Fluorescence lifetime sensor using optical fiber and optical signal processing

by

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A thesis submitted in conformity with the requirements for the Degree of Master of Applied Science, Graduate Department of Aerospace Science and Engineering, in the University of Toronto

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Abstract

I propose in theory and demonstrate in experiment a fibre optic sensor which measures fluorescent lifetime and fiber length using an all optical signal processing scheme. The scheme uses a double modulation homodyne method to produce auto or cross correlation in frequency domain. Because it eliminates the need for high bandwidth optical detection and electronic processing, it has the potential of superior performance with less complexity than conventional time or frequency domain fluorometers. To demonstrate, fiber length of 50.28 ± 0.27m and fluorescent lifetimes of 28.3 ± 3.5ns and 360 ± 28ns are measured with only 4MHz bandwidth. Furthermore, the fluorescence collection efficiency of multimode fiber is derived for single and double fiber configurations. And the effect of optical fiber on excitation and fluorescence is analyzed. Also, possible application in the cure monitoring of epoxy resin is addressed.

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Abbreviations

AO    acousto-optic
AOD   driver for AOM
AOM   acousto-optic modulator
BS    beamsplitter
BW    bandwidth
DFT   Discrete Fourier Transform
EOM   electro-optic modulator
FT    Fourier Transform
IF    intermediate frequency
IOM   integrated optic modulator
LO    local oscillator
MMF   multimode fiber
NA    numerical aperture
OFDR  optical frequency domain reflectometer
OTDR  optical time domain reflectometer
QY    quantum yield
RF    radio frequency
RU    ruthenium tris-bipyridyl dichloride, hexahydrate
SMF   singlemode fiber
SPA   N-(3-sulfopropyl) acridinium, inner salt
## Contents

1 Introduction  

2 Multimode Fiber  
  2.1 Mode Propagation in Steady-State  
  2.2 Dispersions  
    2.2.1 Modal Dispersion  
    2.2.2 Material Dispersion  
    2.2.3 Waveguide Dispersion  
  2.3 Rayleigh Backscattering  
  2.4 Fresnel Backrefection  
  2.5 Fluorescence Collection by Direct Coupling  
    2.5.1 Liouville's Theorem  
    2.5.2 Collection Efficiency  
    2.5.3 Collection Efficiency of Single Fiber  
    2.5.4 Collection Efficiency of Double Fibers  
  2.6 Fluorescence Collection by Evanescent Coupling  
    2.6.1 Evanescent Excitation  
    2.6.2 Collection Efficiency  

3 Fluorescence System  
  3.1 Fluorescent Response  
  3.2 Determining Lifetime in Time Domain  
  3.3 Determining Lifetime in Frequency Domain  

4 Acousto-Optic Modulator  
  4.1 Acoustic Perturbation  
  4.2 Diffraction Efficiency  
    4.2.1 Raman-Nath AOM  
    4.2.2 Bragg AOM  
  4.3 Modulation Bandwidth  

5 Signal Processing  
  5.1 Signal Flow  
  5.2 Auto and Cross Correlations  
  5.3 Time Domain Responses  
  5.4 Estimation of Parameters  
  5.5 Estimation of Error  

6 Experiment  
  6.1 Experimental Setup  
  6.2 Components  
    6.2.1 Laser
List of Figures

2.1 Fiber tip with an angled cleave ........................................ 12
2.2 Excitation and collection of fluorescence using single fiber ..... 14
2.3 Excitation and collection of fluorescence using double fibers . 14
2.4 Graph of $F(L)$ and $G(L, s)$ ............................................ 16

3.1 3-level model of fluorescence system ................................. 18
3.2 Survey of frequency domain fluorometers ............................ 21

4.1 Diffraction at Bragg angle .................................................. 24
4.2 Diffraction at non-Bragg angle ........................................... 24

5.1 Signal flow diagram ......................................................... 28
5.2 Square-wave excitation and fluorescent response .................. 28
5.3 Simulation of Fresnel backreflection from 50m fiber ............. 31
5.4 Simulation of SPA fluorescence using 50m fiber ................. 32
5.5 Simulation of RU fluorescence using 50m fiber ................... 33

6.1 Schematic of experimental setup .................................... 37
6.2 Pictures of optical (top) and electrical (bottom) equipments ... 38
6.3 Picture of inside AOM ..................................................... 40
6.4 Transfer function of AOM/AOD ........................................ 41
6.5 Chemical formula for SPA (left) and RU (right) ................ 43
6.6 Absorption and emission spectrum for SPA (top) and RU (bottom) 44
6.7 Calibration data for LO's voltage-to-frequency conversion .... 45
6.8 DC-coupled lowpass detector ........................................... 47
6.9 Simpler AC-coupled bandpass detector ............................... 47
6.10 Aligning AOM for first pass (top) and second pass (bottom) ... 47
6.11 Pictures of SPA (top) and RU (bottom) fluorescence ............ 49
6.12 Raw data for Fresnel backreflection from 50m fiber .......... 50
6.13 Raw data for SPA fluorescence using 50m fiber ................. 51
6.14 Raw data for RU fluorescence using 50m fiber ................... 52
6.15 Raw data for Fresnel backreflection from 49m fiber .......... 53
6.16 Fluorescence decay of SPA and Fluorescein ....................... 55
6.17 Fluorescence decay of RU and Fluorescein ....................... 55

B.1 epoxy resins DGEBA and TGDDM ................................... 63
B.2 curing agents DDS and DDM ........................................... 63
Chapter 1

Introduction

Many have used optical fiber sensor and fluorescent lifetime to measure such parameters as temperature, pH, O₂ concentration, etc. The optical fiber allows one to excite and collect the fluorescent signal from a remote medium that may otherwise be difficult to access. The lifetime of fluorescence decay is chosen because the decay mechanism is well understood, is sensitive to its molecular environment, and is mostly independent of the intensity and the wavelength of both excitation and fluorescence. In combining optical fiber and fluorescent lifetime, the sensors represent a significant development not only in fluorescence field, but also in other sensing applications where the parameter of interest can be measured from an optical signal whose profile and mechanism are similar to the fluorescence decay.

Unfortunately, all the sensor implementations, so far, suffers from 3 major problems:

1. The coupling between the optical fiber and the fluorescence has not been studied analytically. The fluorescent coupling is important in determining the amount of fluorescence that is collected by fiber, and in evaluating the performance and potential of optical fiber sensor.

2. The waveguide effect of optical fiber on the excitation and the fluorescence signals has not been quantified. Unlike the free-space which it replaces, the optical fiber generates Rayleigh scattering, Fresnel reflection, dispersions, modal interference, wavelength dependent attenuation, coupling loss, incidental luminescence, etc.

3. Most of signal processing for the lifetime is done electronically in either time or frequency domain, using expensive components and complicated arrangements. The main advantage of electronic signal processing is that amplification, filtering, and other signal conditioning can be done. As the lifetime gets shorter, it requires faster detection and higher bandwidth signal processing. This becomes more difficult to accomplish with the equipments that are currently available.

I address these problems in my thesis with the emphasis on understanding the fundamentals, rather than on reciting a list of possible applications or on performing characterization of actual materials.

In my thesis, I propose in theory and demonstrate in experiment a fibre optic sensor which measures fluorescent lifetime and fiber length using an all optical signal processing scheme. A continuous laser beam is intensity modulated by an acousto-optic modulator and is launched into a multimode fiber. At the far-end,
the optical fiber collects either fluorescent response from which to measure the lifetime or Fresnel backreflection from which to measure the length. The returning signal is intensity modulated again by the modulator and is detected using a lowpass detector. Although the fluorescent lifetime is the main parameter of interest, the fiber length must be also measured because the use of optical fiber inevitably introduces propagation delay.

The first and second problems are addressed by deriving the fluorescence collection efficiency for single and double fiber configurations, and by analyzing the effect of optical fiber on the propagating signals. The results are obtained mainly for multimode fiber, but they can be useful for singlemode fiber as well.

To address the third problem, the signal processing scheme uses a double optical modulation homodyne method to produce auto or cross correlation in frequency domain. The length and lifetime parameters are estimated by data fitting auto and cross correlation model directly in frequency domain. My scheme eliminates the need for wide bandwidth optical detection and high frequency electronic processing, while at the same time, reducing the component count and simplifying the experimental setup. It has the potential of superior performance with less complexity than conventional time or frequency domain fluorometers.
Chapter 2

Multimode Fiber

In this chapter, I will present some basic expressions which are helpful in understanding the workings of multimode fiber (MMF) and occasionally singlemode fiber (SMF). Consider a MMF with core radius \( a \) and index profile

\[
 n(r) = \begin{cases} 
 n_1 \sqrt{1 - 2\Delta \left( \frac{r}{a} \right)^g}, & r < a \\
 n_2, & r \geq a
\end{cases}
\]  

(2.1)

where \( NA = n_1 \sin \theta_{NA} = n_1 \sqrt{2\Delta} = \sqrt{n_1^2 - n_2^2} \) is known as numerical aperture, and \( \theta_{NA} \) is half-angle of acceptance aperture inside the fiber. Two common types of fiber are step-index fiber \((g = 1)\) in which the core index stays constant \( n_1 \) and parabolic-index fiber \((g = 2)\) in which the core index decreases parabolically from \( n_1 \) to \( n_2 \).

2.1 Mode Propagation in Steady-State

At steady-state, optical power can be propagated only in guided mode with \( k_0n_2 < \beta < k_0n_1 \), where \( \beta \) is the propagation constant along the fiber axis. [Marcuse, Ungar, Cheo, Ghatak]. The total number of guided modes is

\[
 N = \frac{\sqrt{V^2}}{g + 2} = \begin{cases} V^2/2, & \text{step} \\
 V^2/4, & \text{parabolic}
\end{cases}
\]  

(2.2)

where \( V = k_0aNA \). Since diffuse source, such as LED and fluorescence, will launch equal power into each mode, fibers with equal number of modes will carry equal amount of power. If \( \beta_m \) is the propagation constant of \( m \)th-order mode, then the number of modes with \( \beta_m < \beta < k_0n_1 \) is

\[
 N_m = N \left( \frac{k_0^2n_1^2 - \beta_m}{k_0^2n_1^2} \right)^{(g+2)/g}
\]  

(2.3)

which can be rewritten for the propagation constant as

\[
 \beta_m = k_0n_1 \sqrt{1 - 2\Delta \left( \frac{N_m}{N} \right)^{g/(g+2)}}
\]  

(2.4)

Since each mode propagates with different velocity \( v_g = d\omega/d\beta_m \), the time that a mode takes to travel distance \( L \) is [Cheo, Marcuse, Ghatak, Gowar]

\[
 t_m = \frac{L}{v_g} = \frac{L}{c} \left( 1 + \frac{g - 2}{g + 2} \Delta \xi + \frac{3g - 2}{2g + 4} \Delta^2 \xi^2 + \cdots \right)
\]  

(2.5)
where $\xi = (N_m/N)^{s/(s+2)}$, and small $\Delta$ is assumed in Eqn 2.4. Ignoring higher order terms, the difference between the fastest time ($\xi = 0$) and the slowest time ($\xi = 1$) is

$$T = \frac{\ln 1}{c} \begin{cases} \Delta, & \text{step} \\ \Delta^2/2, & \text{parabolic} \end{cases}$$ (2.6)

One can also describe propagation conditions in terms of geometric optics. By comparing Eqn 2.4 and $\beta_m = k_0 n_1 \cos \theta_m$ where $\theta_m$ is the angle between ray vector and fiber axis, one obtains [Ungar, Barnoski, Cheo]

$$\frac{\sin^2 \theta_m}{\sin^2 \theta_{NA}} = \left( \frac{N_m}{N} \right)^{s/(s+2)}$$ (2.7)

which explains concisely much of the experimentally observed phenomena in MMF. Furthermore, outside the optical fiber, the far-field angular ($\theta$) dependence and the near-field radial ($r$) dependence of normalized power are [Barnoski]

$$P(\theta) = \left( 1 - \frac{\sin^2 \theta}{\sin^2 \theta_{NA}} \right)^{2/s}, \quad P(r) = 1 - \left( \frac{r}{a} \right)^g$$ (2.8)

where $\theta_{NA}$ is half-angle of fiber aperture in air. As expected, the power distributions for step-index fiber ($g = \infty$) are uniform; that is, $P(\theta) = P(r) = 1$.

### 2.2 Dispersions

In this section, I review only the broadening of rms width ($\sigma$) due to 3 types of dispersion associated with optical fiber: [Cheo, Ghatak]

1. Modal dispersion ($\sigma_1$) is caused by the fact that different mode travels at different speed.
2. Material dispersion ($\sigma_2$) is caused by the fact that refractive index $n(\lambda)$ varies with wavelength.
3. Waveguide dispersion ($\sigma_3$) is caused by the fact that waveguide parameter $V$ depends on $\lambda$.

For MMF, $\sigma_1$ is usually dominant, but $\sigma_2$ makes contribution in fluorescence measurement where source spectral width is large. For SMF, only $\sigma_2$ and $\sigma_3$ are applicable. The dispersions are uncorrelated with one another. And if the fiber is operating linearly, the rms width of total dispersion becomes [Gowar]

$$\sigma^2 = \sigma_1^2 + \sigma_2^2 + \sigma_3^2$$ (2.9)

#### 2.2.1 Modal Dispersion

Fortunately for step-index and parabolic-index MMF, the modal impulse response turns out to be a square pulse [Marcuse]; that is, when an impulse excitation is launched into the fiber, the propagating modes are distributed evenly between the fastest mode and the slowest mode. Then, using Eqns 2.6 and A.53, the rms width of modal dispersion becomes

$$\sigma_1 = \frac{T}{\sqrt{12}} = \frac{\ln 1}{c\sqrt{12}} \begin{cases} \Delta, & \text{step} \\ \Delta^2/2, & \text{parabolic} \end{cases}$$ (2.10)
Since modal coupling tends to reduce pulse broadening in exchange for increased attenuation, the modal dispersion exhibits square-root dependence with distance after travelling critical length of $L_c = 1/4\gamma_\infty$. [Cheo, Gowar]

$$\sigma_1(z) = \begin{cases} \frac{z}{L_c}, & z < L_c \\ \sqrt{z/L_c}, & z > L_c \end{cases}$$ (2.11)

where $\sigma_c$ is the rms width at $L_c$, and $\gamma_\infty$ is steady-state attenuation coefficient.

### 2.2.2 Material Dispersion

The rms width of material dispersion is [Cheo, Ghatak]

$$\frac{\sigma_2}{L\sigma_s} = -\frac{\lambda}{c} \frac{d^2 n(\lambda)}{d\lambda^2}$$ (2.12)

where $n(\lambda)$ is the core index and $\sigma_s$ is the spectral rms width of source. An empirical expression of $n(\lambda)$ and $n''(\lambda)$ for pure silica are [Ghatak]

$$n(\mu) = C_0 + C_1\mu^2 + C_2\mu^4 + C_3p + C_4p^2 + C_5p^3 \pm 10^{-7}$$

$$n''(\mu) = 2C_1 + 12C_2\mu^2 + C_3p'' + 2C_4 (p'p' + pp'') + 3C_5p (2p'p' + pp'')(2.13)$$

where $\mu = \lambda \times 10^6$ is wavelength in [\mu m], $n''(\lambda) = n''(\mu)/\mu m^2$, and

$$p = 1/(\mu^2 - 0.035), \quad C_0 = 1.4508554, \quad C_3 = 3.027 \times 10^{-3},$$

$$p' = -2\mu p^2, \quad C_1 = -3.1268 \times 10^{-3}, \quad C_4 = -7.79 \times 10^{-5},$$

$$p'' = -2p^2 - 4\mu pp', \quad C_2 = -3.81 \times 10^{-5}, \quad C_5 = 1.8 \times 10^{-6}.$$

### 2.2.3 Waveguide Dispersion

Waveguide dispersion is relevant only for SMF. An empirical expression for step-index SMF is [Ghatak]

$$\frac{\sigma_3}{L\sigma_s} = -\frac{n_1\lambda}{c\lambda} \left(0.080 + 0.549(V - 2.834)^2\right), \quad 1.3 \leq V \leq 2.6$$ (2.15)

### 2.3 Rayleigh Backscattering

Consider an impulse excitation of unit energy travelling down an optical fiber. The spatial profile of excitation is simply $E(z) = e^{-\gamma z}$, where $\gamma$ is the total attenuation coefficient due to Rayleigh scattering ($\gamma_\alpha \propto \lambda^{-4}$) and absorption ($\gamma_\alpha$). In $[z,z+dz]$ interval, $\gamma_\alpha E(z)dz$ is lost to scattering, of which only $S\gamma_\alpha E(z)dz$ is collected back into fiber. Assuming Rayleigh scattering is isotropic at least over the acceptance aperture of fiber, the collection efficiency is given as

$$S = \frac{3\gamma_\alpha}{4(g + 1)} \Delta, \quad \text{for MMF [K9, K33, Ungar]}$$ (2.16)

$$= \frac{3\lambda^2}{V^2w^2} \Delta, \quad \text{for SMF [K10, K11, K33]}$$ (2.17)

where $w$ is the spot size of Gaussian profile that approximates SMF's fundamental mode field. Specifically, $S = 3\Delta/4$ for step-index MMF, and $S = 3\Delta \ln V/V^2$ for step-index SMF using Eqn 2.36.

Since the attenuation is same for backward and forward propagation, the backscattered energy arriving at $z = 0$ from the interval $[z,z+dz]$ is $dE_s = S\gamma_\alpha e^{-2\gamma z} dz$. The impulse response $h(t) = dE_s/dt$ is simply the rate of scattered
energy returning back to the launch point. Then, using \(2z = ct/n\) for 2-way propagation in an optical fiber of length \(L\), the impulse and frequency responses of Rayleigh backscattering are [K10, K13, K14, K32]

\[
h(t) = \frac{cS\gamma_s e^{-\gamma ct/n}}{2n}, \quad 0 \leq t \leq \frac{2Ln}{c} \tag{2.18}
\]

\[
H(f) = S\gamma_s \frac{1 - e^{-2AL}}{2A}, \quad A = \gamma + j\frac{2\pi fn}{c} \tag{2.19}
\]

2.4 Fresnel Backreflection

At a cleaved end of MMF as illustrated in Fig 2.1, the cleave angle \(\theta\) determines how much of Fresnel reflection, which is normally produced due to the index difference between \(n_1\) and \(n_0\), is collected back into the fiber. Using Eqn A.63 with small angle approximation (\(\theta, \theta_{NA} < 1\)) or from Ref [K12], the total backreflection coefficient can be written directly as

\[
R = \frac{\phi - \sin \phi}{\pi} \left( \frac{n_1 - n_0}{n_1 + n_0} \right)^2, \quad \cos \frac{\phi}{2} = \frac{\theta}{\theta_{NA}} \tag{2.20}
\]

If \(R_0\) and \(R_L\) are the reflection coefficients respectively at near-end \((z = 0)\) and far-end \((z = L)\), then the impulse and frequency responses of Fresnel backreflection are [K13, K14, K32]

\[
h(t) = R_0\delta(t) + R_L e^{-2\gamma L} \delta \left( t - \frac{2Ln}{c} \right) \tag{2.21}
\]

\[
H(f) = R_0 + R_L e^{-2AL}, \quad A = \gamma + j\frac{2\pi fn}{c} \tag{2.22}
\]

2.5 Fluorescence Collection by Direct Coupling

Coupling between diffuse source and optical fiber has been studied using Liouville's theorem and ray optic assumption, mainly in the context of launching LED into MMF [H1]. By extending the results of LED-to-fiber coupling, I derive the efficiency of collecting fluorescence through MMF, whether the excitation is carried by the same fiber or by another. Others have studied the case of single fiber [H6, H7, H9] and double fibers [H3, H4, H8, H10], all utilizing brute force integration with complicated geometry. My approach yields simpler yet more complete solutions.

2.5.1 Liouville's Theorem

Liouville's theorem uses canonical conjugate variables of Hamiltonian mechanics. For ray optics, it begins with Format's principle on optical path length,
where \( x' = dx/ds, y' = dy/ds \), and \( ds = dx + dy + dz \). This variational problem has well-known solution in terms of Lagrangian \( L(x, y, x', y') \) or in terms of Hamiltonian \( H(x, y, p_x, p_y) = p_xx' + p_yy' - L \), where \( p_x = \partial L/\partial x' = ndx/ds \) and \( p_y = \partial L/\partial y' = ndy/ds \).

The generalized coordinates \((x, y)\) and generalized moments \((p_x, p_y)\) constitute 4-D phase space, in which point \((x, y, p_x, p_y)\) represents an individual ray, density \( \rho(x, y, p_x, p_y) \) represents the local distribution of ray flux, and phase volume \( \Gamma(x, y, p_x, p_y) \) represents the domain of ray ensemble. For a ray distribution propagating along \( \hat{z} \) axis, the phase volume becomes [H1]

\[
d\Gamma = dx \, dy \, dp_x \, dp_y = n^2 \, dA \, d\Omega \tag{2.24}
\]

where \( A \) is normal area and \( \Omega \) is planar solid angle as defined in Eqn A.14.

Liouville's theorem states that, as rays propagate through a lossless optical system according to the equations of motion,

- local density \( (\rho) \) in the neigbourhood of any moving point does not change, and
- local phase volume \( (\Gamma) \) surrounding a fixed ensemble of points does not change even though its boundary and shape may change.

This means that once the ray distribution is established by source, no optical system can decrease the phase volume without excluding some ray points, since by doing so the local density in some region would necessarily increase in violation of Liouville's theorem. The use of lens and other optical components merely changes the shape of phase volume. For uniform \( n \), Liouville's theorem gives

\[
\Gamma = n^2 A \Omega = \text{constant} \tag{2.25}
\]

It is instructive to note that phase volume \( \Gamma \) is proportional to the total number of modes \( N \) as defined in Eqn 2.2.

### 2.5.2 Collection Efficiency

Out of the total rays emitted by a source, an optical fiber can only collect those ray flux which fall within its aperture. If \( \Gamma_s \) and \( \Gamma_f \) are the phase volume of source and fiber, respectively, the collection efficiency of optical fiber is a simple ratio of ray flux,

\[
\eta_c = \frac{\int_{\Gamma_s \cap \Gamma_f} \rho_s \, d\Gamma}{\int_{\Gamma_s} \rho_s \, d\Gamma} = \frac{(A_s \cap A_f)(\Omega_s \cap \Omega_f)}{A_s \Omega_s} \tag{2.26}
\]

assuming that source density \( \rho_s \) and refractive index \( n \) are uniform which is usually the case for diffuse source such as LED and fluorescence.

LED Because the emitting area is smaller than fiber core, LED is usually butt-coupled to MMF [Gowar]. Using \( \Omega_f = \pi \sin^2 \theta_{NA} \) for the fiber's aperture and \( \Omega_s = \pi \) for source emission into 1 hemisphere, the collection efficiency is

\[
\eta_c = \frac{\Omega_f}{\Omega_s} = \frac{NA^2}{n_i^2} \tag{2.27}
\]
Figure 2.2: Excitation and collection of fluorescence using single fiber

\[ A(z) = \pi a^2 (1 + z/b)^2 \]

Figure 2.3: Excitation and collection of fluorescence using double fibers

\[ A(z) = \pi a^2 (1 + z/b)^2 \]
Fluorescence As shown in Fig 2.2, the phase volume of fluorescent source at 
\( z = z \) emitting into 2 hemispheres is \( \Gamma_s = n_0^2 A_s \Omega_s \), where \( A_s = \pi a^2 (1 + z/b)^2 \) and \( \Omega_s = 2\pi \). The phase volume of fiber at \( z = 0 \) is \( \Gamma_f = n_0^2 A_f \Omega_f \) where \( A_f = \pi a^2 \) and \( \Omega_f = \pi \sin^2 \theta_{NA} \). Then, the collection efficiency is

\[
\eta_c = \frac{A_f \Omega_f}{A_s \Omega_s} = \frac{\text{NA}^2}{2n_0^2} \left( \frac{b}{b + z} \right)^2
\]  

(2.28)

2.5.3 Collection Efficiency of Single Fiber

Consider a MMF illuminating a conical volume with half-angle \( \theta_{NA} \) as shown in Fig 2.2. In an absorbing medium, the excitation profile along \( \hat{z} \) axis is \( I(z) = I_0 e^{-\alpha z} \), where \( \alpha \) is bulk absorption coefficient and \( I_0 \) is the initial intensity over the core area \( A_0 \). In \([z, z + dz]\) interval over cross-section area \( A(z) \), \( \alpha I(z) dz \) is absorbed and \( QY \alpha I(z) dz \) is emitted as fluorescence, of which only \( \eta_c QY \alpha I(z) dz \) is collected by the fiber. In travelling back to \( z = 0 \), fluorescence suffers from various attenuations which are lumped into another coefficient \( \beta \) (usually negligible).

Using Eqn 2.28, the overall collection efficiency for single fiber becomes

\[
\overline{\eta}_c = \frac{\int_0^\infty e^{-\beta z} \eta_c QY \alpha I_0 e^{-\alpha z} dz}{\int_0^\infty QY \alpha I_0 e^{-\alpha z} dz} = \frac{\text{NA}^2}{2n_0^2} \frac{\alpha}{\alpha + \beta} F(L)
\]

(2.29)

\[
F(L) = \int_0^\infty e^{-z} \left( \frac{L}{L + z} \right)^2 dz
\]

(2.30)

where \( x = (\alpha + \beta)z \) and \( L = (\alpha + \beta)b \) are normalized distances. Asymptotically, \( F(L) \~ L \) as \( L \to 0 \), and \( F(L) \~ 1 - 2/L \) as \( L \to \infty \). Similar result has been obtained for backscattering from bulk medium [H2, Culshaw].

2.5.4 Collection Efficiency of Double Fibers

Consider 2 identical and parallel MMFs whose cores are separated by \( 2s \) distance as shown in Fig 2.3. Excitation is carried by one fiber and fluorescence is collected by the other. The derivation for double fibers is same as for single fiber, except that the collecting fiber does not "sees" the entire cross-section \( A(z) \) but only the overlap area \( B(z) \) beginning at \( z_0 = s/\tan \theta_{NA} = bs/a \). This reduces \( \eta_c \) in proportion by

\[
\frac{B(z)}{A(z)} = \frac{\phi - \sin \phi}{\pi}, \quad \cos \frac{\phi}{2} = \frac{b + z_0}{b + z}
\]

(2.31)

Then, the overall collection efficiency for double fibers becomes

\[
\overline{\eta}_c = \frac{\int_0^\infty e^{-\beta z} \eta_c QY \alpha I_0 e^{-\alpha z} dz}{\int_0^\infty QY \alpha I_0 e^{-\alpha z} dz} = \frac{\text{NA}^2}{2n_0^2} \frac{\alpha}{\alpha + \beta} G(L, s)
\]

(2.32)

\[
G(L, s) = \int_{z_0}^\infty e^{-z} \left( \frac{L}{L + z} \right)^2 \frac{\phi - \sin \phi}{\pi} dz
\]

(2.33)

where \( z = (\alpha + \beta)z \), \( L = (\alpha + \beta)b \), \( z_0 = Ls/a \) are normalized distances, and \( \cos(\phi/2) = (L + z_0)/(L + z) \).
Figure 2.4: Graph of $F(L)$ and $G(L, s)$ with $s = 0$ and $s = 1.44a$. $F(L)$ is for single fiber. $G(L, 0)$ is for double fibers with no cladding so that the fiber cores touch each other. $G(L, 1.44a)$ is for double fibers with realistic dimensions of 62.5μm cladding radius and 25.6μm core radius, just like the MMF used in my experiment.

$F(L)$ and $G(L, s)$ are evaluated using Romberg numerical integration [Press] and plotted in Fig 2.4. Since $F(L) > G(L, s)$ for all $L > 0$, as expected, single fiber collects more fluorescence than double fibers. The assumption of zero separation ($s = 0$), even though unrealistic because cladding is present in most optical fibers, establishes the upper limit of collection efficiency for double fiber configuration. When a real cladding dimension of MMF ($s = 1.44a$) is considered, the efficiency decreases by an order of magnitude.

2.6 Fluorescence Collection by Evanescent Coupling

In order to maximize evanescent coupling in MMF [19], the cladding is removed so that the core is in direct contact with sample ($n_0$) which acts as the missing "cladding". Similarly in order to maximize evanescent coupling in SMF [111], the fiber is heated and stretched so that the core for all practical purpose disappears, and a new optical waveguide is formed by the cladding ($n_2$) acting as "core" and sample ($n_0$) acting as "cladding". Since typical sample index is not equal to the cladding index, there is coupling loss [114, 115, 116] due to $NA$ mismatch at the beginning and at the end of sensing region.

2.6.1 Evanescent Excitation

The sample absorbs evanescent power just as it does in the case of bulk excitation. But, because the sample experiences only $p$ fraction of the total power, the absorption coefficient decreases to $\gamma = \alpha p$ and the excitation profile becomes $I(z) = I_0 e^{-\gamma z}$. For a SMF or MMF with weakly guiding step-index profile, the
cladding fraction of mode power is [Cheo, Ghatak]

\[
p = \frac{P_{\text{clad}}}{P_{\text{clad}} + P_{\text{core}}} = \frac{u^2a^2}{V^2} \left(1 - \frac{K_l(wa)}{K_{l-1}(wa)K_{l+1}(wa)}\right)
\]  

(2.34)

where \( u^2 = k_0^2n_1^2 - \beta^2 \), \( w^2 = \beta^2 - k_0^2n_2^2 \), \( u^2a^2 + w^2a^2 = V^2 \), and Bessel function \( K_0() \) represents the radial field of mode order \( l \) in the cladding.

By summing over all \( N = V^2/2 \) modes in step-index MMF, the cladding fraction of total power propagating in MMF becomes [18, 19, after minor correction]

\[
p = \frac{\sum P_{\text{clad}}}{\sum P_{\text{clad}} + \sum P_{\text{core}}} \approx \frac{1}{N^2\sqrt{2N-2x}} = \frac{4}{3V}
\]  

(2.35)

In the case of SMF, there is only one mode to consider, namely \( l = 0 \). However, it is much easier to work with a Gaussian approximation than with the exact Bessel function. For step-index SMF, the radial field of fundamental mode can be approximated by [Snyder]

\[
\psi(r) = \sqrt{2} \frac{e^{-r^2/w^2}}{w\sqrt{\pi}}, \quad \frac{a}{w} = \sqrt{\ln V}, \quad V > 1
\]  

(2.36)

where \( w \) is the spot size of Gaussian profile and \( a \) is the core radius. Then, the cladding fraction of mode power in SMF becomes a simple ratio of integrals, [111]

\[
p = \frac{\int_0^\infty \psi^2(r)rdr}{\int_0^\infty \psi^2(r)rdr} = e^{-2a^2/w^2} = \frac{1}{V^2}
\]  

(2.37)

### 2.6.2 Collection Efficiency

The collecting fluorescence through evanescent coupling can be thought of as the inverse of evanescent absorption: the amount that “tunnels” back into a guiding mode is proportional to the evanescent field intensity of that mode. Hence, MMF is expected to have higher collection efficiency than SMF because of the simple fact that MMF supports many modes and SMF supports only one.

Using Eqn 2.36, the collection efficiency for step-index SMF is given as [111]

\[
\eta_c = \frac{\pi}{k_0^2n_1^2} \int_0^\infty \frac{\psi^4(r)rdr}{\psi^2(r)rdr} = \frac{e^{-2a^2/w^2}}{k_0^2n_1^2w^2} = \frac{\ln V}{V^4/2\Delta}
\]  

(2.38)

This expression was derived with 2 assumptions — first, the indexes of fiber and sample are similar (\( n_1 \approx n_2 \approx n_0 \)); second, the wavelengths of excitation and fluorescence are close enough (\( \lambda_{ex} \approx \lambda_f \)) so that their modal fields are similar.

Unfortunately, there is no comparable analytical expression for MMF. The collection efficiency of weakly guiding step-index MMF [110] and general step-index MMF [112, 113] has been studied using numerical simulations. For bulk source distributed uniformly throughout the sample, \( \eta_c \propto V \) since only high-order modes have appreciable penetration into the cladding. For surface source located at the fiber-sample interface, \( \eta_c \propto V^2 \) since every mode has a reasonable power at the interface. But, the exponentially decaying source profile which occurs in evanescent excitation has not been studied so far.
Chapter 3

Fluorescence System

3.1 Fluorescent Response

A single lifetime fluorescence system can be modelled as a 3 level system as depicted in Fig 3.1. When excited, the system moves from ground state $|1\rangle$ to absorption state $|3\rangle$, then quickly settles to emission state $|2\rangle$ via non-radiative relaxations. The system drops down to ground state through radiative ($A$) and non-radiative ($k$) decays. When the excitation is time varying $E(t)$, the emission state $|2\rangle$ will be populated at a rate proportional to the excitation. The rate equation describing the emission state population $N_2(t)$ is [Lakowicz, Birks]

$$\frac{dN_2(t)}{dt} = -(A + k)N_2(t) + E(t)$$

(3.1)

which has well-known solution $N_2(t) = e^{-t/\tau} \ast E(t)$, where $\tau = 1/(A + k)$ is the lifetime of fluorescent decay.

Since the fluorescent intensity that is actually observed is $(hc/\lambda_P)AN_2(t)$, the solution can be rewritten in terms of input $x(t) \sim E(t)$ and output $y(t) \sim N_2(t)$ of linear, time-invariant, and stable system

$$x(t) \rightarrow \left[ h(t) \right] \rightarrow y(t)$$

$$y(t) = h(t) \ast x(t) \leftrightarrow Y(f) = H(f)X(f)$$

(3.2)

which is entirely characterized by the impulse response $h(t) = e^{-t/\tau}$ and the frequency response $H(f) = \tau/(1 + j2\pi f \tau)$. Since optical signals cannot be

Figure 3.1: 3-level model of fluorescence system
The input and output must be biased above 0. This is important for sinusoidal input and output

\[ x(t) = 1 + a \cos(2\pi ft), \quad |a| \leq 1 \]  \hspace{1cm} (3.3)

\[ y(t) = 1 + a \frac{|H(f)|}{|H(0)|} \cos(2\pi ft + \arg H(f)) \]  \hspace{1cm} (3.4)

\[ = 1 + a \frac{\cos(2\pi ft - \tan^{-1}(2\pi f\tau))}{\sqrt{1 + (2\pi f\tau)^2}} \]  \hspace{1cm} (3.5)

where the constant term has been normalized to 1.

### 3.2 Determining Lifetime in Time Domain

The fluorescent lifetime of sample can be measured in time domain using pulse excitation. Because of finite pulse width, finite detector bandwidth, \( \lambda \)-bias, and other problems, it is a standard practice to measure fluorescence from both the sample and a reference using the same setup. This arrangement is outlined in block diagrams,

\[ \begin{align*}
  l(t) &\rightarrow h_r(t) \rightarrow d(t) \rightarrow x(t) \\
  l(t) &\rightarrow h(t) \rightarrow d(t) \rightarrow y(t)
\end{align*} \]

where \( l(t) \) is laser excitation, \( d(t) \) is detector response, \( h(t) = e^{-t/\tau} \) is sample response with lifetime \( \tau \), \( h_r(t) = e^{-t/\tau_r} \) is reference response with lifetime \( \tau_r \), \( x(t) \) is measured reference fluorescence, and \( y(t) \) is measured sample fluorescence. Mathematically, it can be described as

\[ \begin{align*}
  x(t) &= h_r(t) \ast d(t) \ast l(t) \quad \Leftrightarrow \quad X(f) = H_r(f)D(f)L(f) \quad \text{(3.6)} \\
  y(t) &= h(t) \ast d(t) \ast l(t) \quad \Leftrightarrow \quad Y(f) = H(f)D(f)L(f) \quad \text{(3.7)}
\end{align*} \]

This problem is solved by iterative convolution in which the measured data \( \{x_i, y_i\} \) is fitted directly to the convolution models. After convolving \( h_r(t) \) with Eqn 3.7 and \( h(t) \) with Eqn 3.6, the residual function

\[ \chi^2(\tau) = \sum_i (h_r(t_i; \tau_r) \ast y_i - h(t_i; \tau) \ast x_i)^2 \]  \hspace{1cm} (3.8)

is minimized for the least-square estimator of \( \tau \). Since the reference lifetime \( \tau_r \) is a fixed constant, this is a 1-D minimization problem which can be solved by Brent’s method [Press], for example. Although one convolution needs to be calculated for every new \( \tau \) point, the convergence for 1-D problem is usually fast. Because of its simplicity and flexibility, this method is preferred over direct deconvolution methods.

### 3.3 Determining Lifetime in Frequency Domain

The fluorescent lifetime of sample can be measured in frequency domain using sinusoidal or harmonic excitation. Typically, sinusoidal excitation is generated by modulating a continuous wave source; and harmonic excitation is derived from a train of pulses which decomposes into harmonic series. 3 signals are involved
in general — excitation \( z(t) \) with fundamental frequency \( f_1 \) and harmonics \( n f_1 \), fluorescent response \( y(t) \), and LO signal \( z(t) \) at frequency \( f_o \).

\[
x(t) = 1 + \sum_n a_n \cos(2\pi n f_1 t) \\
y(t) = 1 + \sum_n a_n m_n \cos(2\pi n f_1 t - \varphi_n) \\
z(t) = \cos(2\pi f_o t)
\]

(3.9)  
(3.10)  
(3.11)

where, from Eqns 3.3 and 3.5,

\[
m_n = \frac{1}{\sqrt{1 + (2\pi n f_1 \tau)^2}}, \quad \varphi_n = \tan^{-1}(2\pi n f_1 \tau)
\]

(3.12)

If the frequency \( f_1 \) is low enough, the amplitude factor \( (m_1) \) and phase shift \( (\varphi_1) \) are measured directly from Eqns 3.9 and 3.10. But if the frequency is high, heterodyne or homodyne method is used, where LO signal \( z(t) \) is mixed with RF signals \( x(t) \) and \( y(t) \), either inside or outside photodetector. Subsequent lowpass filter produces "down-converted" IF signals \( X(t) \) and \( Y(t) \) at a sufficiently low frequency so that \( m_1 \) and \( \varphi_1 \) can be measured using a lockin amplifier. In heterodyne method, \( f_o \approx f_1 \) and the IF signals are at frequency \( \Delta f = f_1 - f_o \); and in homodyne method, \( f_o = f_1 \) and the IF signals are DC:

\[
\begin{array}{c|c|c}
\text{heterodyne} & \text{homodyne} \\
\frac{x(t)z(t) \rightarrow X(t)}{y(t)z(t) \rightarrow Y(t)} & \frac{\cos(2\pi \Delta f t)}{m_1 \cos(2\pi \Delta f t + \varphi_1)} & \frac{1}{m_1^2}
\end{array}
\]

(3.13)

Usually, the data are sampled at more than one frequency, either by changing \( f_o \) to near any of the harmonics or by changing \( f_1 \) itself.

Once \( \{m, \varphi\} \) data set has been obtained, the lifetime \( \tau \) is determined either through linear regression of [P4]

\[
\tan \varphi = 2\pi f \tau, \quad 1/m^2 = 1 + (2\pi f \tau)^2
\]

(3.14)

against the frequency \( f \), or through minimization of residual function

\[
\chi^2(\tau) = \sum_i (m_i - m(f_i, \tau))^2 + \sum_i (\varphi_i - \varphi(f_i, \tau))^2
\]

where \( m_i \) and \( \varphi_i \) are measured at frequency \( f_i \), and \( m(f_i) \) and \( \varphi(f_i) \) are calculated from Eqn 3.12. For an example, \( m \) and \( \varphi \) that will be produced at \( f = 1\text{MHz} \) are

<table>
<thead>
<tr>
<th>( \tau )</th>
<th>( m )</th>
<th>( \varphi )</th>
</tr>
</thead>
<tbody>
<tr>
<td>30ns</td>
<td>0.983</td>
<td>10.7°</td>
</tr>
<tr>
<td>350ns</td>
<td>0.414</td>
<td>65.5°</td>
</tr>
</tbody>
</table>

Fig 3.2 lists papers, in the order of increasing bandwidth, which use sinusoidal or harmonic excitation to measure fluorescent lifetime in the frequency domain. I should point out that since fluorescence system is treated as linear and time-invariant, a Network Analyzer (some with 20GHz bandwidth) can perform all the signal processing and can in principle replace most of the equipments described in the papers. But, Network Analyzer is very expensive, and the papers basically talk about cheaper alternative of measuring the sinusoidal response. Main advantages of frequency domain method over time domain method are that high detection bandwidth and deconvolution of sample response are not required.
Figure 3.2: Survey of frequency domain fluorometers

<table>
<thead>
<tr>
<th>Refs</th>
<th>max freq</th>
<th>source</th>
<th>data</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sinusoidal Excitation</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[P17]</td>
<td>1kHz</td>
<td>EOM</td>
<td>φ</td>
</tr>
<tr>
<td>[P21]</td>
<td>455kHz</td>
<td>LED</td>
<td>φ</td>
</tr>
<tr>
<td>[P22]</td>
<td>10MHz</td>
<td>LED</td>
<td>φ</td>
</tr>
<tr>
<td>[P18]</td>
<td>40MHz</td>
<td>AOM</td>
<td>φ</td>
</tr>
<tr>
<td>[P1]</td>
<td>50MHz</td>
<td>EOM</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P19]</td>
<td>120MHz</td>
<td>D₂ Lamp</td>
<td>m</td>
</tr>
<tr>
<td>[P2]</td>
<td>160MHz</td>
<td>EOM</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P3, A7, A8]</td>
<td>200MHz</td>
<td>EOM</td>
<td>m, φ</td>
</tr>
<tr>
<td>[N24]</td>
<td>320MHz</td>
<td>Quartz AOM</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P20]</td>
<td>500MHz</td>
<td>LiNbO₃ IOM</td>
<td>φ</td>
</tr>
<tr>
<td>Harmonic Excitation</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[P7]</td>
<td>100MHz</td>
<td>synchrotron radiation</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P9]</td>
<td>400MHz</td>
<td>mode-locked laser + AOM</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P10]</td>
<td>820MHz</td>
<td>mode noise of CW laser</td>
<td>m</td>
</tr>
<tr>
<td>[P4, P5]</td>
<td>1GHz</td>
<td>mode-locked laser</td>
<td>m</td>
</tr>
<tr>
<td>[P11]</td>
<td>2GHz</td>
<td>pulsed laser diode</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P14]</td>
<td>2GHz</td>
<td>mode-locked laser</td>
<td>m, φ</td>
</tr>
<tr>
<td>[P15]</td>
<td>10GHz</td>
<td>mode-locked laser</td>
<td>m, φ</td>
</tr>
</tbody>
</table>
Chapter 4

Acousto-Optic Modulator

I used PbMoO₄ acousto-optic modulator (AOM) in my experiment because it was cheap and available. Others [N9, N10, N12, N15, N16, N17, N18] have used different optical modulators to implement various schemes in which the double modulation of optical signal is the key feature just as in my own correlation scheme. The choice depends on factors like modulation bandwidth, operating wavelength, compatibility with optical fiber, and, most of all, cost and availability. In this section, I review only those aspects of AOM relevant to amplitude modulation, since that is how I use it.

4.1 Acoustic Perturbation

Acousto-optic (AO) interaction can be modelled as inelastic scattering between incident optical photon \((\omega, k)\) and acoustic phonon \((\Omega, K)\), producing a diffracted photon that is “up-shifted” \((\omega_+, k_+\) if the phonon is absorbed or “down-shifted” \((\omega_-, k_-\) if an identical phonon is emitted. [Korpel, Magdich, Yariv]

\[
\hbar \omega \pm \hbar \Omega = \hbar \omega_{\pm}, \quad \hbar k \pm \hbar K = \hbar k_{\pm}
\] (4.1)

The phase-matching conditions determine diffraction angle, polarization, frequency shift, and propagation constant. But the modulation of optical signal is achieved through the diffraction efficiency.

Outside the scattering site, incident wave \(E\) and diffracted wave \(E'\) propagate as separate and uncoupled plane waves. Inside the scattering site, however, \(E\) and \(E'\) waves are coupled through acoustic perturbation which is produced according to the well-known strain-optic relation, [Ghatak, Magdich]

\[
\Delta \begin{bmatrix} 1 \\ n^2 \end{bmatrix}_{kl} \rightarrow \begin{bmatrix} 1 & 6 & 5 \\ 6 & 2 & 4 \\ 5 & 4 & 3 \end{bmatrix} \leftrightarrow \begin{bmatrix} 1 \\ 2 \\ 3 \\ 4 \\ 5 \\ 6 \end{bmatrix} \leftarrow \Delta \left( \frac{1}{n^2} \right)_i = p_{ij} S_j
\] (4.2)

where \(p\) is strain-optic constant and \(S\) is strain field set up by the acoustic wave. Because of symmetry in the tensor components, matrix notation \([\cdot]_{kl}\) and vector notation \((\cdot)_i\) can be converted from one to the other, depending on the context. Then, the coupled wave equation becomes [Magdich, Ghatak, Yariv]

\[
\nabla \times \nabla \times (E + E') + \mu_0 \frac{\partial^2}{\partial t^2} ([\epsilon] + \Delta[\epsilon])(E + E') = 0
\] (4.3)

where

\[
[\epsilon] = \epsilon_0 [n^2], \quad \Delta[\epsilon] = -\frac{1}{\epsilon_0} [\epsilon] \Delta \left[ \frac{1}{n^2} \right] [\epsilon]
\] (4.4)
The degree of coupling between the incident and diffracted waves is given by scalar [Magdich, Ghatak, Yariv]

\[ \Delta \varepsilon = (e'|\Delta \varepsilon|e) \]  

where \( e \) and \( e' \) are unit vectors of \( E \) and \( E' \) respectively. Usually, the acoustic and optical waves are aligned with AOM’s axis, so that \( (e'|\Delta \varepsilon|e) \) selects only one element from \( \Delta \varepsilon \). Using appropriate scalar parameters in \( e = \epsilon_0 n^2 \) and \( \Delta(1/n^2) = pS \), the change in refractive index due to acoustic perturbation is [Magdich, Ghatak, Yariv]

\[ |\Delta n| = \frac{|\Delta \varepsilon|}{2\epsilon_0 n} = \frac{1}{2} n^3 |pS| = \sqrt{M_2} \sqrt{\frac{P_a}{2A}} \]  

where \( M_2 = n^6 p^2 / \rho u_0^3 \) is a figure of merit for AOM, \( A \) is area of transducer, and \( P_a = (A\mu_a)(\rho u_0^2 S^2) \) is acoustic power. From experimental point of view, \( \Delta n \) is the most important parameter because it influences all optical manifestations of AO interaction.

For example, consider longitudinal acoustic wave along \( \hat{z} \) in PbMoO₄ which is an uniaxial crystal with \( 4/m \) point symmetry:

\[
\Delta \left( \frac{1}{n^2} \right) = \begin{bmatrix} p_{11} & p_{12} & p_{13} & 0 & 0 & p_{16} \\ p_{12} & p_{11} & p_{13} & 0 & 0 & -p_{16} \\ p_{31} & p_{31} & p_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & p_{44} & p_{45} & 0 \\ 0 & 0 & -p_{45} & p_{44} & 0 & 0 \\ p_{61} & -p_{61} & 0 & 0 & 0 & p_{66} \end{bmatrix} \Delta \left( \frac{1}{n^2} \right)_0 = \begin{bmatrix} p_{13} S_3 \\ p_{13} S_3 \\ p_{33} S_3 \\ 0 \\ 0 \\ 0 \end{bmatrix}
\]  

Then, the acoustic perturbation as given by Eqn 4.4 is

\[ \Delta [\epsilon] = -\frac{1}{\epsilon_0} [\epsilon] \Delta \left[ \frac{1}{n^2} \right] [\epsilon] = -\epsilon_0 S_3 \begin{bmatrix} n_1^2 & 0 & 0 \\ 0 & n_1^2 & 0 \\ 0 & 0 & n_3^2 \end{bmatrix} \]  

Because \( \Delta [\epsilon] \) is diagonal, the diffracted and incident waves have the same polarization; otherwise, there would be no AO coupling. The scalar \( \Delta \varepsilon \) and \( \Delta n \) that are selected depend on the orientation of \( E \) and \( K \). Therefore, from Eqns 4.5 and 4.6,

\[ |\Delta n| = |\Delta \varepsilon| = \frac{S_3}{2} \left\{ \begin{array}{l} n_1^2 p_{13}, \text{ if } E \perp K \\ n_3^2 p_{33}, \text{ if } E \parallel K \end{array} \right\} \]  

\[ |\Delta n| = \frac{|\Delta \varepsilon|}{2\epsilon_0 n^2} = \frac{S_3}{2} \left\{ \begin{array}{l} n_1^2 p_{13}, \text{ if } E \perp K \\ n_3^2 p_{33}, \text{ if } E \parallel K \end{array} \right\} \]  

\[ Q = \frac{K^2 L}{k} = \frac{2\pi \lambda L}{nA^2} \rightarrow \left\{ \begin{array}{l} Q \ll 1, \text{ for Raman-Nath} \\ Q \gg 1, \text{ for Bragg} \end{array} \right\} \]  

where \( K \) is acoustic wavevector, \( k \) is optical wavevector, and \( L \) is transducer length.
4.2.1 Raman-Nath AOM

In Raman-Nath AOM, multiple scattering takes place because the acoustic divergence is greater than the diffraction angle. If $\theta_0$ is the angle of incidence with respect to the acoustic normal, the diffraction efficiency ($\eta$) and angle ($\theta$) of $m$th-order beam are [Ghatak, Korpel, N25, Yariv]

$$\eta_m = J_m^2 \left( \Delta\varphi \frac{L \tan \theta_0}{\Lambda} \right), \quad \theta_m = \theta_0 + \sin^{-1} \left( \frac{mK}{k} \right)$$

(4.12)

where $J_m()$ is standard Bessel function, and $\Delta\varphi = k_0 \Delta n L / \cos \theta_0$ is optical phase shift caused by $\Delta n$ over the interaction length.

When $\theta_0 = 0$ for example, 100% of 0th-order beam is diffracted out at $\Delta\varphi = 2.40$; maximum 34% diffraction into 1st-order occurs at $\Delta\varphi = 1.84$; and maximum 24% diffraction into 2nd-order occurs at $\Delta\varphi = 3.05$. Naturally, higher orders exhibit progressively lower efficiencies. Due to its low efficiency and low bandwidth, Raman-Nath AOM appears only in specialized application [N24].
4.2.2 Bragg AOM

In Bragg AOM, only single scattering occurs because the acoustic divergence is smaller than the diffraction angle. As depicted in Fig 4.1, the Bragg diffraction produces a single 1st-order beam when the angle of incidence ($\theta_0$) is equal to the Bragg angle

$$\theta_B = \sin^{-1} \left( \frac{K}{2k} \right) = \sin^{-1} \left( \frac{\lambda}{2n\lambda} \right) \tag{4.13}$$

The diffraction efficiency of 0th and 1st order beams are [Ghatak, Korpel, N25, Yariv]

$$\eta_1 = \sin^2 \left( \frac{\Delta \varphi}{2} \right), \quad \eta_0 = 1 - \eta_1 \tag{4.14}$$

where $\Delta \varphi = k_0 \Delta nL / \cos \theta_B$ is optical phase shift caused by $\Delta n$ over the interaction length. Bragg AOM can reach 100% 1st-order diffraction at $\Delta \varphi = 3.14$.

As shown in Fig 4.2, if the angle of incidence increases by $\Delta \theta$ from the Bragg angle, then the 1st-order angle decreases by the same amount, so that the propagation constants become $\beta_0 = k \cos(\theta_B + \Delta \theta)$ and $\beta_1 = k \cos(\theta_B - \Delta \theta)$. The resulting phase mismatch reduces the diffraction efficiency of 1st-order beam to [Ghatak, Korpel, N25, Yariv]

$$\eta_1 = \frac{\Delta^2 \varphi}{\Delta^2 \varphi + \Delta^2 \alpha} \sin^2 \left( \frac{\sqrt{\Delta^2 \varphi + \Delta^2 \alpha}}{2} \right) \tag{4.15}$$

where $\Delta \alpha = K \Delta nL / \cos \theta_B$ is acoustic phase shift caused by the angular change $\Delta \theta$, and $\Delta \varphi = k_0 \Delta nL / \cos \theta_B$ is optical phase shift caused by $\Delta n$. The phase mismatch usually produces higher orders, as well.

4.3 Modulation Bandwidth

Intensity modulation in AOM is accomplished by modulating the amplitude of acoustic carrier wave, which in turn modulates the diffraction efficiency. If $f_m$ and $f_c$ are modulation and carrier frequencies respectively, amplitude modulation (AM) creates 2 sidebands at $f_c \pm f_m$. Experimentally, maximum modulation frequency $f_{max}$ is reached when modulation wavelength $\Lambda_m$ is equal to optical beam width $D$, at which point there is zero net modulation. [Korpel]

$$f_{max} = \frac{v}{\Lambda_{max}} = \frac{v}{D} = \frac{1}{\tau} \tag{4.16}$$

where $\tau = D/v$ is transit time for the acoustic wave to cross the optical beam, and $\nu = \Lambda_c f_c = \Lambda_m f_m$ is the acoustic speed. As the beam width is reduced, the modulation bandwidth increases, the optical divergence increases, and the diffraction efficiency decreases because greater proportion of incident beam is phase mismatched.

When acoustic wave with sinusoidal envelope $s(z) = 1 + a \cos(2\pi z/\Lambda_m)$ passes through optical beam with spatial profile $p(z)$, the diffracted signal $\eta(t)|_{z=vt} = p(z) * s(z)$ is also sinusoidal. This situation resembles the sinusoidal excitation of a linear system as defined in Eqns 3.3 and 3.4, where $s(z)$ acts as “input”, $p(z)$ acts as “system response”, and $\eta(t)$ acts as “output”. If the optical beam has uniform profile with $D$ width, $p(z) = \Pi(z/D)$; and if it has Gaussian profile with $D$ full width at $e^{-2}$ point, $p(z) = e^{-8z^2/D^2}$. Then,
the diffraction output becomes [Magdich, Goutzoulis]

\[
\eta(t) = 1 + a \left| \frac{P(1/\Lambda_m)}{P(0)} \right| \cos(2\pi f_m t) = 1 + a \text{sinc}(f_m / f_{max}) \cos(2\pi f_m t), \quad \text{uniform beam} \tag{4.18}
\]

\[
= 1 + ae^{-r^2 / 2f_{m0}^2} \cos(2\pi f_m t), \quad \text{Gaussian beam} \tag{4.19}
\]

where \( P(f) \) is the spatial FT of optical beam profile \( p(z) \). The 3dB point or 50% modulation depth occurs at

\[
f_{3dB} = \begin{cases} 
0.60f_{max}, & \text{uniform beam} \\
0.75f_{max}, & \text{Gaussian beam} 
\end{cases} \tag{4.20}
\]
Chapter 5

Signal Processing

5.1 Signal Flow

Fig 5.1 depicts the block diagram of my all optical signal processing scheme:

1. LO signal $x(t)$ is modulated onto DC laser intensity by AOM.

2. 1-way propagation in optical fiber introduces $\Delta t/2$ delay.

3. At the far-end, Fresnel reflection $x(t - \Delta t/2)$ or fluorescent response $y(t - \Delta t/2)$ couples back into the fiber.

4. On the return path, the fiber introduces another $\Delta t/2$ propagation delay.

5. Second optical modulation by AOM mixes the LO signal $x(t)$ with the returning signal, giving $x(t)x(t - \Delta t)$ or $x(t)y(t - \Delta t)$.

6. The mixed signal is integrated by lowpass detector, producing auto correlation $R_{xx}(\Delta t)$ or cross correlation $R_{xy}(\Delta t)$.

The goal is to determine the fiber length ($L$) by analyzing the auto correlation which comes from Fresnel backreflection with impulse response $h(t) = \delta(t)$, and to determine the fluorescent lifetime ($\tau$) by analyzing the cross correlation which comes from SPA or RU fluorescence with impulse response $h(t) = e^{-t/\tau}$.

All signals are periodic, either sinusoidal or square-wave, with frequency $f = 1/T$. Since the propagation delay $\Delta t = 2Ln/c$ is fixed by the fiber length, and since it is the modulation frequency that is being varied in my experiment, the correlations turn out to be functions of $f$:

\[
\frac{1}{T} \int_{0}^{T} x(t)x(t-\Delta t)dt = R_{xx}(\Delta t) \rightarrow R_{xx}(f) \quad (5.1)
\]

\[
\frac{1}{T} \int_{0}^{T} x(t)y(t-\Delta t)dt = R_{xy}(\Delta t) \rightarrow R_{xy}(f) \quad (5.2)
\]

This represents a fundamental change of domain which does not easily lend itself to the conventional techniques of (stationary) random variable. Although time domain responses can be inferred from inverse FT of correlations, it is simpler to estimate $L$ and $\tau$ directly in frequency domain.
Figure 5.1: Block diagram of signal flow in my experiment. Auto correlation (top) is produced when measuring Fresnel backreflection, and cross correlation (bottom) is produced when measuring SPA or RU fluorescence.

\[
R_{xx}(-\Delta t) = \frac{1}{T} \int_0^T dt
\]

\[
R_{xy}(-\Delta t) = \frac{1}{T} \int_0^T dt
\]

Figure 5.2: Square-wave excitation \( x(t) \) and fluorescent response \( y(t) \) of single lifetime system
5.2 Auto and Cross Correlations

Square-wave excitation is used extensively in my experiment because the simple "on/off" states are easier to generate, especially with nonlinear modulator like AOM. But its analysis is based on sinusoidal case. The sinusoidal input $x(t)$ and the fluorescent response $y(t)$ are given by Eqns 3.3 and 3.5. Now, consider an ideal (equal on/off) square-wave excitation at frequency $f = 1/T$, as illustrated in Fig 5.2. The waveform is decomposed into more convenient Fourier series using Eqns A.48 and A.49.

$$x(t) = 2 \sum_{m=-\infty}^{\infty} \delta(t - mT)$$

$$= 1 + \sum_{m=1}^{\infty} 2 \text{sinc} \left( \frac{m}{2} \right) \cos(2\pi mt/T)$$

$$= 1 + \sum_{m=1,3,5,...}^{\infty} \frac{4(-1)^{(m-1)/2}}{\pi m} \cos(2\pi mfT)$$

By superposing Eqn 3.5, the fluorescent response to the square-wave excitation is

$$y(t) = 1 + \sum_{m=1}^{\infty} \frac{4(-1)^{(m-1)/2}}{\pi m} \cos \left( 2\pi mfT - \tan^{-1} (2\pi mf) \right) \sqrt{1 + (2\pi mf)^2}$$

which, as shown in Fig 5.2, rises and falls exponentially between the maximum $B = 1 + \tanh(T/4\tau)$ and the minimum $A = 1 - \tanh(T/4\tau)$.

Then, the correlations defined by Eqns 5.1 and 5.2 become, for sinusoidal case

$$R_{xx}(f) = 1 + \frac{a^2}{2} \cos(2\pi f \Delta t)$$

$$R_{xy}(f) = 1 + \frac{a^2}{2} \cos \left( 2\pi f \Delta t + \tan^{-1} (2\pi f \tau) \right) \sqrt{1 + (2\pi f \tau)^2}$$

and, for square-wave case

$$R_{xx}(f) = 1 + \sum_{\text{odd } m} \frac{8}{\pi^2 m^2} \cos(2\pi mf \Delta t)$$

$$R_{xy}(f) = 1 + \sum_{\text{odd } m} \frac{8}{\pi^2 m^2} \cos \left( 2\pi mf \Delta t + \tan^{-1} (2\pi mf) \right) \sqrt{1 + (2\pi mf)^2}$$

The effect of finite bandwidth on the shape of correlation cannot be ignored. First, the AOM frequency response is $\text{sinc}(0.6f/f_{3dB})$ from Eqns 4.18 and 4.20, where beam profile for both the laser beam and the MMF output are assumed sufficiently uniform. After 2 passes through AOM, the frequency signal is attenuated by $\text{sinc}^2(0.6f/f_{3dB})$. Second, the cosine series is truncated after 36MHz because very little contribution comes from higher frequencies due to bandwidth and lifetime attenuations. Therefore, the auto and cross correlations must be corrected to, for sinusoidal case

$$R_{xx}(f) = 1 + \frac{a^2}{2} \text{sinc}^2(u) \cos(v)$$

$$R_{xy}(f) = 1 + \frac{a^2}{2} \text{sinc}^2(u) \cos(v + \tan^{-1}(u)) \sqrt{1 + u^2}$$
and, for square-wave case
\[
R_{zz}(f) = 1 + \sum_{m=1}^{mf=36MHz} \frac{8}{\pi^2 m^2} \text{sinc}^2(mw) \cos(mu) (5.13)
\]
\[
R_{yz}(f) = 1 + \sum_{m=1}^{mf=36MHz} \frac{8}{\pi^2 m^2} \text{sinc}^2(mw) \frac{\cos (mu + \tan^{-1}(mu))}{\sqrt{1 + m^2 w^2}} (5.14)
\]
where \( w = 0.6 f / f_{dB}, v = 2\pi f \Delta t, u = 2\pi f \tau, \) and \( \Delta t = 2L / c. \)

Using parameter values which are close to the actual experimental conditions — \( f_{dB} = 8MHz, L = 50m, \tau = 30ns \) (SPA), and \( \tau = 350ns \) (RU) — both square-wave and sinusoidal excitation have been simulated. The AC component of \( R_{zz}(f) \) for Fresnel backreflection is plotted in Fig 5.3; and the AC component of \( R_{yz}(f) \) for SPA and RU fluorescence are plotted in Figs 5.4 and 5.5, respectively.

### 5.3 Time Domain Responses

Ignoring the bandwidth attenuation and using Eqns A.43 and A.44, inverse FT of correlations are, for sinusoidal case
\[
\mathcal{F}^{-1}\{R_{zz}(f)\} = \delta(t) + \frac{a^2}{4} \delta(t - \Delta t) + \frac{a^2}{4} \delta(t + \Delta t) \tag{5.15}
\]
\[
\mathcal{F}^{-1}\{R_{yz}(f)\} = \delta(t) + \frac{a^2}{4\tau} \begin{cases} 0, & |t| < \Delta t \\ e^{-(|t| - \Delta t)/\tau}, & |t| > \Delta t \end{cases} \tag{5.16}
\]
and, for square-wave case
\[
\mathcal{F}^{-1}\{R_{zz}(f)\} = \delta(t) + \sum_{m=1}^{\infty} \frac{4}{\pi^2 m^2} \left( \delta(t - m\Delta t) + \delta(t + m\Delta t) \right) \tag{5.17}
\]
\[
\mathcal{F}^{-1}\{R_{yz}(f)\} = \delta(t) + \sum_{m=1}^{\infty} \frac{4}{\pi^2 m^2} \begin{cases} 0, & |t| < m\Delta t \\ e^{-(|t| - m\Delta t)/\tau}, & |t| > m\Delta t \end{cases} \tag{5.18}
\]

Therefore, at least for \( t > 0 \) (sinusoidal) or for \( 0 < t < 3\Delta t \) (square-wave), the inverse FT correctly represents the impulse response of Fresnel reflection \( \delta(t - \Delta t) \) and fluorescent sample \( e^{-(t - \Delta t)/\tau} \), with propagation delay \( \Delta t \) and lifetime \( \tau \).

The inverse FT was done analytically because \( R_{zz}(f) \) and \( R_{yz}(f) \) are known for all \(-\infty < f < \infty\). But in real world, the inverse FT must be done by DFT because the correlations are known only at discrete points over \( -f_{\max} < f < f_{\max} \). That is, \( N \) data points are sampled at \( \delta f \) interval in the positive frequency range, and then flipped onto the negative frequency range by taking advantage of the fact that the correlations are even functions. This gives us \( 2N \) data points for DFT whose fundamental relation between time domain resolution \( \delta t \) and frequency domain resolution \( \delta f \) is
\[
(2N) \delta f \delta t = 2f_{\max} \delta t = 1 \tag{5.19}
\]
Since \( f_{\max} = 4MHz \) in my experiment, the time and spatial resolutions are limited to
\[
\delta t = 125ns, \quad \delta L = (c/2n)\delta t = 13m \tag{5.20}
\]
Clearly, these resolutions are inadequate for fiber length of 50m and fluorescent lifetimes of 30ns and 350ns, if I were to follow the traditional DFT method.
Figure 5.3: Simulated AC component of $R_{zz}(f)$ for Fresnel backreflection under square-wave excitation (top) and sinusoidal excitation (bottom), with $L = 50m$ and $f_{3dB} = 8MHz$
Figure 5.4: Simulated AC component of $R_{xy}(f)$ for SPA fluorescence under square-wave excitation (top) and sinusoidal excitation (bottom), with $L = 50\,\text{m}$, $f_{3\text{dB}} = 8\,\text{MHz}$, and $\tau = 30\,\text{ns}$
Figure 5.5: Simulated AC component of $R_{xy}(f)$ for RU fluorescence under square-wave excitation (top) and sinusoidal excitation (bottom), with $L = 50\mu m$, $f_{3dB} = 8\text{MHz}$, and $\tau = 350\text{ns}$
5.4 Estimation of Parameters

The auto correlation of Eqns 5.11 and 5.13 and the cross correlation of Eqns 5.12 and 5.14 can be modelled as \( y = R_{xx}(f; L) \) and \( y = R_{xy}(f; L, \tau) \), where fiber length \( L \) and fluorescent lifetime \( \tau \) are unknown parameters. Fortunately, the 2 parameters can be determined sequentially. First, the least-square estimator of \( L \) is found by minimizing residual function

\[
\chi^2(L) = \sum_i (y_i - R_{xx}(f_i; L))^2
\]

where \( y_i \) is the actual measurement of auto correlation due to Fresnel backreflection at frequency \( f_i \), and \( R_{xx}(f_i) \) is calculated from the analytical model.

Second, the least-square estimator of \( \tau \) is found by minimizing residual function

\[
\chi^2(\tau) = \sum_i (y_i - R_{xy}(f_i; L, \tau))^2
\]

where \( y_i \) is the measurement of cross correlation due to SPA or RU fluorescence at frequency \( f_i \), and \( R_{xy}(f_i) \) is calculated from the model.

In each case, the data fitting reduces to 1-D minimization problem which is solved by Brent's method [Press]. When evaluating the residual in Eqns 5.21 and 5.22, the measured and the calculated data are standardized, first by subtracting linear regression line from the data, and then by normalizing the data to zero mean and unit standard deviation.

5.5 Estimation of Error

The accuracy and the confidence interval of estimation can only be established through ensemble average. Conceptually, the “true” parameter \( a_{true} \) is statistically realized, along with other random processes or errors, as an experimental data set \( D_1 \) from which an estimation \( a_1 \) is obtained by data fitting to a model. However, because of the random components, \( D_1 \) is not an unique realization of \( a_{true} \). There are infinitely many other realizations \( D_2, D_3, \cdots \) from which estimations \( a_2, a_3, \cdots \) are obtained. After sufficient number of data sets, the ensemble of \( a_i \) would describe some probability distribution about \( a_{true} \) in the parameter space. [Press]

If \( \{a_i\} \) is a sample of parameter estimations, then the true parameter is approximated by ensemble average: [Guttman]

\[
a = \bar{a} \pm (t_{N-1; \alpha/2}) \frac{s_a}{\sqrt{N}}
\]

where \( t_{N-1; \alpha/2} \) is a point in Student-\( t \) distribution with \( N - 1 \) degree of freedom and 100(1 - \( \alpha \))% confidence interval, and \( \bar{a} \) and \( s_a \) are the usual mean and standard deviation,

\[
\bar{a} = \frac{1}{N} \sum_{i=1}^{N} a_i, \quad s_a^2 = \frac{1}{N-1} \sum_{i=1}^{N} (a_i - \bar{a})^2
\]
Also, if \( \{b_i\} \) is a sample of estimations for another parameter \( b \), the difference \( a - b \) is approximated by [Guttman]

\[
a - b = \bar{a} - \bar{b} \pm (t_{m; \alpha/2}) \sqrt{\frac{s_a^2}{N_a} + \frac{s_b^2}{N_b}}
\]  

(5.25)

where

\[
\frac{1}{m} = \frac{c^2}{N_a - 1} + \frac{(1 - c)^2}{N_b - 1}, \quad c = \frac{s_a^2/N_a}{s_a^2/N_a + s_b^2/N_b}
\]  

(5.26)
Chapter 6

Experiment

6.1 Experimental Setup

The experimental setup of my correlation scheme is outlined in Fig 6.1; and the optical and electrical equipments are shown in Fig 6.2. The purpose of this experiment is to demonstrate a novel optical signal processing scheme for measuring the lifetime of fluorescence decay. Because the optical fiber introduces round-trip propagation delay, the fiber length is also measured using Fresnel backreflection from the far-end.

A continuous Ar laser beam passes through halfwave rotator (λ/2) and polarizing beamsplitter (BS), and is focused by a lens onto acousto-optic modulator (AOM) at its Bragg angle. AOM modulates the intensity of laser beam in sinusoidal or square-wave whose frequency is ramped in time by local oscillator (LO). The resulting 0th-order beam is spatially filtered by adjustable iris and launched into multimode optical fiber (MMF) by a standard fibre optic coupler equipped with 10X objective.

At the far-end, the optical fiber collects either Fresnel backreflection or fluorescence from SPA or RU solution, and guides it back to AOM. The returning signal is focused by the fibre optic coupler onto AOM again at its old Bragg angle. The 0th-order beam of this second intensity modulation is collimated by the lens, filtered by iris and interference filter, and finally detected by a lowpass PIN diode detector. A digital oscilloscope collects the voltage input to LO as “input” data and the voltage output from PIN detector as “output” data, and uploads to computer for data fitting.

When measuring Fresnel backreflection as the auto correlation, my scheme resembles a hybrid of modulation OFDR and correlation OTDR, because the correlation is measured as function of modulation frequency. When measuring fluorescent response as the cross correlation, my scheme resembles frequency domain fluorometer, because the lifetime is determined through its sinusoidal response.

6.2 Components

6.2.1 Laser

Argon laser was pre-configured for single-line and single-frequency operation at wavelength λ = 457.9nm, with bandwidth Δν = 3MHz, and coherence length \( L_{coh} = c/Δν = 100m \). Its output was linearly polarized and was stable at 15mW ± 0.5% without optical feedback problem. The laser polarization was
Figure 6.1: Schematic diagram of experimental setup. All components are from Ontario Laser and Lightwave Research Center (OLLRC) or Department of Electrical Engineering at U of T.

10X 10X objective fibre optic coupler (Newport)
AOD 140MHz quartz oscillator driver (NEC OD-8802)
AOM Lead Molybdate (PbMoO₄) acousto-optic modulator (NEC OD-8813)
Ar laser Argon laser (Spectra-Physics 2020-05)
BS Broadband polarization beamsplitter cube (Newport 10FC16/PB.3)
detector Low-noise J-FET input Quad Op-Amp (TL 074)
filter Interference bandpass filters (Melles Griot 460 ±10nm, 550 ±20nm, 600 ±20nm)
λ/2 Broadband polarization rotator (Newport PR-550)
 lens Bi-convex optical glass lens (Newport KBX 076)
LO 4MHz Pulse/Function Generator (Wavetek 187)
MMF 51.2μm diameter core multimode fiber (Corning 1517)
oscilloscope 100MHz digital storage oscilloscope (Tektronics 2230)
PIN Blue enhanced PIN diode (Silicon Detector Corp. SD 200-12-12-041)
ramp Programmable Ramp Generator (Burleigh RC-43)
RU Ruthenium tris-bipyridyl dichloride, hexahydrate (Molecular Probes R-1498)
SPA N-(3-sulfopropyl) acridinium, inner salt (Molecular Probes S-460)
Figure 6.2: Pictures of optical (top) and electrical (bottom) equipments
rotated by halfwave ($\lambda/2$) plate for maximum transmission through beamsplitter (BS).

Because of very long coherence length, Fresnel backreflection coming out of MMF contains speckle pattern which is local fluctuation of intensity due to interference amongst modes. In addition, 60Hz "humming" vibration from the laser cooling mechanism contributes to the fluctuation by inducing an acoustic perturbation of fiber index. But, since there is no mode dependent coupling anywhere in the signal path, the total intensity over entire optical beam should be constant despite the local phase noise. Nonetheless, some intensity noise may be introduced because, as the fiber optic launcher experiences the vibration, its focused spots move about over the fiber end-face and over the AOM aperture.

6.2.2 AOM and AOD

Acousto-optic modulator (AOM) and its matching driver (AOD) were salvaged from old NEC laser fax machine. AOM, shown in Fig 6.3, consists of Lead Molybdate (PbMoO$_4$) crystal with 633nm anti-reflection coating and 10mm Lithium Niobate piezoelectric transducer. AOD is 140MHz quartz oscillator which is amplitude modulated by a RF mixer. Since the acoustic speed is $v_a = 3.63$km/s along $\hat{z}$ axis [B12, CRC, N25], 140MHz carrier frequency produces acoustic wavelength of $\Lambda = 25.9\mu$m. The AOM is Bragg type because $Q = 17 \gg 1$.

PbMoO$_4$ is tetragonal (4/m point group) system with strain-optic coefficient at 633nm [B12, CRC, Yariv]

$$p_{ij} \equiv [p] = \begin{bmatrix} 0.240 & 0.240 & 0.255 & 0 & 0 & 0.017 \\ 0.240 & 0.240 & 0.255 & 0 & 0 & -0.017 \\ 0.175 & 0.175 & 0.300 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.067 & -0.01 & 0 \\ 0 & 0 & 0 & 0.01 & 0.067 & 0 \\ 0.013 & -0.013 & 0 & 0 & 0 & 0.050 \end{bmatrix}$$

(6.1)

It is interesting to note that $n_1^3 p_{13} \approx n_3^3 p_{33}$ holds true for PbMoO$_4$ throughout its optical spectrum [B12]. This means that, from Eqn 4.10, the Bragg diffraction is isotropic and independent of polarization between $E$ and $K$. Since BS reflects (TE) wave polarized along $\hat{z}$ and transmits (TM) wave polarized along $\hat{z}$, $n_3$ acts on the laser excitation on the first pass, and $n_1$ acts on the returning signal that actually reaches PIN detector. The relevant refractive indexes (in bold) and their Bragg angles are [Bass]

<table>
<thead>
<tr>
<th></th>
<th>458nm</th>
<th>550nm</th>
<th>600nm</th>
<th>633nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_1, n_2$</td>
<td>2.553</td>
<td>2.436</td>
<td>2.403</td>
<td>2.386</td>
</tr>
<tr>
<td>$n_3$</td>
<td>2.363</td>
<td>2.294</td>
<td>2.273</td>
<td>2.262</td>
</tr>
<tr>
<td>$\theta_B$</td>
<td>0.214°</td>
<td>0.250°</td>
<td>0.276°</td>
<td></td>
</tr>
</tbody>
</table>

The transfer function of AOM/AOD is plotted in Fig 6.4. In order to obtain a linear response from AOM, the electrical input to AOD must be biased at the linear portion of transfer function. The best sinusoidal output with 25dB harmonic suppression was observed with $2.2 \pm 1.0$V input to AOD. For square-wave modulation, biasing does not matter since it works on simple "on/off" principle. Few parameters of 0th-order beam that have been measured at 458nm are

| $n_1, \Delta \varphi, \Delta n$ | 90.5%, 2.51, $18.3 \times 10^{-8}$ |
| $f_{3dB}$ | 8MHz |

39
Figure 6.3: Picture of inside AOM
The Bragg condition is not fully satisfied in my experiment mainly due to optical divergence and λ-dependence. Especially, the λ-dependence of diffraction efficiency becomes important when measuring SPA and RU fluorescence at 550nm and 600nm respectively, since AOM was aligned at 458nm for both the laser excitation and the returning signals. The efficiency decreases not only because of the longer wavelengths, but also because the old incident angle ($\theta_0 = 0.214^\circ$) no longer matches the new Bragg angles. Using small angle approximation in Eqn 4.15, $\eta_i$ decreases from 90.5% at 458nm to

<table>
<thead>
<tr>
<th></th>
<th>550nm (SPA)</th>
<th>600nm (RU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta \alpha = K \Delta \theta L$</td>
<td>1.50</td>
<td>2.62</td>
</tr>
<tr>
<td>$\Delta \varphi \propto 1/\lambda$</td>
<td>2.09</td>
<td>1.92</td>
</tr>
<tr>
<td>$\eta_i$</td>
<td>61%</td>
<td>35%</td>
</tr>
</tbody>
</table>

The optical divergence of returning signal will reduce the diffraction efficiency even further, because AOM diffracts only those portion of the focused beam that satisfies Bragg condition. In addition, since the half-angle divergence ($0.86^\circ$) of 10X objective is greater than the Bragg angle ($0.51^\circ$), 0th and 1st order beams of returning signal overlaps. This reduces the signal modulation depth and contributes to intensity noise, because the PIN detector is not integrating over the entire 0th-order beam.

The double pass through AOM is complicated by

- different Bragg angle and efficiency for excitation (at 458nm) and fluorescence signal (at 550nm or 600nm),
- different focal length for 10X objective (1.48cm) and lens (20cm), and
- different beam profile for first pass (nearly parallel laser beam) and second pass (11.5° MMF aperture).

These asymmetries mean that optimal coupling condition for one way does not automatically lead to optimal coupling for the other way. A compromise is unavoidable.

### 6.2.3 MMF

I used 50m long step-index MMF with 25.6μm core radius and 62.5μm cladding radius. Some of material data at 458nm are
The attenuation coefficient at 458 nm was calculated from 2.3 dB/km given at 850 nm, using the fact that most of the propagation loss occurs through Rayleigh scattering where $\gamma \propto \lambda^{-4}$. From Eqn 2.18, the impulse response of Rayleigh backscattering is an exponential decay with "lifetime" of 0.77 $\mu$s. Although the critical length in Eqn 2.11 is 40 m, it is convenient to take $L_c = 50$ m which is equal to the fiber length.

From Eqns 2.10 and 2.11, 1-way propagation in MMF produces modal dispersion of $\sigma_1 = 0.66$ ns, and 2-way modal dispersion increases to $\sigma_1 \sqrt{2} = 0.93$ ns. This limits the spatial resolution to

$$\delta L = \frac{c}{2n_1} \sigma_1 \sqrt{2} = 0.10$$

Also, the use of 550 ± 20 nm and 600 ± 20 nm interference filters to isolate SPA and RU fluorescence is equivalent to having 40 nm spectral source which produces a substantial material dispersion according to Eqn 2.12. Then, using Eqn 2.9, the total dispersion for fluorescent signal adds up to $\sqrt{2\sigma_1^2 + \sigma_2^2}$.

<table>
<thead>
<tr>
<th></th>
<th>550 nm (SPA)</th>
<th>600 nm (RU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_1$</td>
<td>0.66 ns</td>
<td>0.66 ns</td>
</tr>
<tr>
<td>$\sigma_2$</td>
<td>0.77 ns</td>
<td>0.59 ns</td>
</tr>
<tr>
<td>$\sqrt{2\sigma_1^2 + \sigma_2^2}$</td>
<td>1.2 ns</td>
<td>1.1 ns</td>
</tr>
</tbody>
</table>

Fortunately, the total dispersion of SPA and RU signals are insignificant compared to their lifetimes of about 30 ns and 350 ns.

Maximum scattering takes place when the excitation is continuous and constant, because the entire length of fiber acts as scattering source. And maximum reflection occurs when the fiber end-face is clean and perpendicular. If the excitation input $x(t) = 1$ is launched into MMF, then the steady-state output coming back is $y(t) = h(t) \ast x(t) = H(0)$. Using Eqns 2.19 and 2.22, the Fresnel and Rayleigh components of the output are

<table>
<thead>
<tr>
<th></th>
<th>in air</th>
<th>in water</th>
</tr>
</thead>
<tbody>
<tr>
<td>near-end: $R_0$</td>
<td>0.034</td>
<td>0.034</td>
</tr>
<tr>
<td>far-end: $R_L e^{-2\gamma L}$</td>
<td>0.018</td>
<td>0.0010</td>
</tr>
<tr>
<td>scattering: $(1 - e^{-2\gamma L})S/2$</td>
<td>0.0017</td>
<td>0.0017</td>
</tr>
</tbody>
</table>

When the fiber tip is held in air ($n_0 = 1.0$), Fresnel reflection is about 10 dB above Rayleigh scattering. When the fiber tip is immersed in aqueous solution ($n_0 = 1.33$), Fresnel reflection falls below scattering; but, this is irrelevant because fluorescence is measured at different wavelength. Fresnel reflection from the far-end and Rayleigh scattering are depolarized for all practical purpose and are split 50/50 by BS. However, Fresnel reflection from the near-end is still polarized and passes straight through BS.

Because the laser excitation has shorter penetration in RU solution than in SPA, the RU fluorescence is "closer" and has better coupling to the fiber end-face. Nonetheless, $Q_f$ must be included when calculating the total amount of fluorescence that is collected. Using $n_0 = 1.33$ and $b = a/\tan \theta_M = 168 \mu$m in water, the total collection efficiencies for SPA and RU fluorescence become
There is about 5dB difference between SPA and RU signals. Considering that fiber attenuation, diffraction efficiency, AOM attenuation, filter attenuation, and PIN diode responsivity are all \( \lambda \)-dependent, mostly in RU's favour, the final signals should be of similar magnitude. Furthermore, had I used double fiber configuration in my experiment, the fluorescent signals would be 9-13dB lower since \( G(L, 1.44\alpha) = 0.0023 \) for SPA and \( G(L, 1.44\alpha) = 0.0078 \) for RU.

Comparing the collection efficiency of Fresnel reflection (\( R_L \)) and fluorescence (\( Q' \eta_c \)) at the far-end, the Fresnel backreflection is 23dB higher than SPA fluorescence and 27dB higher than RU fluorescence. This is consistent with experimental observation of few \( \mu \)W for Fresnel signal and few nW for both SPA and RU signals.

Yellow-orange luminescence [B16] is observed from the fiber's plastic buffer. Fortunately, even though this luminescence can be seen easily by naked eyes, its contribution at 550nm and 600nm is below the noise floor of PIN detector. This is due to very poor evanescent coupling between the cladding source and the propagating modes in the core.

### 6.2.4 SPA and RU

SPA and RU compounds were selected based on their single exponential decay, long lifetime, visible absorption and emission spectrum, large \( \lambda \) shift, high \( Q' \), stable photochemistry, easy preparation, and low cost. While SPA is a regular fluorescent compound, RU is fundamentally a phosphorescent compound whose emission is greatly enhanced by spin-orbit coupling due to heavy-atom perturbation. Structurally, RU is a \( d^2 \)-transition metal complex with 2,2'-bipyridine ligand in octahedral configuration. Pertinent transitions in the visible spectrum are metal-to-ligand charge-transfer states (MLCT) which are formed when \( e^- \) is excited from metal's \( d \)-orbital to ligand's \( \pi^* \)-antibonding orbital. Referring to Fig 3.1, the absorption state is \( ^1 \text{(MLCT)} \) and the emission state is the spin-forbidden \( ^3 \text{(MLCT)} \). [B13]

Chemical formula for SPA and RU are shown in Fig 6.5, and their absorption and emission spectrum are included in Fig 6.6. The SPA and RU solutions are prepared as air-saturated aqueous solutions at room temperature, and in rather heavy concentration to maximize the fluorescence. Some pertinent material data are [Probes].
Figure 6.6: Absorption and emission spectrum for SPA (top) and RU (bottom)
Figure 6.7: Calibration data for LO's voltage-to-frequency conversion

\[ f = 1023.5V + 13.98 \]
\[ R^2 = 0.999949 \]

<table>
<thead>
<tr>
<th>molecular mass concentration ( c )</th>
<th>SPA/water</th>
<th>RU/water</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \epsilon, \alpha ) at 458nm</td>
<td>301g/mole</td>
<td>748g/mole</td>
</tr>
<tr>
<td>QY</td>
<td>458( \mu )M</td>
<td>468( \mu )M</td>
</tr>
<tr>
<td>( \tau )</td>
<td>1000/cm M, 105/m</td>
<td>14000/cm M, 1510/m</td>
</tr>
<tr>
<td></td>
<td>1.033 [B8]</td>
<td>363.5 ( \pm ) 0.4ns [B3]</td>
</tr>
<tr>
<td></td>
<td>31.6 ( \pm ) 0.1ns [B1]</td>
<td>350 ( \pm ) 25ns [B5]</td>
</tr>
</tbody>
</table>

where \( 10^{-cz} = e^{-\alpha z} \).

6.2.5 LO

LO is basically a voltage controlled oscillator, converting input voltage \( V \) to output frequency \( f \). The calibration data for this conversion is recorded manually using lockin amplifier\(^1\) and frequency counter\(^2\), and is plotted in Fig 6.7. Linear regression gives [Guttman]

\[ f = 1023.5V + 13.98, \quad R^2 = 0.999949 \quad (6.3) \]

with \( f \) in [kHz] and \( V \) in [V]. A stable and accurate frequency generation is essential to my correlation scheme; regrettably, LO exhibited some thermal drift which limits the degree of accuracy that can be achieved in my experiment.

Since LO frequency is swept 0–4MHz every second, the modulation frequency that the laser excitation experiences in the first pass is different from that the returning signal experiences in the second pass due to propagation delay \( \Delta t = 2Ln/c \). For 50m fiber, the difference is \( \Delta f = (4\text{MHz}/1s)0.5\mu s = 2\text{Hz} \). This beat frequency is difficult to deal with because it appears as slow swing of the entire signal baseline.

\(^1\)Stanford Research System SR530
\(^2\)Fluke 1900A
6.2.6 PIN Detector

Fig 6.8 shows the schematic diagram of detector that I used in the experiment. Essentially, it is DC-coupled lowpass transimpedance amplifier with the PIN diode in photovoltaic mode (0V bias) so that diode current \( I \) is proportional to optical power \( P \), [Jenkins]

\[
I = I_0 \left( e^{V/kT} - 1 \right) - R_\Lambda P = -R_\Lambda P
\]

(6.4)

where \( R_\Lambda = 0.2-0.3 \text{A/W} \) is the responsivity of diode for 458–600nm. The final output signal becomes

\[
V_3 = 2V_r + \left( \frac{R_2}{R_1} + 1 \right) R_0 I, \quad f \ll f_0, f_3
\]

(6.5)

By trial and error, the upper limit of \( f_0, f_3 \approx 160\text{Hz} \) seemed sufficient to filter out noise without excessive smoothing. However, the circuit in Fig 6.8 is “home-made”; and, as such, the effects of improper grounding and stray capacitance are unavoidable even under the best of circumstances.

Unfortunately, after all the experiments were done, I realized that detector in Fig 6.9 would have been simpler and better. It is AC-coupled bandpass circuit with the final output signal

\[
V_2 = -\left( \frac{R_3}{R_2} + 1 \right) R_0 I, \quad f_1 \ll f \ll f_0
\]

(6.6)

Since the correlation output is repeated every ramp interval, this ramp frequency constitutes the lowest frequency component of output signal. Also, the output sits on top of large DC bias. The bandpass detector, with low cut-off \( f_1 \) set to the ramp frequency and with high cut-off \( f_0 \) set sufficiently high but not too high, would filter out the large DC bias, slow drifts, and fluctuations which plagued my experiment.

6.3 Experimental Procedures

Preparations involving sample solutions, fiber, laser, LO, and AOM alignments are summarized as follows:

- SPA/water (458\text{\textmu}M) and RU/water (468\text{\textmu}M) solutions are prepared using laboratory grade water.
- Both ends of approximately 50m long MMF are stripped of plastic buffer with Polystripper\textsuperscript{R}, cleaned with acetone, and cleaved with a fiber cleaver. Since fiber coupling depends critically on the quality of end-face, it is important to obtain a cut that is flat and perpendicular at least across the fiber core.
- The Ar laser output is set to 15mW at 458nm.
- Ramp generator is set to ramp 0–4V at every 1 second interval, and LO converts this to a corresponding frequency ramp 0–4MHz every second.
- Fig 6.10 (top) illustrates the aligning of AOM for the first pass. The Ar laser beam is focused by lens so that the spot size at AOM is within its active aperture. The fiber coupler is adjusted for maximum coupling into MMF by monitoring the optical power coming out of the far-end.
Figure 6.8: DC-coupled lowpass detector, using blue enhanced PIN diode and low-noise J-FET input Quad Op-Amp

\[
V_0 = \frac{-R_0 I}{1 + jf/f_0} \quad V_1 = \left( \frac{R_2}{R_1} + 1 \right) V_0 \quad V_2 = 2V_r - V_1 \quad V_3 = \frac{V_2}{1 + jf/f_3} \quad f_3 = \frac{1}{2\pi R_3 C_3}
\]

Figure 6.9: Simpler AC-coupled bandpass detector

\[
V_0 = \frac{-R_0 I}{1 + jf/f_0} \quad V_1 = \frac{jf/f_1}{1 + jf/f_1} V_0 \quad V_2 = \left( \frac{R_2}{R_2} + 1 \right) V_1
\]

Figure 6.10: Aligning AOM for first pass (top) and second pass (bottom)
• Fig 6.10 (bottom) illustrates the aligning of AOM for the second pass. Ar laser is launched from the far-end to simulate and make visible the path of returning signal. The fiber coupler is adjusted until the focused spot at AOM is within the same aperture that the first pass went through, thereby maintaining the same Bragg angle. Basically, AOM is geometrically aligned for optimum performance at 458 nm; the subsequent decrease in performance at 550 nm and 600 nm is accepted as a compromise in return for easier alignment.

The experimental procedures of actually measuring Fresnel backreflection (for fiber length) and fluorescent response (for SPA and RU lifetimes) are as follows:

1. Decide on the type of measurement. When measuring Fresnel reflection, the fiber tip is held in air. When measuring fluorescent response, the fiber tip is immersed in SPA or RU solution as shown in Fig 6.11. This maximizes the collection efficiency, and represents the simplest configuration because there are no external components to align. Of course, when changing from one measurement to another, the fiber tip must be washed thoroughly with distilled water.

2. To reduce contamination of signal, 460 $\pm$ 10 nm interference filter is placed in front of PIN detector when measuring Fresnel reflection, 550 $\pm$ 20 nm filter when measuring SPA fluorescence, and 600 $\pm$ 20 nm filter when measuring RU fluorescence.

3. The gain, reference bias, and bandwidth of PIN detector, described in Fig 6.8, are determined by trial and error. Generally, Fresnel signal was few $\mu$W, and SPA and RU signals were both about few nW.

4. Finally, both the ramp input and the correlation output are sampled with digitizing oscilloscope using built-in exponential averaging algorithm. The data set is then uploaded to computer for data analysis and ensemble averaging. Raw data for Fresnel reflection, SPA fluorescence, and RU fluorescence are shown in Figs 6.12, 6.13, and 6.14, respectively.

5. So far the optical fiber is nominally 50 m long. In order to assess the resolution in fiber length measurement, 1.01 m is cleaved off the far-end, making the fiber 49 m long nominally. Fresnel reflection is then measured by repeating steps 1–4. A raw data for this is shown in Fig 6.15.

I tried to collect as many sets of data as possible within the time allocated by OLLRC lab. Unfortunately, I managed to collect only 35 data sets for 50 m fiber, 16 data sets for SPA fluorescence, 13 data sets for RU fluorescence, and 10 data sets for 49 m fiber. Collecting more data sets will help, of course. But a fundamental improvement in the accuracy and confidence of parameter estimation can only occur when better quality equipments are used.

6.4 Data Analysis

When the bottom graphs in Figs 6.12, 6.13, and 6.14 are compared to the top graphs in Figs 5.3, 5.4, and 5.5, one can see that there is very good correspondence between the experimental measurements and the calculated models. Using Student-$t$ values for 95% confidence interval ($\alpha = 0.05$), the length of
Figure 6.11: Pictures of SPA (top) and RU (bottom) fluorescence
Figure 6.12: Raw data for Fresnel backreflection from 50m fiber. The voltage input (top) to LO and the correlation output (bottom) from PIN detector are shown for one complete ramp period. The excitation waveform was square-wave.
Figure 6.13: Raw data for SPA fluorescence using 50m fiber. The voltage input (top) to LO and the correlation output (bottom) from PIN detector are shown for one complete ramp period. The excitation waveform was square-wave.
Figure 6.14: Raw data for RU fluorescence using 50m fiber. The voltage input (top) to LO and the correlation output (bottom) from PIN detector are shown for one complete ramp period. The excitation waveform was square-wave.
Figure 6.15: Raw data for Fresnel backreflection from 49m fiber. The voltage input (top) to LO and the correlation output (bottom) from PIN detector are shown for one complete ramp period. The excitation waveform was square-wave.
50m and 49m optical fibers are estimated to be

\[
L_{50} = 50.28 \pm 0.27m \left( t_{34:0.025} \frac{0.773}{\sqrt{35}} \right)
\]

\[
L_{49} = 48.53 \pm 0.24m \left( t_{9:0.025} \frac{0.329}{\sqrt{10}} \right)
\]

\[
L_{50} - L_{49} = 1.75 \pm 0.34m \left( t_{36:0.025} \sqrt{\frac{0.773^2}{35} + \frac{0.329^2}{10}} \right)
\]

The 0.27m uncertainty for \( L_{50} \) is close to (rms) modal dispersion limit of 0.10m in Eqn 6.2, and represents almost 2 orders of magnitude improvement over the DFT resolution of 13m in Eqn 5.20. Unfortunately, the true difference of 1.01m between 50m and 49m fibers lies outside the confidence interval.

The lifetime of SPA and RU fluorescence are estimated to be

\[
(\text{SPA}) \tau = 28.3 \pm 3.5ns \left( t_{15:0.025} \frac{6.64}{\sqrt{16}} \right)
\]

\[
(\text{RU}) \tau = 360 \pm 28ns \left( t_{12:0.025} \frac{46.7}{\sqrt{13}} \right)
\]

The SPA and RU lifetimes were also determined in time domain, using a conventional pulse excitation setup in Chemical Sensor Group at Erindale campus of University of Toronto. The excitation source was \( N_2 \) laser with 10ns pulse width at 337nm. Fluorescein was used as reference because it exhibits a stable single lifetime of 3.8-4.3ns [A7, A8] over the emission spectrum of interest. The fluorescent decays are shown in Fig 6.16 for SPA-Fluorescein pair and in Fig 6.17 for RU-Fluorescein pair. Setting the reference lifetime at 4.0ns in Eqn 3.8, the estimated lifetimes are

\[
(\text{SPA}) \tau = 29.2ns \pm 0.1\%
\]

\[
(\text{RU}) \tau = 379ns \pm 0.1\%
\]

which are consistent with Eqs 6.10 and 6.11, as well as, with the values cited from literature in Section 6.2.4.
Figure 6.16: Fluorescence decay of SPA sample $y(t)$ and Fluorescein reference $x(t)$ at 550nm

Figure 6.17: Fluorescence decay of RU sample $y(t)$ and Fluorescein reference $x(t)$ at 600nm
Chapter 7

Conclusions

In my thesis, I demonstrate a fibre optic sensor for measuring fluorescence lifetime, in which the main signal processing is performed entirely by optical devices. It has several distinct characteristics and advantages when compared to the conventional time domain and frequency domain fluorometers:

- A single optical fiber is used to carry the laser excitation and to collect fluorescent response or Fresnel backreflection. This gives remote measurement capability which is important for the development of fibre optic sensors.

- The optical signal passes twice through AOM which acts as an optical bidirectional mixer. Since the double modulation occurs at almost same frequency, the signal processing is basically a homodyne scheme. Because AOM is the only active component, system bandwidth can be easily upgraded by replacing it with another optical modulator of higher bandwidth.

- The DC output from the PIN detector becomes auto or cross correlation. The detector's bandwidth is independent of modulator's bandwidth due to somewhat unusual change of domain brought about by the correlation operation. Because only DC component from AOM mixing is measured, the PIN detector does not have problems associated with wide bandwidth optical detection and high frequency electronic signal processing.

- Since the modulation frequency of AOM is ramped, the input and output data are repeated at every ramp period and are measured as functions of frequency.

- Continuous wave excitation has lower peak power than pulse excitation, so that there is less likelihood of damaging a sample.

- Although the experiment uses bulk components, the system is simple enough to be integrated into a single “in-line” device.

Furthermore, I address theoretical problems involved in using optical fiber to excite and collect fluorescence from remote medium. The fluorescence coupling efficiency of multimode and singlemode fiber is derived, and the waveguide effect of optical fiber on the propagating signals is analyzed in detail. I hope that the foundation laid in this thesis helps reader to understand better the workings of optical fiber sensor, whether it is for measuring temperature, pH, O₂ concentration, or even epoxy resin cure [See Appendix B].
From experiment, I have successfully determined the fluorescent lifetime of N-(3-sulfopropyl) acridinium (SPA) and ruthenium tris-bipyridyl dichloride (RU), and the fiber length of multimode optical fiber sensor. SPA and RU compounds were chosen because they have a stable single lifetime, visible spectrum, and relatively high quantum yield; and multimode fiber was chosen because it has large core which facilitates better coupling. The main results of data analysis are as follows:

<table>
<thead>
<tr>
<th>Fiber</th>
<th>Estimation</th>
<th>DFT Limit</th>
<th>Dispersion</th>
</tr>
</thead>
<tbody>
<tr>
<td>SPA L</td>
<td>50.28 ± 0.27m</td>
<td>13m</td>
<td>0.10m</td>
</tr>
<tr>
<td>SPA τ</td>
<td>28.3 ± 3.5ns</td>
<td>125ns</td>
<td>1.2ns</td>
</tr>
<tr>
<td>RU τ</td>
<td>360 ± 28ns</td>
<td>125ns</td>
<td>1.1ns</td>
</tr>
</tbody>
</table>

My correlation scheme gives an order of magnitude improvement over the conventional DFT method. The uncertainties represent the usual 95% confidence interval for ensemble average. Ultimately, the resolutions are limited by optical fiber dispersions; only modal dispersion is present in fiber length measurement, but both modal and material dispersions are present in fluorescent lifetime measurement.

The lifetimes are consistent with literature values and with the values deconvoluted from pulse excitation method. Unfortunately, however, the fiber length shows poor internal consistency, since the actual 1.01m that is cleaved off the far-end is estimated as 1.75 ± 0.34m at 95% confidence. This inconsistency is attributed to the drift and fluctuation of modulation frequency coming from LO.

The coupling efficiency of Fresnel backreflection is at least 10dB greater than Rayleigh backscattering, and 23–27dB greater than SPA and RU fluorescence. This is consistent with experimental observation of few µW for Fresnel signal and few nW for both SPA and RU signals. In collecting SPA and RU fluorescence, as expected, the single fiber configuration is 9–13dB more efficient than double fiber configuration.

AOM is the key component in my correlation scheme, because it performs the modulation of laser excitation in the first pass and the optical mixing necessary for auto and cross correlations in the second pass. Using one AOM for both functions allows the signal processing to be done as much as possible in optical domain with minimum external components. Diffraction efficiency and Bragg angle depend on the input wavelength. Due to phase-mismatch, the Bragg diffraction decreases from 90.5% efficiency for laser excitation (458nm) to 61% efficiency for SPA fluorescence (550nm) and to 35% efficiency for RU fluorescence (600nm). The efficiency decreases even more if optical divergence is considered.

The merits of my fibre optic fluorescence lifetime sensor become obvious when one realizes that an experimental setup with only 4MHz bandwidth is capable of measuring fluorescent lifetime of 28.3 ± 3.5ns and 360 ± 28ns. Shorter lifetimes can be measured by simply upgrading LO and AOM to a higher bandwidth, with no changes to the other components, experimental layout, or data analysis. Although the 4MHz bandwidth is rather low for practical applications, it is sufficient for the purpose of my thesis which is to demonstrate a new concept in optical fiber sensor, fluorescence lifetime measurement, and optical signal processing.
Appendix A

Formula and Tables

A.1 Definitions

\[ x(t) * y(t) = \int_{-\infty}^{\infty} x(u)y(t-u)du \] \hspace{1cm} (A.1)

\[ R_{xx}(t) = \int_{-\infty}^{\infty} x(u)x(u+t)du \] \hspace{1cm} (A.2)

\[ R_{xy}(t) = \int_{-\infty}^{\infty} x(u)y(u+t)du \] \hspace{1cm} (A.3)

\[ \eta(t) = \begin{cases} 1, & |t| < 1/2 \\ 0, & |t| > 1/2 \end{cases} \] \hspace{1cm} (A.4)

\[ \omega(t) = \begin{cases} 1 - |t|, & |t| < 1 \\ 0, & |t| > 1 \end{cases} \] \hspace{1cm} (A.5)

\[ \text{sinc}(x) = \frac{\sin \pi x}{\pi x} \] \hspace{1cm} (A.6)

\[ \sum_{n=1,3,5} \frac{y}{1 + (ny)^2} = \frac{\pi}{4} \tanh \left( \frac{\pi}{2y} \right) \] \hspace{1cm} (A.7)

A.2 Impulse Function

The impulse function \( \delta(t) \) has the following properties: [Brigham]

\[ \delta(t) = 0, \quad t \neq 0 \] \hspace{1cm} (A.8)

\[ \int_{-\infty}^{\infty} \delta(t) = 1 \] \hspace{1cm} (A.9)

\[ |a| \delta(at) = \delta(t) \] \hspace{1cm} (A.10)

\[ h(t)\delta(t-a) = h(a)\delta(t-a) \] \hspace{1cm} (A.11)

\[ \delta(t-a) * \delta(t-b) = \delta(t-(a+b)) \] \hspace{1cm} (A.12)

A.3 Solid Angles

Consider a conical aperture along \( \hat{z} \) axis with half-angle \( \Theta \), so that \( dS \) is differential spherical area normal to the radius vector \( \hat{r} \), \( \theta \) is angle between \( \hat{z} \) and \( \hat{r} \), and \( dS \cos \theta \) is differential planar area normal to \( \hat{z} \). The spherical solid angle of
aperture is
\[
\omega = \int_S \frac{dS}{r^2} = \int_\theta=0 \int_\phi=0 \frac{(r \, d\theta)(r \sin \theta \, d\phi)}{r^2} = 2\pi (1 - \cos \Theta)
\] (A.13)

and the planar solid angle is defined as [Wyatt]
\[
\Omega = \int_S \frac{dS \cos \theta}{r^2} = \int_\theta=0 \int_\phi=0 \frac{(r \, d\theta)(r \sin \theta \, d\phi) \cos \theta}{r^2} = \pi \sin^2 \Theta
\] (A.14)

### A.4 Table of Fourier Transform Pairs

The following Fourier Transform pairs
\[
h(t) = \int_{-\infty}^{\infty} H(f) e^{j2\pi ft} df \quad \leftrightarrow \quad H(f) = \int_{-\infty}^{\infty} h(t) e^{-j2\pi ft} dt
\] (A.15)

have been selected from Refs [Brigham, Bracewell]. If a function is defined with conditional domain, then it is zero outside the domain and is equal to the midpoint at the discontinuity.

\[
\begin{align*}
\text{(even)} + j(0) & \leftrightarrow \text{(even)} + j(0) \\
\text{(odd)} + j(0) & \leftrightarrow \text{(odd)} + j(0) \\
(0) + j(\text{even}) & \leftrightarrow (0) + j(\text{even}) \\
(0) + j(\text{odd}) & \leftrightarrow (0) + j(\text{odd})
\end{align*}
\] (A.29)

\[
\begin{align*}
\delta(t) & \leftrightarrow 1 \\
\sum_{n=-\infty}^{\infty} \delta(t - n) & \leftrightarrow \sum_{n=-\infty}^{\infty} \delta(f - n) \\
\sum_{n=-\infty}^{\infty} \delta(t - nT) & \leftrightarrow \frac{1}{T} \sum_{n=-\infty}^{\infty} \delta \left( f - \frac{n}{T} \right) \\
1 & \leftrightarrow \delta(f) \\
1, \ t > 0 & \leftrightarrow \frac{\delta(t)}{2} + \frac{1}{j2\pi f}
\end{align*}
\] (A.30)
\begin{align*}
\tau(t) & \iff \text{sinc}(f) \\
\Lambda(t) & \iff \text{sinc}^2(f) \\
\cos(2\pi at) & \iff \frac{\delta(f - a) + \delta(f + a)}{2} \\
\sin(2\pi at) & \iff \frac{\delta(f - a) - \delta(f + a)}{2j} \\
e^{-t/r}, \ t > 0 & \iff \frac{\tau}{1 + j2\pi fr} \\
e^{-|t|/r} & \iff \frac{2\tau}{1 + (2\pi fr)^2} \\
e^{-\pi r^2} & \iff e^{-\pi r^2} \\
e^{-t^2/\alpha^2} & \iff \alpha \sqrt{\pi} e^{-\pi \alpha^2 r^2} \\
\frac{\delta(t - \Delta t) + \delta(t + \Delta t)}{2} & \iff \cos(2\pi f \Delta t) \\
\frac{1}{2\tau} e^{-|(t - \Delta t)/r|}, \ |t| > \Delta t & \iff \frac{\cos (2\pi f \Delta t + \tan^{-1}(2\pi f r))}{\sqrt{1 + (2\pi f r)^2}} \tag{A.44}
\end{align*}

**A.5 Decomposition into Fourier Series**

Consider a waveform \( x(t) \) which consists of \{ \( h(t) \), \( |t| < T/2 \} \) repeated at every \( T \) interval or at frequency \( 1/T \). The time domain expression and the corresponding complex Fourier series are [Brigham]

\[
x(t) = h(t) \ast \sum_{n=-\infty}^{\infty} \delta(t - nT) = \sum_{n=-\infty}^{\infty} \alpha_n e^{j2\pi nt/T} \tag{A.45}
\]

Applying FT to both sides and solving for \( \alpha_n \),

\[
H(f) \sum_{n=-\infty}^{\infty} \delta \left( f - \frac{n}{T} \right) = \sum_{n=-\infty}^{\infty} \alpha_n \delta \left( f - \frac{n}{T} \right) \tag{A.46}
\]

\[
\sum_{n=-\infty}^{\infty} \frac{1}{T} H \left( \frac{n}{T} \right) \delta \left( f - \frac{n}{T} \right) = \sum_{n=-\infty}^{\infty} \alpha_n \delta \left( f - \frac{n}{T} \right) \tag{A.47}
\]

\[
\alpha_n = \frac{1}{T} H \left( \frac{n}{T} \right) \tag{A.48}
\]

If \( h(t) \) is real and even, \( H(f) \) is also real and even. Then, the complex series becomes a cosine series,

\[
x(t) = \alpha_0 + \sum_{n=1}^{\infty} 2\alpha_n \cos \left( \frac{2\pi nt}{T} \right) \tag{A.49}
\]

**A.6 rms Width**

Root-mean-square (rms) width of function \( h(t) \) is defined as [Bracewell]

\[
\sigma^2 = \langle t^2 \rangle - \langle t \rangle^2 \tag{A.50}
\]

where

\[
\langle t \rangle = \frac{\int_{-\infty}^{\infty} t h(t) dt}{\int_{-\infty}^{\infty} h(t) dt} = -\frac{H'(0)}{2\pi j H(0)} \tag{A.51}
\]

60
\[ (t^2) = \frac{\int_{-\infty}^{\infty} t^2h(t)dt}{\int_{-\infty}^{\infty} h(t)dt} = -\frac{H''(0)}{4\pi^2 H(0)} \]  

(A.52)

The rms width of few common functions are

\[
\begin{align*}
\Gamma(t) & \rightarrow \sigma^2 = 1/12 \\
\Lambda(t) & \rightarrow \sigma^2 = 1/6 \\
e^{-t/r}, \ t > 0 & \rightarrow \sigma^2 = \tau^2 \\
e^{-|t|/r} & \rightarrow \sigma^2 = 2\tau^2 \\
e^{-t^2/2a^2} & \rightarrow \sigma^2 = a^2
\end{align*}
\]  

(A.53)  

(A.54)  

(A.55)  

(A.56)  

(A.57)

### A.7 Fiber-to-Fiber Coupling

Coupling efficiency \( \eta \) from MMF\(_1\) to MMF\(_2\) is important not only for splices but also for many fiber-tip sensor configurations. Using subscript 1 and 2 to denote the parameter of interest from respective fibers, [Ungar, Etten]

- diameter \( 2a \):
  \[ \eta = \min \left\{ \frac{a_2^2}{a_1^2}, 1 \right\} \]  
  (A.58)

- aperture NA:
  \[ \eta = \min \left\{ \frac{NA_2^2}{NA_1^2}, 1 \right\} \]  
  (A.59)

- index profile \( g \):
  \[ \eta = \min \left\{ \frac{1 + 2/g_1}{1 + 2/g_2}, 1 \right\} \]  
  (A.60)

- core index \( n \):
  \[ \eta = 1 - \left( \frac{n_1 - n_2}{n_1 + n_2} \right)^2 \]  
  (A.61)

- axial gap \( z \):
  \[ \eta = \frac{a^2}{(a + z \tan \theta_{NA})^2}, \quad \text{if } g = \infty \]  
  (A.62)

- radial shift \( 2r \):
  \[ \eta = \frac{\phi - \sin \phi}{\pi}, \quad \text{if } g = \infty \]  
  (A.63)

- angular tilt \( 2\theta \):
  \[ \eta = \frac{\phi}{\pi} - \frac{4 \sin \phi}{3\pi} + \frac{\sin 2\phi}{6\pi}, \quad \text{if } g = 2 \]  
  (A.64)

where \[ \frac{\cos \phi}{2} = \frac{r}{a} \quad \text{or} \quad \frac{\sin \theta}{\sin \theta_{NA}} \]  

(A.65)
Appendix B

Fluorescence Application in Cure Monitoring

B.1 Epoxy Resin and Composite

The term *epoxy* refers to molecule containing \( \text{C} \equiv \text{C} \) ring structure. The cure process breaks open the epoxy rings and converts epoxy monomers into network of crosslinked polymers which is characterized [Ency] by high adhesive strength, low shrinkage, good mechanical and thermal properties, high chemical and corrosion resistance, and good electrical insulation. To bring about some optimum characteristics, most commercial epoxy resins contain proprietary mixture of epoxies, curing agents (hardeners), accelerators, diluents, fillers, flexibilizers, and impurities (dyes, solvents, etc.) [Ency, Lubin]. The generic chemical formula of 2 popular epoxies and curing agents are shown in Figs B.1 and B.2, for which the ring opening reaction occurs mainly between the terminal glycidyl group and the primary amine group as follows:

\[
-\text{NH}_2 + H_2\text{C} \equiv \text{CHCH}_2 \rightarrow \text{OH} \]

One important application of epoxy resin is in *composite* [Ency] where strong and stiff fibers (usually glass, boron-tungsten, graphite, or Kevlar\(^1\)) are aligned in epoxy resin matrix. Most composites are manufactured by baking in an oven under pressure. Temperature provides heat needed to initiate and maintain the epoxy cure reactions, and pressure compacts the fiber matrix by squeezing out excess resin and gas pockets. Typically, the cure cycle follows a fixed time-temperature-pressure profile [08], even in the presence of chemical variation in resin mixture, unequal distribution in composite laminates, difference in preparation or storage, and other practical situations that come up in the real world.

The fixed cycle is simple and cheap to implement, but the quality of composite varies from batch to batch. Obviously, the composite does not attain its optimum properties when the epoxy resin is not fully cured; furthermore, unreacted epoxy resins serve as sites [O13] for crack initiation and propagation. The quality can be improved by using some feedback control which requires monitoring of parameters (temperature, pressure, viscosity, capacitance, conductance, etc.).

\(^1\)Kevlar, also known as PPTA or poly(p-phenyleneterephthalamide), is trademark of Dupont
Figure B.1: Generic formula of 2 epoxy resins: (top) DGEBA or diglycidyl ether of bisphenol A, and (bottom) TGDDM or tetraglycidyl diamino diphenyl methane

\[
\begin{align*}
\text{(top)} & \quad \text{H}_2\text{C} & \quad \text{CHCH}_2 & \quad \text{O} & \quad \text{C(} & \quad \text{CH}_3 & \quad \text{)}_2 & \quad \text{O} & \quad \text{CH}_2\text{HC} & \quad \text{CH}_2 \\
\text{(bottom)} & \quad \text{H}_2\text{C} & \quad \text{CHCH}_2 & \quad \text{N} & \quad \text{CH}_2\text{HC} & \quad \text{CH}_2 \\
\text{ } & \quad \text{H}_2\text{C} & \quad \text{CHCH}_2 & \quad \text{O} & \quad \text{CH}_2\text{HC} & \quad \text{CH}_2 \\
\end{align*}
\]

Figure B.2: Generic formula of 2 curing agents: (top) DDS or diamino diphenyl sulfone, and (bottom) DDM or diamino diphenyl methane

\[
\begin{align*}
\text{(top)} & \quad \text{H}_2\text{N} & \quad \text{SO}_2 & \quad \text{NH}_2 \\
\text{(bottom)} & \quad \text{H}_2\text{N} & \quad \text{CH}_2 & \quad \text{NH}_2 \\
\end{align*}
\]

acoustic impedance, refractive index, IR absorption, fluorescence, etc.) that affect or indicate the state of cure.

B.2 Fluorescence during Cure

If \( A \) and \( k \) are, respectively, rates of radiative and non-radiative decays from an emitting state of molecule, the quantum yield (\( QY \)) and the lifetime (\( \tau \)) of fluorescence are defined as

\[
QY = \frac{A}{A + k}, \quad \tau = \frac{1}{A + k}
\]  

(B.2)

The radiative decay remains fairly constant, but the non-radiative decays, such as rotational and vibrational relaxations, are influenced by the molecular environment. At the beginning, an un cured epoxy resin exhibits high molecular mobility which means fast \( k \), low \( QY \), and short \( \tau \). After full crosslinking polymerization, a cured epoxy resin exhibits low molecular mobility which leads to slow \( k \), high \( QY \), and long \( \tau \).

If the conversion from uncured to cured state occurs homogeneously throughout the resin, then \( QY \) and \( \tau \) would increase smoothly during cure. However, from phosphorescence studies [O10, O11, O12], ESR spectroscopy [O12], and electron and optical microscopy [O13], the initial formation of network and the subsequent growth in crosslinking are found to be nucleation process, in which the cure process occurs heterogeneously or locally with distinct boundary between the 2 states. Therefore, \( QY \) increases because the cured domain grows at the expense of the uncured domain, but \( \tau \) remains constant because the underlying nature of \( A \) and \( k \) within each domain do not change.
B.3 Cure Monitoring

Usually, one adds a dye or probe to an epoxy resin mixture, as long as it does not interfere with the cure reactions and does not overlap with other fluorescence already present. In some cases, one can take advantage of intrinsic fluorescence from epoxy resin itself. So far, the following mechanisms or parameters have been used for cure monitoring:

- increase in $QY$ which depends on local viscosity [O8, O31, O34, O39]
- shift in emission spectrum [O39, O56]
- increase in $QY$ of reactive probe that has similar structure and reactivity as the curing agent [O8, O31, O55]
- decrease in excimer fluorescence which depends on local “free volume” [O8, O31]
- increase in fluorescence polarization which depends on rotational diffusion [O31]
- longer recovery time after photobleaching due to slow diffusion [O54]

An accurate interpretation of data is difficult because of impurities and complex chemical reactions. But, fluorescence method is uniquely suited for fibre optic application [O39, O55, O56], since typical fluorescent spectrum of near-UV to near-IR wavelength is easily supported by many optical fibers.

Even though the fluorescent lifetime mechanism does not change during epoxy resin cure, if $\tau$ is measured using a fluorescent lifetime sensor that assumes single lifetime, as I have done in demonstrating my experiment, then $\tau$ will appear to increase during the cure, because the numerical algorithm cannot differentiate signals from 2 different domains and simply follows the growing (shrinking) contribution from the cured (uncured) domain. Consequently, the fluorescent lifetime can be used for cure monitoring of epoxy resin.

A more precise analysis requires distributed lifetime model where one assumes that varying molecular environments produce a distribution of fluorescent lifetimes. As the molecular compositions changes with cure, the profile of lifetime distribution changes accordingly. However, this requires more complicated data analysis than the one I used in my experiment.
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