. (3.13b). The solid curves have been obtained by convoluting Eq. (3.13b) with a Gaussian intensity distribution in $\theta_i$, see text.
will be scattered from the oscillatory refractive index profile into other propagating modes with mode numbers being uniquely determined by the oscillation period (see section 6.2). Hence, wall-induced layering effects in fluids [3] can be sensitively detected by exciting a single mode at the waveguide entrance and measuring the distribution of intensity over the modes emerging from the waveguide exit. Random density variations within the medium give rise to scattering of intensity into all propagating modes. In either case the multiple scattering phenomena inherent to the waveguiding geometry result in significant mode coupling even for low refractive index contrast.