#### Appendix 1

# The estimation of SOF<sub>4</sub> and SO<sub>2</sub>F<sub>2</sub> refractive indices

For this calculation the equation for molar refractivity A (or atomic refractivity in the case of monatomic substances) will be  $\iota$  sed. It is defined as (Born & Wolf, 1980):

$$A = \frac{4\pi}{3} N_m \, o_{\cdot} = \frac{RT}{p} \frac{n^2 - 1}{n^2 + 2} \tag{A1.1}$$

where:

 $N_m = 6.022045 \text{ x } 10^{23} \text{ mol}^{-1} \text{ is the Avogadro rumber}$ 

 $\alpha$  is the mean polarisability,

R is the molar gas constant,

T is the temperature,

p is the pressure.

In equation (A1.1), the compressibility of the gases and the nonlinearity of the index of refraction have been neglected; that is, it is assumed that coefficient B(T) (equation 4.4) is equal to zero. This assumption matches reality for pressures below 1 atm at room temperatures. When a molecule is made up of  $N_1$  atoms with molar refractivity of  $A_1$ , and  $N_2$  atoms of molar refractivity  $A_2$ , then the molar refractivity of the molecule is (Born & Wolf, 1980)

$$A = A_1 N_1 + A_2 N_2 \tag{A1.2}$$

Suppose that, in order to calculate the refractive index of  $SOF_4$ , we assume that it is the combination of one  $SOF_2$  molecule and one  $F_2$  molecule. The index of refraction of  $F_2$  is 1.000214 at 101325 Pa and 0 °C measured at 589.4 nm (Landolt - Börnstein, 1962). As molar refractivity is constant for a particular gas and does not depend on temperature and pressure, equation (A1.1) can be used to calculate the index of refraction at 25 °C when it is known at 0° C. This gives the refractive index of 1.000196 at 25 °C and 101325 Pa. Also, 589.4 nm is close to 632.99 nm and it can be assumed that the index of refraction is the same at both wavelengths. The index of refraction of  $SOF_2$  is 1.00062166 at 101325 Pa and 25 °C (this work, chapter 4). Using these values and equations (A1.1) and (A1.2), the value of 1.000818 is obtained for the refractive index of  $SOF_4$ .

A similar approach is used to calculate the refractive index of  $SO_2F_2$ ; as the combination of a molecule of  $SO_2$  and a molecule of  $F_2$ . The refractive index of  $SO_2$  is 1.000628 at 101325 Pa and 25 °C, calculated from 1.000686 at 101325 Pa and 0 °C at 589.6 nm (Gray, 1972). This gives 1.000877 for the refractive index of  $SO_2F_2$  at 101325 Pa and 25 °C.

In the determination of the refractive indices of  $SOF_4$  and  $SO_2F_2$ , it has been assumed that the gases are linear. Any non-linear contributions to the refractive indices are likely to be small: for example, in the case of  $SF_6$  the contribution of B(T) is only  $8x10^{-6}$ , although  $SF_6$  is a very non-linear gas in terms of refractive index and compressibility in comparison to other gases. Therefore, although the values of 1.000877 and 1.000817 for the refractive indices of  $SO_2F_2$  and  $SOF_4$ , respect vely, may not be exact, the conclusion that their refractive indices are higher than the refractive index of  $SF_6$ , which is 1.00070296 at 1 atm and 25 °C at 632.99 nm, should be valid.

#### Appendix 2

# Data acquisition programs for the refractive index measurement

Programs, "fill\_in" and "pump\_out", were written for the acquisition of data during the refractive index measurement when the gas is bled in and out of the invar tube. These programs control the A/D converter, receive data from the A/D converter, store the data and receive data from the VAISALA barometer via a serial port. The A/D converter is in a CONTEC ADC-10 card. It is a 12-bit A/D converter with a maximum speed of 30 ksamples/s. With this program the sampling speed is 15 ksamples/s on an IBM PC compatible computer with INTEL 386SX-16 processor, math cooprocessor and 4 MB memory. This speed is sufficient, because the amplified signal from the photodiode which is a part of the polarisation interferometer (see figure 4.1) is around 1 Hz. The programs were compiled with Borland Turbo C++ ver. 3.0 for DOS. The VAISALA PTB 200A digital barometer sends ASCII string data via the serial port. An interrupt 0x14 from BIOS was used to receive this string.

The photodiode signal I from the interferometer is given by (Born & Wolf, 1980):

$$I = A\cos^2\left(\frac{\delta}{2}\right) \tag{A2.1}$$

where A is a constant that depends on beam intensities and the sensitivities of the photodiode,  $\delta$  is the phase difference between the two beams of the interferometer, and  $\delta$  changes with time as the pressure in the invar tube changes. The programs monitor the maxima and minima of this function. When the signal passes through a quadrature point the computer acquires the pressure information from the VAISALA barometer. If the pressure is

not recorded at the quadrature point the program makes the required correction. The fringe number and pressure are stored on the hard disk.

#### A2.1. FILL\_IN program

The flow chart in figure A2.1 represents the program for fringe counting and pressure. The fringe-intensity signal is the signal from the photodiode (see figure 4.1) measured with a 12-bit analog/digital converter CONTEC in the computer. The program for controlling this converter is part of the FILL\_IN program. It also displays the signal and the pressures. When the measuring system is evacuated, the program is started and the valve is opened.

A driver for the A/D converter was not available for Turbo C++, so a driver was written and is shown in the listing A2.1.

```
// driver for A/D converter on the CONTEC board ADC-10
int
              get_val()
              datal, datah:
unsigned
              val;
imt
              val=outp(port,ox87);
              for(;;)
              datal = inp (port);
              datah = datal & ox80;
                if(datah != ox8o)
                break:
              datah= inp(port);
              datal= inp(port-1);
              datah=datah & oxF;
              val = (datah*256+datal);
              return(val);
```

Listing A2.1. The driver in TURBO C++ for the A/D converter channel 7 in the CONTEC ADC-10 card.

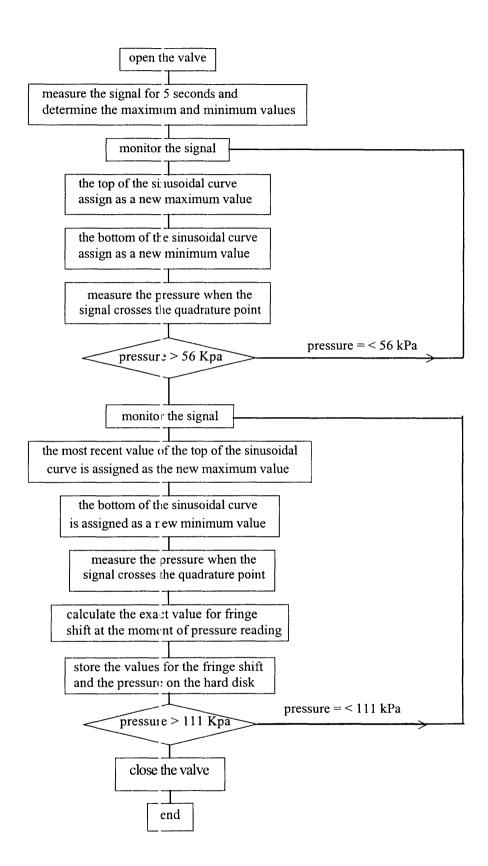


Figure A2.1. The flow chart of the FILL\_IN program for refractivity data acquisition.

The pressure from the VAISALA PTB-200A barometer via the serial interface is read using a BIOS interrupt 0x14. The subroutines for retrieving data from this port are shown in listing A2.2. The data from the barometer are sent as ASCII data. These are taken from the computer at a time determined by the main program (FILL\_IN). The subroutine receive\_line takes 40 successive characters and the actual pressure is extracted in the main program.

```
char receivech(char chs)
               ou.h.ah=2;
               ou.x.dx=COMport;
               int86(0x14, &ou, &out);
               chs=out.h.al;
               return (chs);
}
void receive_line(char line[])
                       receivech(char ch);
               char
                      ch='z';
               int
                      i=o;
               for(i=0; i < 40; i++)
                       ch=receivech(ch);
                       if((ch == '\r')) ch = '?';
                         if(ch=='\xo') ch='?';
               line[i]=ch;
               line[i]='\xo';
               ch=receivech(ch);
               ch=receivech(ch);
}
```

Listing A2.2. The subroutines for receiving a character from the serial interface and for receiving 40 successive characters from the same port.

#### A2.2. PUMP\_OUT program

This program collects data when gas is pumped out of the measuring cylinder of the interferometer. The program is similar to the program FILL\_IN except that it runs in the opposite direction: it starts at 115 kPa and stops at 57 kPa.

#### Appendix 3

## Equations for light propagation in an optical fibre

An optical fibre is a cylindrical dielectric waveguide. If the dielectric constants of the core and cladding are  $\varepsilon_1$  and  $\varepsilon_2$  respectively, then  $\varepsilon_1 > \varepsilon_2$ . The radius of the fibre is  $\rho$  and it can be assumed that the radius of the cladding is infinite and both regions are perfect insulators. If a cylindrical coordinate system is chosen with the z - axis lying along the axis of the fibre, then the solutions for the electric and magnetic components of the electromagnetic wave travelling in the fibre are (Snitzer, 1961):

$$E_{zcore} = A_m J_m(h_1 r) \cos(m \theta + \varphi_m) \exp[i(\beta z - \omega t)]$$
(A3.1)

$$H_{zcore} = B_m J_m(h_1 r) \cos(m \theta + \psi_m) \exp[i(\beta z - \omega t)]$$
 (A3.2)

$$E_{zclad} = C_m K_m(h_2 r) \cos(m \theta + \varphi_m) \exp[i(\beta z - \omega t)]$$
 (A3.3)

$$H_{zclad} = D_m K_m(h_2 r) \cos(m \theta + \psi_m) \exp[i(\beta z - \omega t)]$$
 (A3.4)

where  $\omega$  is the angular frequency;  $\beta = 2\pi/\lambda_g$  is the propagation constant along the axis of the fibre and  $\lambda_g$  is the wavelength along the fibre (see figure A3.1);  $\phi_m$  and  $\psi_m$  are phase factors;  $\beta$ ,  $\phi_m$  and  $\psi_m$  are determined by the boundary conditions. At the boundary  $r = \rho$ , the tangential components of the field are the same on both sides of the boundary.  $J_m(h_1r)$  and  $K_m(h_2r)$  are the Bessel function and the modified Hankel function of the first kind, respectively. The propagation constants  $k_1$  and  $k_2$ , and  $k_3$  are defined by  $k_4$  and  $k_4$  are constants for a fibre, but  $k_1$  and  $k_2$  have different values for each mode)

$$k^2 = \omega^2 \mu \varepsilon \tag{A3.5}$$

$$h_1^2 = k_1^2 - \beta^2 \tag{A3.6}$$

$$h_2^2 = \beta^2 - k_2^2 \tag{A3.7}$$

where  $\mu$  is free-space magnetic permeability.

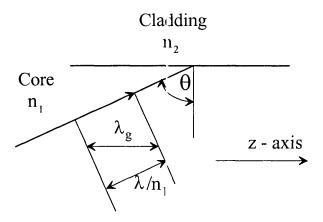


Figure A3.1. Considering geometrical optics propagation, the wavelength  $\lambda_g$  along a fibre (z - axis) of a certain mode is  $\frac{\lambda}{n_1 \sin \theta}$ ;  $\lambda$  is the free-space wavelength.

The transverse components of the fields  $E_r$ ,  $E_\theta$ ,  $H_r$  and  $H_\theta$  can be expressed in terms of  $E_z$  and  $H_z$  (Stratton, 1941 Chapter V). After taking the boundary conditions (4 equations) into account, expressions for the field components in the core are obtained:

$$E_z = J_m(h_1 r) F_c \tag{A3.8}$$

$$E_r = i \frac{\beta}{h_1} \left[ J_m' - P \frac{m J_m}{h_1 r} \right] F_c \tag{A3.9}$$

$$E_{\theta} = i \frac{\beta}{h_1} \left[ P J_{n} - \frac{m J_m}{h_1 r} \right] F_s \tag{A3.10}$$

$$H_z = -\frac{\beta}{\mu \omega} P J_m F_s \tag{A3.11}$$

$$H_r = -i \frac{k_1^2}{\mu \omega h_1} \left[ P \frac{3^2}{k_1^2} J_m' - \frac{m J_m}{h_1 r} \right] F_s$$
 (A3.12)

$$H_{\theta} = i \frac{k_1^2}{\mu \omega h_1} \left[ J_m' - P \frac{h^2}{k_1^2} \frac{m J_m}{h_1 r} \right] F_s$$
 (A3.13)

where

$$P = -\frac{\mu\omega}{\beta} \frac{B_m \cos(m\theta + \psi_m)}{A_m \sin(m\theta + \phi_m)}$$

$$= m \left[ \frac{1}{(h_1 \rho)^2} + \frac{1}{(h_2 \rho)^2} \right] \left[ \frac{J'_m(h_1 \rho)}{h_1 \rho J_m(h_1 \rho)} + \frac{K'_m(h_2 \rho)}{\lambda_2 \rho K_m(h_2 \rho)} \right]^{-1}$$
(A3.14)

$$F_c = A_m \cos(m\theta + \varphi_m) \exp[i(\beta z - \omega t)]$$
 (A3.15)

$$F_s = A_m \sin(m\theta + \varphi_m) \exp[i(\beta z - \omega t)]$$
 (A3.16)

and  $J'_m$  is the first derivative of the Bessel function with respect to its argument. For a multimode fibre there are many values for  $\beta$  (propagation constant along the fibre axis) that satisfy these equations. Each of them represents a propagation mode. For every cross-section geometry and refractive-index profile it may be stated that (Snyder and Love, 1983):

$$n_{cl}\frac{2\pi}{\lambda} < \beta \le n_{co}\frac{2\pi}{\lambda}$$
 (A3.17)

where  $n_{cl}$  and  $n_{co}$  are the refractive indices of the cladding and the core, respectively. The lower limit for  $\beta$ , namely  $n_{cl}\frac{2\pi}{\lambda}$ , is called the modal cutoff (Snyder and Love, 1983). At the cutoff,  $h_2\rho=0$ , and equations A3.6 and A3.7 become

$$\beta = k_2 \tag{A3.18}$$

$$h_{1mj}\rho = \frac{2\pi}{\lambda} \rho \sqrt{n_{co}^2 - n_{cl}^2}$$
 (A3.19)

where  $\lambda$  is the free-space wavelength and  $h_{1m_j}$  is the value of  $h_1$  at cutoff; that is, the *j*th root of the cutoff condition involving the *m*th order Bessel function (Snitzer, 1961). The right-hand term in equation (A3.19) is called the waveguide parameter, the waveguide frequency or the *V*-number of a fibre, and is written as (Snyder and Love, 1983):

$$V = \frac{2\pi}{\lambda} \rho \sqrt{n_{co}^2 - n_{cl}^2}$$
 (A3.20)

For multimode fibres V >> 1 and for step-profile circular single-mode fibres V < 2.405.

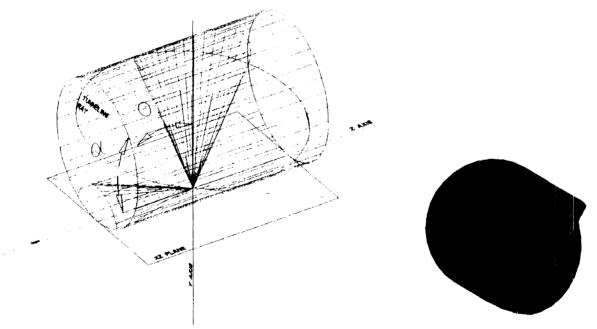


Figure A3.2. Left: Schematic picture of bound (within red semi-cone) rays, refracting (within magenta cone) rays and a sample tunnelling ray. Right: Actual picture within the fibre showing the cones of bound and refracting rays touching along a common edge line. The geometry of the fibre restricts the bound rays to a small section of cone. In reality, the critical angle  $\theta_c$ , and hence the angle of the magenta cone, is larger than depicted in the figure.

The rays that propagate through an optical fibre are called bound rays. Other rays are leaky rays. When a ray hits a point on the boundary core-cladding it will be a bound ray if it comes within a red semi-cone depicted in figure A3.2. Rays in the red cone satisfy the condition

$$0 \le \alpha < \left(\frac{\tau}{2} - \theta_c\right) \tag{A3.21}$$

where  $\theta_c$  is the critical angle for the core-cladding interface, that is,  $\theta_c = \sin^{-1}(n_{co} / n_{cl})$ . A refracting ray in the magenta cone in figure A3.2 is defined within the range

$$0 \le \theta < \theta_c. \tag{A3.22}$$

All other rays are called tunnelling rays and they are defined within the ranges

$$\left(\frac{\pi}{2} - \theta_c\right) \le \alpha \le \frac{\pi}{2}$$
 and  $\theta_c \le \theta \le \frac{\pi}{2}$  (A3.23)

Rays in an optical fibre can also be divided into meridional and skew rays. The meridional rays pass through the axis of the fibre while the skew rays do not pass through the axis. Refracting and bound rays can be either meridional or skew rays, while tunnelling rays are always skew rays.

When light is launched into a fibre all three types of ray (bound rays, tunnelling rays and refracting rays) are present. Refracted rays irradiate their energy into the cladding and the space around the fibre. According to Fresnel's equations, only a small fraction of a refracted ray is reflected back into the fibre. The power carried by refracted rays can be neglected after  $100\rho$  (Pask, 1975).

Exact solutions to equations A3.1-A3.4 require detailed numerical computation. However, some simplifications and approximations can be made in order to obtain useful and relatively simple equations.

For example, two such equations are relevant for the present work, as follows. For a multimode fibre, the number of modes that can propagate through a fibre, is given by (Gloge, 1971; Janata, 1989)

$$N \approx \frac{\rho^2}{2} \left( k_1^2 - k_2^2 \right) = 2 \left( \frac{\pi \rho}{\lambda} \right)^2 \left( n_{co}^2 - n_{cl}^2 \right)$$
 (A3.24)

If incoherent light is launched into a multimode fibre, all modes will be excited and will carry equal amounts of power. In this case the power that travels in the cladding is given by (Gloge, 1971)

$$P_{clad} = \frac{4}{3} \frac{1}{\sqrt{N}} P_{tot}. \tag{A3.25}$$

## Appendix 4

# Evanescent field of totally reflected light at a flat interface

When light arrives at the interface of two materials, it is reflected and refracted according to Snell's law, as shown in figure A4.1.

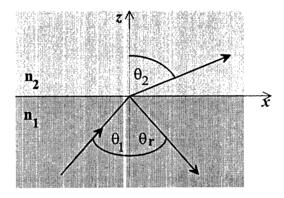


Figure A4.1. Reflection and refraction of an electromagnetic wave;  $n_1 > n_2$ .

Boundary conditions require the tangential field components of the electromagnetic field to be continuous. If the electric field component of the refracted electromagnetic wave in the second medium is  $E_0$  then the equation of the wavefront in the second medium is

$$E_2 = E_0 \exp \left[ -i \frac{2\pi n_2}{\lambda_0} (x \sin \theta_2 + z \cos \theta_2) \right]$$
 (A4.1)

From Snell's law and

$$\cos \theta_2 = \pm \sqrt{1 - \left(\frac{n_2}{n_2}\right)^2 \sin^2 \theta_1},$$
 (A4.2)

we get:

$$E_{2} = E_{0} \exp \left[ -i \frac{2\pi}{\lambda_{0}} \left( x n_{1} \sin \theta_{1} \pm z \sqrt{1 - \left(\frac{n_{1}}{n_{2}}\right)^{2} \sin^{2} \theta_{1}} \right) \right]$$
(A4.3)

Since for total reflection the term in the square root is negative, equation A4.3 needs to be written as

$$E_{2} = E_{0} \exp \left[ -i \frac{2\pi}{\lambda_{0}} \left( x \, n_{1} \sin \theta_{1} \pm iz \, \sqrt{\left(\frac{n_{1}}{n_{2}}\right)^{2} \sin^{2} \theta_{1} - 1} \right) \right]$$
 (A4.4)

or

$$E_2 = E_0 \exp\left[-ix\frac{2\pi}{\lambda_0}n_1\sin\theta_1\right] \exp\left(\pm z\frac{2\pi}{\lambda_0}\sqrt{\left(\frac{n_1}{n_2}\right)^2\sin^2\theta_1 - 1}\right)$$
(A4.5)

The solution with negative sign in the second exponential term is the real solution. It shows that the electric field decays exponentially in the z direction; that is, the direction perpendicular to the interface. This field is called the evanescent field. The first exponential term shows that this field travels along the x-axis in the same direction and with the same phase as the wavefront in the waveguide. The depth of the evanescent-field penetration is defined as the distance over which the amplitude of the field falls to 1/e of its value at the interface, and is given by

$$d_p = \frac{\lambda_0}{2\pi \sqrt{\left[\frac{\sin\theta_1}{\sin\theta_c}\right]^2 - 1}}.$$
 (A4.6)

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129

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