The observable effects of stimulated emission on the optical spectra of an active medium include peak gain, fluorescence band narrowing and fluorescence lifetime shortening. The detection of these effects allows a decisive evaluation of the potential use of a given material as a lasing medium. The stimulated emission parameters of a laser material are the gain coefficient, the pumping threshold energy density for the beginning of stimulated emission, the population inversion density at threshold and stimulated emission cross section. Experimentally, the measurable quantities are the gain coefficient, the pumping threshold energy density and the fluorescence decay time as a function of pump power. From these quantities the stimulated emission parameters can be deduced. Optical gain of the material, in general expresses its efficiency to couple the incident input radiation to the output. Hence this parameter can be considered as an ideal probe to study the various atomic or molecular processes occurring in an optical system. Optical gain spectroscopy, a branch of laser spectroscopy deals with the measurement of optical gain through various spectroscopic techniques. Some of the well established methods for optical gain measurements are (i) pump-probe method, (ii) two channel technique and (iii) amplified spontaneous emission (ASE) technique.
2.1 Generalities

When a parallel beam of monochromatic light, of intensity \( I_p \), is incident on the surface of an optical material having a length \( X \), a part of the incident beam emerges out. To a large extent the intensity of the emergent beam depends on the nature of the impurities present in the crystal matrix. It also depends on the crystal matrix as a whole. Let \( I(l) \) be the intensity of the emergent beam. Now the ratio of \( I(l) \) to \( I_p \), gives the optical gain \( G \) of the crystal. The interaction of the incident radiation with the crystal can be expressed by the equation\(^1\)

\[
I(l) = I_p \exp(\alpha \rho X) \tag{2.1}
\]

where \((\alpha \rho)\) represents the effective gain coefficient while \( \alpha \) and \( \rho \) the gain coefficient and re-absorption coefficient respectively. For a high-gain media \( \rho \) is assumed to be very small, i.e., \( \rho \approx 0 \) and hence the optical gain equation can be written as

\[
G = \frac{I(l)}{I_p} = \exp(\alpha X) \tag{2.2}
\]

For a homogeneous isotropic medium, in the absence of re-absorption, the gain coefficient \( \alpha \) is defined as

\[
\alpha = \frac{1}{I(l)} \frac{dI(l)}{dX} \tag{2.3}
\]

that is, the percentage increase of intensity per unit length of the active medium.

The parameters \( \alpha \) can be expressed in terms of the ratio between photon input rate for unit length and output photon rate from unit length. Considering both the absorption and emission in resonance, the process can be treated either as a co-operative process or as a stimulated emission process (Einstein's treatment).
When considering it as a co-operative process, \( \alpha \) can be written as

\[
\alpha = \frac{1/(\tau_R X)}{1/T_2}
\]  

(2.4)

where \( T_2 = g(0) \) is the decay time of the non-diagonal elements of the density matrix, \( g(\nu-\nu_0) \) being the normalised line shape function (\( \nu \) is the frequency and \( \nu_0 \) the resonance frequency) and \( \tau_R \), the collective decay time, is given by

\[
\tau_R = \frac{8 \pi n^2}{\Delta N n^2 X} \tau_{sp}
\]  

(2.5)

where \( \tau_{sp} \) represents the spontaneous emission decay time, \( \Delta N = (N_2 - N_1) \) the population inversion, \( n \) the refractive index and \( \lambda \) the wavelength. Combining equations (2.4) and (2.5) we get

\[
\alpha = \frac{\lambda^2 \Delta N g(0)}{8\pi n^2 \tau_{sp}}
\]  

(2.6)

In Einstein's procedure we can write

\[
\alpha = \frac{\gamma^* \Delta N/X}{1/T_d}
\]  

(2.7)

where \( \gamma^* \) represents the stimulated emission coefficient, \( T_d = X/c \) the photon decay time in the material and \( c \) the speed of light.

The gain coefficient can be rewritten in terms of the spontaneous emission coefficient \( \gamma \) given by the equation

\[
\gamma = \frac{\gamma^* 8 \pi \nu^2}{g(0)c^3}
\]  

(2.8)

By replacing \( \gamma^* \) in equation (2.7) by means of equation (2.8) and putting \( \gamma = 1/\tau_{sp} \), we get the same expression (2.6) for \( \alpha \).
The normalised line shape at resonance \( g(0) \) is related to \( \Delta v \) (HWHM) in a different way depending on the line shape. For a Gaussian profile \( g(0) = 1/(1.064 \Delta v) \), while for a Lorentzian \( g(0) = 1/(1.571 \Delta v) \).

The presence of non-radiative de-excitations with time constant \( \tau_{NR} \) causes the effective emission decay time \( \tau_{EFF} \) to be shorter than \( \tau_{sp} \) according to the relation

\[
\frac{1}{\tau_{EFF}} = \frac{1}{\tau_{sp}} + \frac{1}{\tau_{NR}}
\]  

(2.9)

The quantum yield \( (Q) \) of the system is given by

\[
Q = \frac{\tau_{EFF}}{\tau_{sp}}
\]  

(2.10)

Thus for a Gaussian profile

\[
\alpha = \frac{\lambda^2 Q \Delta N}{8\pi \ 1.064 \ n^2 \tau_{EFF} \ \Delta v}
\]  

(2.11)

while for a Lorentzian profile

\[
\alpha = \frac{\lambda^2 Q \Delta N}{8\pi \ 1.521 \ n^2 \tau_{EFF} \ \Delta v}
\]  

(2.12)

Furthermore, the emission cross-section \( \sigma_e \) is defined as

\[
\sigma_e = \frac{\alpha}{\Delta N}
\]  

(2.13)

which depends only on the luminescence characteristics of the material.

2.2. Mathematical treatment of the optical gain coefficient

The intensity \( I(l) \) emitted by the pumped region, for a given wavelength, can be expressed as

\[
I(l) = K \int_0^l \frac{\exp(\alpha x)}{(P+x)^2} \ dx
\]  

(2.14)
where $K$ is a constant for a given pumping intensity and $I, P, x$ are defined in Figure 2.1.

![Diagram of optical gain measurements](image)

**Figure 2.1.** Schematic representation of optical gain measurements. $I_1$, incident beam; $I_2$, emergent beam; C, optical material; D, detector.

For $I << P$ or $x << P$, equation (2.14) can be written as

$$I(l) = K \int_0^l \frac{\exp(\alpha x)}{P^2} \, dx$$

i.e.,

$$I(l) = \frac{K}{\alpha P^2} [\exp(\alpha l) - 1]$$

Equation (2.15) implies that $I(l)$ is a constant only if $\alpha$ is a constant, that is independent of $x$. Since $\alpha = \alpha_0 N$, where
N is population of the excited centres and $\alpha_0$ is a coefficient depending on the luminescent properties of the material, the independence of $\alpha$ of $x$ requires N independent of $x$, or a uniform pumping intensity. In practice, the pumping laser beam is not perfectly homogeneous thus influencing the behaviour of $I(l)$. This variation of $I(l)$ affects the value of $\alpha$ and hence it is found to be a function of $x$. This dependence of $\alpha$ on $x$ can be written (for moderate inhomogeneities) as 

$$\alpha(x) = \alpha_0 N(x) = \alpha_0 N_0 \left(1 + A x + B x^2 + C x^3\right)$$

where the higher order terms in $x$ are small with respect to unity and $A$, $B$, $C$ are constants which are to be determined from the experimental analysis. Thus the exponential term in equation (2.14) can be written as

$$e^{\alpha x} = e^{\alpha_0 N_0 x} \left(1 + A x + B x^2 + C x^3\right)$$

$$= e^{\alpha_0 N_0 x} e^{\alpha_0 N_0 x^2 A} e^{\alpha_0 N_0 x^3 B} e^{\alpha_0 N_0 x^4 C}$$

(2.16)

Let $\alpha_0 N_0 A = \alpha_1$, $\alpha_0 N_0 B = \alpha_2$, $\alpha_0 N_0 C = \alpha_3$ and

$$P(x) = e^{\alpha_1 x^2} e^{\alpha_2 x^3} e^{\alpha_3 x^4}$$

(2.17)

Substitution of equation (2.17) in equation (2.16) gives

$$e^{\alpha_0 N_0 x} P(x)$$

(2.18)

On expanding each terms of $P(x)$ in terms of the corresponding exponential series we get

$$P(x) = [1 + \alpha_1 x^2 + (1/2!) \alpha_1 x^4 + \ldots] [1 + \alpha_2 x^3 + (1/2!) \alpha_2 x^6 + \ldots]$$

$$[1 + \alpha_3 x^4 + (1/2!) \alpha_3 x^8 + \ldots]$$

$$= 1 + \alpha_1 x^2 + \alpha_2 x^3 + \alpha_3 x^4 + \frac{1}{2} \alpha_1 x^4 + \alpha_1 \alpha_2 x^5 + \alpha_1 \alpha_3 x^6 + \ldots$$

i.e., $P(x) \approx 1 + \alpha_1 x^2 + \alpha_2 x^3 + \alpha_3 x^4$

(2.19)

$$I(l, N_0) = \frac{K}{P^2} \int_0^l e^{\alpha_0 N_0 x} P(x) \, dx$$

$$= \frac{K}{P^2} \int_0^l e^{\alpha_0 N_0 x} P(x) \, dx$$
\[ l(l,N_0) = \frac{K}{P^2} \int_0^1 e^{\alpha_0 N_0 x} (1 + \alpha_1 x^2 + \alpha_2 x^3 + \alpha_3 x^4) \, dx \]

\[
= \frac{K}{P^2} \frac{1}{\alpha_0 N_0} \left[ e^{\alpha_0 N_0 x} \left( \frac{P'(x)}{\alpha_0 N_0} + \frac{P''(x)}{(\alpha_0 N_0)^2} + \frac{P'''(x)}{(\alpha_0 N_0)^3} + \frac{P^{IV}(x)}{(\alpha_0 N_0)^4} \right) \right]_0^1
\]

where

\[ P'(x) = 2\alpha_1 x + 3\alpha_2 x^2 + 3\alpha_2 x^2 \]

\[ P''(x) = 2\alpha_1 + 6\alpha_2 x + 12\alpha_3 x^2 \]

\[ P'''(x) = 6\alpha_2 + 24\alpha_3 x \]

\[ P^{IV}(x) = 24\alpha_3 \]

(2.20)

The calculation of gain depends on the determination of \((\alpha_0 N_0)\) from equation (2.20) which is rather cumbersome. An analytical way of calculating \((\alpha_0 N_0)\) is to obtain the ratio \(R = l(l,N_2)/l(l,N_1)\) where \(l(l,N_2)\) and \(l(l,N_1)\) are the intensities corresponding to two different values of the pumping.

From equation (2.15) it can be seen that the output intensity \((l_o)\) varies exponentially with the excitation length \((l)\).

On differentiating equation (2.15) with respect to \(l\) we get

\[
\frac{dl_o}{dl} = \frac{K}{P^2} \exp(\alpha l)
\]

i.e., \(\ln [dl_o/dl] = \ln (K/P^2) + \alpha l\)

This represents a straight line equation with slope \(\alpha\) and \(y\) intercept \(\ln (K/P^2)\). Thus by plotting a graph between \(\ln(dl/dl)\) and the excitation length \((l)\) and by measuring its slope \((\alpha)\), and the gain \((G)\), of the material can be calculated.
2.3 Special cases

2.3.1 Large intensity coefficient gain ($\alpha_0 N_0 l \gg 1$)

If the variation of the beam inhomogeneity is limited, but not negligible, and $\alpha_0 N_0$ is sufficiently high, then equation (2.18) turns out to be the product of a strongly varying function $\exp(\alpha_0 N_0 l)$ times a moderate varying function $P(x)$, so that we can neglect the derivatives of $P(x)$ in equation (2.20). Then equation (2.20) turns out to be

$$I(l, N_0) = \frac{K}{p^2 \alpha_0 N_0} \left[ \exp(\alpha_0 N_0 l) P(l_1, N_0) - 1 \right]$$  \hspace{1cm} (2.21)

$$\approx \frac{K}{p^2 \alpha_0 N_0} \exp(\alpha_0 N_0 l) P(l_1, N_0)$$

Also

$$R(l) = \frac{l_2(l_1 N_2)}{l_1(l_1 N_1)} = \frac{K_2/K_1}{N_1/N_2} \exp[\alpha_0(N_2-N_1)l]$$  \hspace{1cm} (2.22)

Since $K \propto N$

$$R(l) = \exp[\alpha_0(N_2-N_1)l]$$  \hspace{1cm} (2.23)

Thus by measuring $R(l)$, $\alpha_0$ can be calculated.

2.3.2 Small intensity coefficient gain ($\alpha_0 N_0 l \ll 1$)

On substituting the values of $P(x)$, $P^T(x)$, $P^T(x)$, $P^{III}(x)$ and $P^{IV}(x)$ in equation (2.20) and simplifying we obtain

$$I(l, N_0) = \frac{Ke^{\alpha_0 N_0 l}}{p^2 \alpha_0 N_0} \left[ 1 + \frac{2A}{(\alpha_0 N_0)} - \frac{6B}{(\alpha_0 N_0)^2} + \frac{24C}{(\alpha_0 N_0)^3} \right]$$
\[
+ \left[ -2A + \frac{6B}{(\alpha_0 N_0)} - \frac{24C}{(\alpha_0 N_0)^2} \right] + \left[ A - 3B + \frac{12C}{(\alpha_0 N_0)^2} \right] + (B - 4C) l^3 + c l^4
\]

\[
- \frac{K}{P^2 \alpha_0 N_0} \left[ 1 + \frac{2A}{\alpha_0 N_0} - \frac{6B}{(\alpha_0 N_0)^2} + \frac{24C}{(\alpha_0 N_0)^3} \right]
\]

(2.24)

Since \( \alpha_0 N_0 l \ll 1 \), we can simplify equation (2.24) by considering that the terms multiplying \( l, l^2, l^3 \) and \( l^4 \) are negligible. Thus we obtain

\[
I(l N_0) = \frac{K e^{\alpha_0 N_0 l}}{P^2 \alpha_0 N_0} \left[ 1 + \frac{2A}{\alpha_0 N_0} - \frac{6B}{(\alpha_0 N_0)^2} + \frac{24C}{(\alpha_0 N_0)^3} \right] (e^{\alpha_0 N_0 l} - 1)
\]

(2.25)

\[
R(l) = \frac{l_2(l N_2)}{l_1(l N_1)} = \frac{(e^{\alpha_0 N_2 l} - 1)}{(e^{\alpha_0 N_1 l} - 1)} \quad (2.26)
\]

where \( P(N_2, N_1) = \frac{1 + 2A(\alpha_0 N_2) - 6B/(\alpha_0 N_2)^2 + 24C/(\alpha_0 N_2)^3}{1 + 2A(\alpha_0 N_1) - 6B/(\alpha_0 N_1)^2 + 24C/(\alpha_0 N_1)^3} \)

The values of \( N_0 \) corresponding to the measurements, besides satisfying the condition \( \alpha_0 N_2 l, \alpha_0 N_1 l \ll 1 \), have to be sufficiently different so as to make the ratio \( R(l) \) significantly dependent on the intensity coefficient \( g \). In this condition an accurate description of equation (2.26) can be achieved by expanding the numerator up to the third order and the denominator up to the second order. By retaining in the ratio \( R(l) \) terms up to the second order in \( l \) we get

\[
R(l) = N_0/N_1 \left[ 1 + \frac{1}{2} \alpha_0 l (N_2 - N_1) + \alpha_0^2 l^2 \left( N_2^2/6 - (N_1 N_2)/4 \right) \right] \quad (2.27)
\]
From the experimental data of \( R(l) \) we can determine, by a best fitting procedure, the parameters \( N_2/N_1, P(N_2N_1) \) and \( \alpha_0N_2 \). Hence the gain \( G(l) = \exp(\alpha_0N_2l) \) can be calculated as a function of length \( l \).

### 2.3.3 Intermediate case \((\alpha_0N_0l = 1)\)

In this case equation (2.9) can be used for finding the intensity \( I(l,N_0) \) of the emergent beam.

### 2.4 Experimental techniques

Some of the common experimental techniques for optical gain measurements are (i) the pump-probe method, (ii) two channel technique, and (iii) the amplified spontaneous emission (ASE) technique which consists of (a) L-L/2 method, and (b) movable screen technique.

#### 2.4.1 The pump-probe method

The basic principle of this method\(^5\) is that, while the system is pumped by an electromagnetic wave of some frequency (pump beam), a second beam at another frequency (probe beam) propagates through the pumped area. Due to stimulated emission the output from the system at the probe beam frequency can be amplified, with respect to the input.

In the experimental arrangement the probe beam must be sufficiently weak in order not to vary significantly the populations of the system to prevent saturation. In this condition, the ratio of the input intensities, with and without pumping, is a direct measurement of the optical amplification. For measurements of the optical gain by this method two arrangements can be adopted: (i) transverse configuration (Figure 2.2a) where the probe beam propagates near the surface orthogonally to the pumping beam, and (ii) longitudinal configuration (Figure 2.2b), where the probe beam propagates in the same direction as the pumping beam.\(^6\,7\)
Figure 2.2. Optical gain measurement using pump-probe method
(a) Transverse configuration:
P$_1$, pump beam; P$_2$, probe beam; S, sample.
(b) Longitudinal configuration:
P$_1$, pump beam; P$_2$, probe beam; L, focusing lens; S, sample;
BS, beam splitter; D, detector; O, oscilloscope; MR, monochromator;
M, mirror.
In principle, under pumping, the optical gain for a single passage is given by the ratio between the intensities of the output signal \( I(I) \) and the probe intensity \( I_0 \).

The disadvantage of this method is that in the UV region this is not applicable due to the scarcity of tunable lasers in this region.

### 2.4.2 Two channel technique

This method\(^8\) is based on the comparison between the luminescence intensity \( I_F \) and a reference signal proportional to the pump beam intensity \( I_P \). Figure 2.3 shows the experimental setup of optical gain measurement by two channel technique. A continuous recording of \( I_F \) versus \( I_P \) is achieved by varying the pump intensity by means of a variable slit in such a way that the inhomogeneity of the pumping beam does not affect the measurement.

The intensity coefficient \( \alpha_0 N_0 \) can be obtained by means of the fitting of \( I_F \) expressed by the equation

\[
I(N_0) = K(\alpha_0 N_0 P^2) \left[ \exp(\alpha_0 N_0) - 1 \right]
\]

Knowing \( \alpha_0 N_0 \) and the length of the crystal \( L \), the gain of the crystal can be calculated. The main disadvantage of this method is the difficulty of maintaining the linearity of the system. It is found that a high power density is required for the incident beam to maintain the linearity of the system. But this may damage to the material surface. For less power densities, which do not damage the material, the deviation from the linearity is too small to be detectable, and hence the resulting gain data are not reliable.
Figure 2.3. Experimental setup of two channel technique.
1, crystal; 2, spherical lens; 3, beam splitter; 4, variable slit;
5, laser beam; 6, to fluorescence detector; 7, to reference detector.
2.4.3 Amplified spontaneous emission (ASE) techniques

Amplified spontaneous emission occurs when the spontaneous emission from a distribution of inverted atoms is amplified by stimulated emission without the help of a resonant cavity to provide feed back.

As an alternative to the pump-probe method optical gain can be obtained from the determination of the stimulated emission under the influence of the system's own fluorescence. The analysis of ASE offers an indirect but simpler method because it requires only the pump beam.

The ASE technique involves a comparison of the emission from different lengths of the pumped region of the sample under test. Commonly the comparison is made for discrete values of the length of the activated medium or, in improved methods, the emission is measured as a continuous function of the pumped region length. Both these techniques are commonly known as movable screen techniques.\(^5\)

a) \textit{L-L/2 method}

In this technique the length of the gain region may be varied from L to L/2 by blocking off one-half of the pumping beam by a shutter.

Figure 2.4 gives a sketch of the measurement apparatus. The emission, through a pinhole, enters a monochromator. With reasonable assumptions on the pumping stability and the geometry and on the hypothesis that the gain is small enough so that the ASE does not saturate the transition, by simply measuring the emission from a full and a half length, the gain coefficient can be computed from the ratio of the resultant ASE signals \(I_L\) and \(I_{L/2}\) and is given by\(^4\)

\[
\alpha = \frac{2}{L} \ln \left( \frac{I_L}{I_{L/2}} - 1 \right) \tag{2.28}
\]
b) Movable screen technique

While in the previous methods the light intensities considered correspond to two fixed lengths of the pumped region, another technique has been developed in which the emission is measured as a function of the pumped length.

The optical setup of this technique is given in Figure 2.5. The emission from the sample is continuously recorded while a screen moves slowly perpendicular to the pumping beam. Further, the use of two different pump intensities in alternate sequence permits compensation of the inhomogeneities of the pumping beam, also minimising the effect of pump intensity drifts, radio-frequency interferences and efficiency loss of the active medium. From a best fit of the ratio of the intensities (at full and reduced pumping) the optical gain can be derived. The main advantage of this method is the increased accuracy in the optical gain evaluation by means of a best fit of the whole experimental curve of the emission intensity as a function of the pumped length. This is particularly useful when materials with small values of optical amplification are investigated. The detailed experimental arrangements for optical gain measurement by the above described method is given in Figure 2.6.
Figure 2.5. Optical set-up (schematic) for ASE gain measurement by movable screen technique.
1. laser beam, 2. cylindrical lens; 3. movable screen; 4. crystal.
Figure 2.6. Detailed experimental arrangement for optical gain measurement by movable screen technique.
BS, beam splitter; MS, movable screen.

In conclusion, even if ASE techniques have less sensitivity compared to the pump-probe method, they have the advantage of employing a simpler setup with only one pump laser and an easier experimental procedure.
References


Chapter Three

Optical Amplification In CdI$_2$ and CsI:In(Tl)
Ionic Crystals

Ever since the discovery of optical amplification a lot of efforts have been done by scientists to reveal this phenomenon in a variety of optical materials. Since optical amplification is a fundamental process behind laser action a lot of theoretical as well as experimental investigations have been carried out in this field over the past few decades. The results of all these investigations helped in the design and development of a lot of laser materials. Nowadays, a lot of materials are found to exhibit laser action in the solid, liquid and gaseous state. Solid state laser materials can be either crystalline or amorphous in nature. An important condition for laser action in any material is the existence of a number of states whose mean lifetimes are relatively long so that it will be possible to pile up a considerable amount of energy in the excited state. Higher values of cross section, long lifetime of the levels and the sharpness of the spectral line are the characteristic features of a good optical (laser) material. However, on the other hand, optical materials having short lived excited states are found to be good laser materials because of the fact that the stored energy should be released in the shortest possible interval realising high optical gain. Among the crystalline laser materials, rare earth doped crystals, colour centre laser crystals and semiconductors are of special interest. This chapter deals with the spectroscopic investigations on the optical amplification of