CHAPTER III

STUDY OF FOUR-WAVE MIXING PROCESSES

1. INTRODUCTION

In this chapter, the nonlinear optical process of four-wave mixing is studied both theoretically and experimentally.

1.1. Nonlinear phenomena

Nonlinear optics [1-3] deals with phenomena in which the electric polarization of a medium depends on second and higher powers of the electric field strength incident on the medium. Rich variety of nonlinear optical processes have been observed, such as harmonic generation, optical mixing including sum and difference frequency generation, parametric oscillation, multiple photon absorption, Raman scattering, self-focussing and photoelastic effect. These phenomena are all nonlinear as they all arise from a modulation of the optical susceptibility of the medium by the electromagnetic wave. There is an additional interesting feature that the rate of the nonlinear optical processes in general depends upon the degree of coherence or the photon statistics of the participating beams. The nonlinear rates thus change with time as the nonlinear process itself changes the photon statistics of the light beams, leading to temporal variation of the beam intensities which is
sensitive to the initial statistics. Some of the nonlinear processes are capable of producing squeezed light of the kind discussed in the following Chapter.

Consider an electromagnetic field $E$ incident on a non-magnetic, non-conducting medium. For incident fields of very low strengths, the induced electric polarization is given as $P = \varepsilon_0 \chi E$, where $\varepsilon_0$ is the dielectric permittivity of vacuum and $\chi$ is the dielectric susceptibility of the medium. For sufficiently strong fields, the susceptibility $\chi$ is field dependent and can be written as a power series expansion in $E$. Under weak and nonresonant excitation of the medium, the induced polarization of the medium is given by

$$P_i = \varepsilon_0 \left[ \chi^{(1)} E_j + \chi^{(2)}_{jk} E_j E_k + \chi^{(3)}_{jk} E_j E_k E_l + \ldots \right].$$  \(\text{III.1.1}\)

where $\chi^{(n)}$ is the nth-order susceptibility tensor (of rank $n+1$ with $3^{n+1}$ components) characteristic of the medium. Symmetry considerations reduce the number of nonzero and distinct components of the susceptibility tensor. For media with inversion symmetry, $\chi^{(2)}$ is zero and hence second- (and all even-) order effects are not seen. Details of various nonlinear phenomena can be found in several books on nonlinear optics [1-3].

The generation of new waves in a nonlinear optical interaction can be interpreted intuitively in terms of a grating picture. When the light waves interfere inside a medium, a spatially periodic pattern of excitation is created. Such a pattern of medium excitation is called a laser-induced grating (LIG) [4]. These dynamic gratings are created only in the presence of two or more incident beams. In terms of macroscopic parameters, the LIG can be viewed as a modulation in the refractive index and absorption coefficient of the
medium. The former is called a *phase grating* and later an *amplitude grating*. In general, both types of gratings coexist in a medium. A light wave incident on the medium will be scattered by the LIG in specific directions in a manner similar to the diffraction from an ordinary static grating. In addition, the LIG can scatter the light beams that created the grating in the first place. This process is known as *self-diffraction*.

Nonlinear mechanisms can be classified into two types: resonant and nonresonant. The nonresonant nonlinearities may occur as a result of the nonlinear response of bound electrons to an incident electric field. In this case the size of nonlinearity is usually not very large. Large nonresonant nonlinearities due to orientation of molecules may occur in liquids comprising of anisotropic molecules. The tendency of the anisotropic molecules to get oriented along the incident electric field direction leads to the orientational nonlinearity. Light induced distortions of molecular, electronic and nuclear moments also lead to changes in the optical properties. These nonlinearities can be seen in some liquids, such as CS$_2$. The incident electric field can also cause stress changes in the medium leading to the changes in refractive index. The nonlinearities due to this ‘electrostriction’ are present in a medium composed of aerosols or dielectric spheres.

The resonant mechanisms, on the other hand, are relatively slow as they involve the actual redistribution of populations of the associated levels but are large in strength. By tuning the incident frequency around the material response, one can selectively enhance the susceptibility. In fact the largest observed nonlinearities are based on resonant mechanisms. Saturation of optical transitions in various materials like dyes and semiconductors falls
under this category. In the experimental section of this chapter, we employ
the resonant nonlinearities due to saturation in organic dyes.

1.2. Four-wave mixing

Four-wave mixing refers to the nonlinear process with four interacting
electromagnetic waves. In the weak interaction limit, it is a third-order
process and is governed by the third-order nonlinear susceptibility $\chi^{(3)}$. As
the susceptibility tensor is invariant under reflection, only 21 components of
$\chi^{(3)}$ (out of a possible $3^4 = 81$) will be nonzero. For an isotropic medium with
rotational symmetry, $\chi^{(3)}$ has only four nonzero components, $\chi_{xxyy}^{(3)}$, $\chi_{xyyx}^{(3)}$, $\chi_{xyyx}^{(3)}$, and $\chi_{xxyz}^{(3)}$, out of which only three are independent as $\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)} + \chi_{xyyx}^{(3)} = \chi_{xxyz}^{(3)}$. The typical susceptibility element $\chi_{xyxy}^{(3)}$ couples the three
fields which are along $y$, $x$ and $y$ directions to generate a polarization which
radiates along the $x$-direction. Unlike second-order processes, a third-order
process is allowed in all media, with or without inversion symmetry. Being
flexible and easily observable in all media, four-wave mixing has many
interesting applications. In the degenerate case (i.e., four waves having the
same frequency) it is used for wavefront reconstruction. In a resonant medium,
it can be adopted as a powerful spectroscopic and analytical tool for material
studies.

In media with inversion symmetry, third-order nonlinearity is the lowest
order nonlinearity allowed under the electric-dipole approximation. The micro-
scopic expression of a third-order nonlinear susceptibility is derived from a
perturbation calculation and is given in ref. [2]. For simplicity, let us
consider three fields $E_m(\omega_m) = \xi_m \exp(i k_m \cdot r - i \omega_m t)$ with $m = 1, 2, 3$, incident
on an isotropic medium. The output field, \( E_s(\omega_s) = \varepsilon_s \exp(\textbf{i}k_e \cdot \textbf{r} - i\omega_s t) \) with \( \omega_s = \omega_1 + \omega_2 + \omega_3 \), is governed by the wave equation

\[
\left[ \nabla^2 + \mu_0 \omega_s^2 \varepsilon(\omega_s) \right] E_s = -\mu_0 \omega_s^2 P(3)(\omega_s), \tag{III.1.2}
\]

where \( \varepsilon(\omega) = \varepsilon_0 \left( 1 + \chi^{(1)} \right) \), \( P(3)(\omega_s) = \varepsilon_0 \chi^{(3)}_{1j} \chi^{(3)}_{j1} (\omega_s = \omega_1 + \omega_2 + \omega_3) E_j(\omega_1) \times E_k(\omega_2) E_l(\omega_3) \). With the slowly-varying amplitude approximation [2], negligible pump depletion, and assuming the nonlinear medium to be semi-infinite with a plane boundary surface, the solution of (III.1.2) is [2]

\[
\varepsilon_{s1}(z) = -\frac{\mu_0 \omega_s^2}{2(\Delta k \cdot \hat{z}) k_s} \chi^{(3)}_{1j} \chi^{(3)}_{j1} \varepsilon_{2k} \varepsilon_{31} (1 - e^{i\Delta k \cdot z}) e^{-\alpha_s z}. \tag{III.1.3}
\]

where \( \Delta k = \Delta k' + i\Delta k'' = (k'_1 + k'_2 + k'_3 - k'_s) + i\hat{z} (\alpha_{1j} + \alpha_{2k} + \alpha_{31} - \alpha_{s1}) \), \( k'_i \) is the real part of \( k_i \), \( \Delta k' \) is the wavevector mismatch, and \( \alpha \)'s are the absorption coefficients of the waves along \( \hat{z} \).

For a nonlinear interaction to take place with appreciable probability, phase matching (energy and momentum conservation) is required. In four-wave mixing, phase matching \((\Delta k' = 0)\) may be achieved in an infinite number of ways by properly adjusting the directions of propagation of the three pump waves, and it greatly enhances the signal output whose amplitude is given by Eq. (III.1.3).

We now consider the special case of degenerate four-wave mixing (DFWM) in which all the four waves have the same frequency \( \omega \). In this case \( \chi^{(3)} \) has at least a singly-resonant term arising from the two-photon zero-frequency
resonance, i.e., a term with \((\omega - \omega + i/T_1)\) in the denominator where \(T_1\) is the longitudinal relaxation (population decay) time of the medium. It may also have a two-photon resonant term if \(\omega + \omega\) is in resonance with a transition of the medium. This corresponds to a two-photon absorption process in which two photons are simultaneously absorbed to excite a material medium. Finally \(\chi^{(3)}\) can be triply resonant if \(\omega\) is very near a resonance. Because of the strong resonant enhancement, \(\chi^{(3)}\) for degenerate four-wave mixing can be very large in some media. In our work in this thesis, we consider such a resonant third-order process which is observable with low-power CW laser beams.

1.3. Optical phase conjugation

Phase conjugation is defined as the process in which the phase of the output wave is complex conjugate to the phase of an input wave. In other words, the process reverses the phase of an input wave. The phase conjugate wave can be generated in a number of ways and the backward DFWM is a method often used. It involves the interaction of three coherent waves of the same frequency in a nonlinear medium to generate a fourth wave which is phase conjugate to one of the input waves [5]. The forward wave \(f\) and the backward pump wave \(b\) propagate opposite to each other. The probe wave \(p\) is incident at an angle (usually less than 45°) to \(f\). The three inputs stimulate a third-order nonlinear polarization, which radiates an output wave \(s\) that is the phase conjugate of the probe wave \(p\). The conjugate wave \(s\) propagates exactly opposite of \(p\) and retraces its wavefronts (see Fig. III.3 later).

In terms of the grating picture, the spatial interference of \(f\) and \(p\) waves gives rise to a medium excitation grating that is phase-matched to
scatter the b wave along s, counterpropagating to p. Similarly, the b and p waves form another grating that scatters the f wave along the direction of s. There are other diffraction processes taking place in the DFWM geometry but they do not lead to the conjugate of the probe wave.

2. FOUR-WAVE MIXING WITH CHAOTIC FIELDS

Various line broadening processes in a source of light (discussed in Chapter II) cause the electric field of the beam to fluctuate around its mean value on time scales that are inversely proportional to the frequency bandwidth of the light. In a gas discharge lamp, different atoms are excited by an electrical discharge and emit radiation independently of one another. The shape of an emission line is determined by the statistical spread in atomic velocities and random occurrence of collisions. Such a light source produces chaotic light. Thermal cavity, filament lamp, and multimode laser are some other examples of a chaotic light source. The light beams from any variety of chaotic light sources have similar statistical distributions; only parameters of the statistical distribution vary from one chaotic light beam to another.

Recently, there has been considerable interest in the study of atoms interacting with chaotic laser fields. It has been recognized that the incoherence and in general partial coherence of the fields incident on a nonlinear medium can lead to important spectroscopic applications. For example, Morita and Yaiima [6] proposed that extremely fast dephasing phenomena can be studied by four-wave mixing with incoherent broadband light. These authors established that the shortest dephasing times that can be
measured with incoherent light are limited by the correlation time of the fluctuating field. Since it is much simpler to produce incoherent light with a short correlation time than to produce ultrashort pulses, this technique is indeed a powerful one. They observed that "even a CW light, which has infinite duration, enables us to observe the subpicosecond or femtosecond relaxation process in the time domain if only it has an adequate spectral width". Morita and Yajima dealt with weak, chaotic, broadband fields, i.e., δ-function correlated (Markovian) fluctuations, and derived expressions for the FWM signal from perturbative solution of optical Bloch equations. Related work in this context is that of Agarwal [7] on weak, chaotic fields with finite bandwidths using the third-order perturbation theory. For strong fields, solution exists when excitation is provided by uncorrelated, broadband light pulses [8]. This situation can be illustrated by a stimulated photon-echo experiment [9] in which three different lasers are used to produce the three broadband excitation pulses. On the other hand, when two strongly correlated pulses derived from same source interfere inside a medium, they create a spatial grating in the atomic inversion (difference in the atomic level population). This grating can be probed by a third pulse which is generally weak, uncorrelated with the first two, and is delayed in time so that it does not overlap with them. A number of papers [10-12] have been devoted to this subject. In these calculations, the response of an atomic ensemble subjected to two time-delayed pulses has been studied as a function of noise properties of the fields. In such a system, the atoms retain some memory of the first field when the second field acts. Even if the noise of each field is Markovian, the combined effect of two fields is non-Markovian in general owing to memory effects.
Vemuri et al. [13] presented Monte Carlo results for four-wave mixing with time-delayed CW chaotic fields of arbitrary bandwidths. Subsequently, these calculations were extended for phase-diffusing fields for which the amplitude of the field is a constant but the phase or frequency is a stochastic quantity [14]. Agarwal et al. [15] did a comparative study of chaotic and phase-diffusing fields. In all these papers [13-15], the authors considered the "probe" to be one of the two correlated pump beams which create the dynamical grating, and dealt with the case when "probe" is weak and calculated the four-wave mixing signal to first order in the weak "probe" field and all orders in the other pump field. In most of the experiments [10,16], however, the grating is created by two equally strong pump beams. These beams themselves get diffracted from the grating (self-diffraction), and sometimes a third uncorrelated weak beam is used to probe the grating.

We consider the self-diffraction geometry in which the two chaotic pump beams which create the grating in a resonant material can be arbitrarily strong. These pump beams in turn get diffracted from the grating and generate output light in new directions. In Section 2.1, we present the basic equations for nonlinear mixing with chaotic pumps. The calculation of the total output light reduces to the numerical solution of three coupled Langevin equations with multiplicative noise. The treatment of this section can easily be extended to three incident beams. In Section 2.2, we numerically simulate a chaotic field with arbitrary fluctuation parameters. The total output is obtained by Monte Carlo simulation methods. Here the two input beams are allowed to be arbitrarily strong, but the output lacks detailed information on signal components along specific directions. In Section 2.3, we outline a
method for calculating the component of the output that is solely due to four-wave mixing interactions. In Section 3, we report the experimental work in which a nonlinear signal is produced in a self-diffraction geometry and the diffracted beam is studied for different conditions of the pump beam [17]. The transient behavior of the signal when the pump beams are switched on and off is also reported. Finally in Section 4, we summarize the results.

2.1 Basic equations

We consider a degenerate four-wave mixing interaction in a forward geometry for which a light beam of frequency $\omega$ is spatially divided into two beams with wave vectors $k_1$ and $k_2$, and the split-beams are then mixed in a thin, resonant material. We treat the medium as an ensemble of stationary two-level atoms with no degeneracy, interacting with the field consisting of the superposition of two correlated beams. For simplicity, we take the path lengths travelled by the two pump beams to be equal so that there is no delay between them. Note that here, unlike in the phase-conjugate geometry, any effects due to a standing wave in the medium are negligible. The total field incident on the medium at position $r$ and at time $t$ can be written in the form

$$E(r,t) = \varepsilon_1(r,t)e^{i(k_1 \cdot r - \omega t)} + \varepsilon_2(r,t)e^{i(k_2 \cdot r - \omega t)} + \text{c. c.}$$  \hspace{1cm} (III.2.1)

where $\varepsilon_1(r,t)$ and $\varepsilon_2(r,t)$ are the electric field amplitudes of the two incident beams. Four-wave mixing signals at the same frequency $\omega$ are generated in new directions along $k_3 = 2k_2 - k_1$ and $k_4 = 2k_1 - k_2$ owing to first-order diffraction from the LIG, i.e., due to third-order nonlinearity of the
material (see Fig. III.1). In the case of chaotic fields, the stochastic envelope $\xi_i(\mathbf{r},t)$. $i = 1, 2$, is a Gaussian random process with the properties

$$
\langle \xi_1(\mathbf{r},t) \rangle = 0 .
$$

$$
\langle \xi_1(\mathbf{r},t)\xi_1^*(\mathbf{r},t') \rangle = |\xi_1(\mathbf{r},0)|^2 e^{-\Gamma|t-t'|}.
$$

$$
\langle \xi_1(\mathbf{r},t)\xi_1(\mathbf{r},t') \rangle = 0 = \langle \xi_1^*(\mathbf{r},t)\xi_1^*(\mathbf{r},t') \rangle .
$$

where $|\xi_1(\mathbf{r},0)|^2$ is the variance of the Gaussian random process and gives the incident intensity of the field. The chaotic field model as described by Eq. (III.2.2) has a Lorentzian spectral profile with a full width at half maximum (FWHM) of $2\Gamma$, and is an accurate representation of the field from a multimode laser. In the case when the two beams incident on the sample are derived from the same laser, the two incident beams are fully correlated and can be written as

$$
\xi_2(\mathbf{r},t) = g \xi_1(\mathbf{r},t) .
$$

In most experiments, $g \approx 1$. The time evolution of the atoms is conveniently expressed in terms of the density-matrix elements $\rho_{11}$, $\rho_{22}$, $\rho_{12}$, and $\rho_{21}$ that characterize the atoms. The relaxation processes in the medium are usually described by two kinds of phenomenological parameters, the longitudinal relaxation time $T_1$ and the transverse relaxation time $T_2$, representing the decay of the population difference and coherence between the two levels, respectively. The dynamical behavior of the two-level atoms interacting with the field (III.2.1) is governed by the optical Bloch equations. In the frame rotating with the frequency $\omega$ of the pump field, the Bloch equations for the
Fig. III.1 Schematic diagram showing self-diffraction geometry of degenerate four-wave mixing. The pump beams $k_1$ and $k_2$ and diffracted beams $k_3$ and $k_4$ are in the x-z plane. The resonant material surface at the point 0 where pump-beam centers coincide is taken to be in the x-y plane.
atomic dipole moments and the population inversion can be written as

\[
\frac{d\psi}{dt} = M \psi + I .
\]  

(III.2.4)

where

\[
\psi = \begin{bmatrix}
\rho_{21} e^{i(k_1 \cdot r - \omega t)} \\
\rho_{12} e^{-i(k_1 \cdot r - \omega t)} \\
(p_{11} - p_{22})/2
\end{bmatrix} .
\]  

(III.2.5)

\[
M = \begin{bmatrix}
-1/T_2 + i\Delta & 0 & -2i\Omega^*(r,t) \\
0 & -1/T_2 - i\Delta & 2i\Omega(r,t) \\
-i\Omega(r,t) & i\Omega^*(r,t) & -1/T_1
\end{bmatrix} .
\]  

(III.2.6)

\[
I_1 = I_2 = 0, \quad I_3 = -1/2T_1 .
\]  

(III.2.7)

\(\Delta\) is the atom-field detuning given by

\[
\Delta = \omega_0 - \omega .
\]  

(III.2.8)

\(\omega_0\) is the atomic transition frequency. \(\Omega\) corresponds to the Rabi frequency in the frame rotating with the field frequency \(\omega\) and is given by

\[
\Omega(r,t) = \frac{\mu \cdot \hat{e}}{\hbar} \left[ \xi_1(r,t) + \hat{g} \xi_1(r,t) \exp \left[ i(k_2 - k_1) \cdot r \right] \right] .
\]  

(III.2.9)

\(\mu\) is the electric dipole matrix element of the transition between the two levels, and \(\hat{e}\) is the unit polarization vector. The output light field in the
mixing process is proportional to the induced polarization in the medium. The output light intensity in the steady-state, to all orders in the pump fields, is thus proportional to $S$, which, for a homogeneously-broadened medium, is defined by

$$S = \lim_{t \to \infty} \left\langle \rho_1^* \rho_2 \right\rangle. \quad \text{(III.2.10)}$$

where $\left\langle \right\rangle$ denotes stochastic averaging over fluctuations of the pump beams. The stochastic averaging is done for chaotic input field fluctuations by the Monte Carlo technique.

2.2. Numerical study of nonlinear mixing signals

The inclusion of arbitrary pump intensities and bandwidths makes the problem of atoms interacting with chaotic field very difficult to solve analytically and one resorts to numerical methods to obtain the dynamical behavior of the atom. Using the Monte Carlo technique in the form developed by Fox et al. [18] to numerically integrate the Langevin (Bloch) equations (III.2.4) in which $\varOmega(r,t)$ represents a random process, one can calculate the FWM signals for a wide range of pump intensities and bandwidths. To carry out the Monte Carlo simulations, one first needs to produce the complex, stochastic electric field, a method for which is outlined here. As the electric field amplitude is chaotic in nature with the properties (III.2.2), $\varOmega(r,t)$ defined by (III.2.9) is taken to be an exponentially correlated (colored) Gaussian process, with the properties

$$\left\langle \varOmega(r,t) \right\rangle = 0. \quad \text{(III.2.11)}$$
\begin{equation}
\left< \Omega(r,t)\Omega^*(r,t') \right> = D\Gamma e^{-\Gamma|t-t'|} .
\tag{III.2.12}
\end{equation}

where \( \Gamma \) is the inverse of the correlation time (half-width at half-maximum of the spectral profile) as before (see III.2.2b), and the variance of \( \Omega(r,t) \) is

\begin{equation}
D\Gamma = \left( \frac{\mu e}{h} \right)^2 |\xi_1(r,0)|^2 \left[ 1 + 2g \cos \left( (k_2 - k_1) \cdot r \right) + g^2 \right].
\tag{III.2.13}
\end{equation}

The algorithm used by Vemuri et al. [13] to generate \( \Omega(r,t) \) is briefly reproduced here. The first step is to produce the complex, Gaussian \( \delta \)-correlated (white) noise \( g_w \), which is the source term for the colored noise. \( g_w \) has the well known properties

\begin{align*}
\left< g_w(t) \right> &= 0 , \tag{III.2.14a} \\
\left< g_w(t)g^*_w(t') \right> &= 2D\delta(t-t') . \tag{III.2.14b}
\end{align*}

which completely determine all the statistical properties. It is easily produced by the Box-Mueller algorithm,

\begin{equation}
g_w = \left[ -2D\Delta t \ln (a) \right]^{1/2} \exp(2\pi ib) . \tag{III.2.15}
\end{equation}

where \( a \) and \( b \) are computer generated, uniformly distributed random numbers between 0 and 1, and \( \Delta t \) is the integration step size [used in integrating (III.2.4)]. Exponentially correlated colored noise as described in Eq. (III.2.12) is obtained from the equation

\begin{equation}
d\Omega/dt = -\Gamma \Omega + \Gamma g_w . \tag{III.2.16}
\end{equation}
in which \( g_w \) is the Gaussian white noise as defined before. On integrating (III.2.16) we get

\[
\Omega(r, t + \Delta t) = \Omega(r, t) \exp(-\Gamma \Delta t) + h(t). \tag{III.2.17}
\]

where \( h \) is the source term producing colored noise. \( h \) depends on \( g_w \) and obeys Gaussian statistics, has a zero mean and a second moment given by

\[
\langle |h(t, \Delta t)|^2 \rangle = D \Gamma \left[ 1 - \exp(-2\Gamma \Delta t) \right]. \tag{III.2.18}
\]

Thus to generate the colored noise \( \Omega(r, t) \), \( h \) is first produced by the formula

\[
h = \left[ -D \Gamma \{1 - \exp(-2\Gamma \Delta t)\} \ln (\alpha) \right]^{1/2} \exp(2\pi ib). \tag{III.2.19}
\]

where, as before, \( \alpha \) and \( b \) are computer generated, uniformly distributed random numbers between 0 and 1. The exponentially correlated noise is then obtained from expression (III.2.17). For \( \delta \)-correlated fluctuations, one can use either the Box-Mueller algorithm to produce white noise, or the above noise algorithm in the limit \( \Gamma \gg D \).

The three Langevin equations given by (III.2.4) are of the form

\[
\dot{\psi}(t) = \nu(\psi(t)) + \omega(\psi(t)) X(r, t). \tag{III.2.20}
\]

where \( \nu(\psi(t)) \) and \( \omega(\psi(t)) \) are functions of \( \psi \), and \( X(r, t) \) is the colored-noise term with zero mean and a variance of \( D \Gamma \), which appears in the multiplicative
form. For numerical integration of this equation to first-order in the time-step $\Delta t$, one uses the following step:

$$\psi(t + \Delta t) = \psi(t) + \psi(t) \Delta t + \psi(t) \chi(t, t) \Delta t.$$ (III.2.21)

One solves the set of coupled Langevin equations (III.2.4) numerically, with initial conditions $\psi_1(t=0) = \psi_2(t=0) = 0$ and $\psi_3(t=0) = -0.5$. An Euler method is used for numerical integration. The accuracy of the results is dependent on the time-step size (which should be much smaller than all other relevant time scales in the problem) and the number of trajectories over which the stochastic averaging is performed, and hence appropriate choices for these parameters are made. All the time units are normalized to $T_2$. For the given set of parameters $\mu = T_2 / T_1$, $\Delta T_2$, $DT_2$ and $\Gamma T_2$, the computed value of $\psi_2$ is allowed to reach a steady state, and the output $S$ is obtained as in (III.2.10). The signal represents an averaging over 1000 trajectories, each with a different set of random numbers. This is to ensure that the results are not effected by small number statistics.

2.3. Calculation of four-wave mixing signals

Writing the solution of Eq. (III.2.4) to all orders in the pump fields as

$$\rho = F_1 e^{i k_1 \cdot r} + F_2 e^{i k_2 \cdot r} + F_3 e^{i k_3 \cdot r} + F_4 e^{i k_4 \cdot r} + \ldots.$$ (III.2.22)

one can see that the output light intensity along the direction of $k_1$ in the steady-state is proportional to $S(k_1)$ where

$$S(k_1) = \lim_{t \to \infty} \langle (F_{11}^*)^2 (F_{12}^*)^2 \rangle.$$ (III.2.23)
As before, $\langle \rangle$ denotes stochastic averaging over fluctuations of the pumps. The FWM signals are then proportional to $S(k_3)$ and $S(k_4)$.

Vemuri et al. [13] and Agarwal et al. [15] assumed one of the pump beams to be weak and they calculated the FWM signal $S(k_4)$ to first order in the weak field. The Block vector $\psi$ of (III.2.5) is expanded in powers of the weak field $\xi_2 = g\xi_1$, with $g \ll 1$ [13]:

$$\psi = \psi^{(0)} + \psi^{(1)} + \psi^{(2)} + \ldots \ldots \ . \quad \text{(III.2.24)}$$

where

$$\frac{d\psi^{(0)}}{dt} = M^{(0)} \psi^{(0)} + I \ , \quad \text{(III.2.25)}$$

$$\frac{d\psi^{(1)}}{dt} = M^{(0)} \psi^{(1)} + M^{(1)} \psi^{(0)} \ . \quad \text{(III.2.26)}$$

Here $M^{(0)} + M^{(1)} = M$ (given by (III.2.6)). $M^{(0)}$ is obtained from (III.2.6) by setting $g = 0$, i.e., for no probe, and $M^{(1)}$ is given by (III.2.6) with $1/T_2 = \Delta = 1/T_1 = 0$, and no pump, i.e., from (III.2.9), for $M^{(1)}$.

$$\Omega(r,t) = \frac{\mu \cdot \hat{e}}{\hbar} g \xi_1(r,t) \exp \left[ i(k_2 - k_4) \cdot r \right] . \quad \text{(III.2.27)}$$

$\psi^{(0)}$ has contribution from the beam along $k_1$, and $\psi^{(1)}$ has contributions from the beams along $k_2$ and $k_4$. So the solution of (III.2.26) for $\psi^{(1)}$ is written in the form

$$\psi^{(1)} = \left( F_2^{(1)} e^{ik_2 \cdot r} + F_4^{(1)} e^{ik_4 \cdot r} \right) e^{-ik_1 \cdot r} . \quad \text{(III.2.28)}$$
From (III.2.26) and (III.2.28), one gets

\[ \frac{dF_4^{(1)}}{dt} = M^{(0)} F_4^{(1)} + i \left( \frac{\mu \cdot \sigma}{\hbar} \right) \begin{pmatrix} -2 \psi_3^{(0)} \\ 0 \\ \psi_2^{(0)} \end{pmatrix} \]  

(III.2.29)

where \( \psi^{(0)} \) is the solution of (III.2.25). The steady state FWM signal in the direction \( k_4 \) is then obtained from

\[ S = \lim_{t \to \infty} \langle (F_4^{(1)})^* (F_4^{(1)}) \rangle_2. \]  

(III.2.30)

where the brackets refer to stochastic averaging with respect to the fluctuations of the field \( \xi_4 \). Vemuri et al. [13] solved six coupled Langevin equations (III.2.25) and (III.2.29) by numerical integration using Monte Carlo methods, as outlined in Section 2.2. In this method, however, one can not obtain a solution for the FWM signal to all orders in the incident fields. When the expansion (III.2.24) is not valid. Here, the signal along \( k_3 \) can be estimated by solving the Bloch equation for \( \psi^{(2)} \) which involves terms which are second-order in the weak field \( \xi_2 \). Hence, the basic symmetry of the FWM signals generated along \( k_3 \) and \( k_4 \) in a typical experimental configuration [10,16] with two equally strong pumps is not recoverable from these calculations. One should ideally proceed with a solution of the type (III.2.22) for such a fully nonperturbative treatment of the problem, and extract information on the signals generated along particular directions to all orders in the pump fields. Further work needs to be done along this direction.
3. EXPERIMENTAL STUDY OF FOUR-WAVE MIXING SIGNALS

In this section, we report the experimental work on the nonlinear signal generated in the degenerate four-wave mixing process in a resonant material in two different set-ups. We consider the self-diffraction geometry and study the behavior of the signal diffracted from the laser-induced grating for various pump parameters. The grating can be produced very easily in some organic dyes doped in a solid environment. In Section 3.1, we discuss the preparation of the Rhodamine 6G-doped boric acid glass samples. In Section 3.2, we report the generation of nonlinear signal in forward DFWM (self-diffraction) geometry as well as in backward DFWM (optical phase-conjugation) geometry using a narrow-band laser. In Section 3.3, the transient behavior of the nonlinear signal is studied using appropriate modulation of the pump laser. In Section 3.4, we use the self-diffraction geometry for the generation of nonlinear signal and report the dependence of the diffracted signal on the relative strength of the pump beam amplitudes. In Section 3.4, we study the dependence of diffracted signal on the pump beam wavelength.

3.1. Dye-doped glass as a nonlinear medium

Strong absorption of organic dyes in the visible region makes them particularly suited for nonlinear optical investigations using saturated absorption. The radiative lifetime of the lowest triplet state in dyes is quite long, but quenching mechanisms in the liquid state lead to considerable shortening of the excited-state lifetime. In the rigid environment of a solid host, the triplet state regains its radiative lifetime. This results in lowering of saturation intensity and thus allows the observation of nonlinear
effects in dye-doped solids (DDS) at very low power levels. The progress made in recent years on the realization and understanding of optical nonlinearities in DDS has been reviewed by Sharma et al. [21].

Most of the studies reported in the following sections are conducted on thin films of Rhodamine 6G dye-doped boric acid. The samples are prepared by the same method as in Kumar et al. [22]. $10^{-3}$M concentration of the dye in orthoboric acid powder is mixed and heated in a test-tube to around $270^\circ$C. The resultant homogeneous melt is poured and pressed between two glass plates preheated to $130^\circ$C. While pressing, care is taken to apply pressure uniformly, so that the thickness variations in the sample are minimal. The samples obtained were of good quality and dye was spread uniformly in the sample. These samples lasted for several months.

3.2. Generation of nonlinear signal

To generate a nonlinear signal in the self-diffraction geometry considered in Section 2, the path lengths of the two pump beams at the point of mixing in the sample should be within the coherence length of the laser. We use a CW argon ion laser (Spectra Physics, 2040-15S) with a temperature-stabilized etalon (Spectra Physics, 586) to produce a narrow bandwidth ($\Gamma \approx 20$ MHz) beam of coherence length of about 15 m at wavelength $\lambda = 514.5$ nm. For such a large value of the laser coherence length, the optical alignment for coherent overlap of the two pump beams at the sample becomes a noncritical criterion. A simple experimental setup shown in Fig. III.2 is used to generate the nonlinear signals. The entire setup is mounted on a vibration isolation table (Newport, RS510). The angle between the two pump beams is about 3° and
Fig. III.2 Experimental set-up for self-diffraction. $M_1$, $M_2$ are mirrors, and BS is a 50/50 beam-splitter. The path-difference between $k_1$ and $k_2$ beams at the sample is about 10.5 cm.
the beam radius is 1 mm for each. Using this configuration, the first-order diffraction signals from a sample of Rhodamine 6G-doped boric acid glass are obtained very easily with a laser output power of only 100 mW. The experimental setup and diffracted signals are recorded on a photographic plate. Relatively weak signals are obtained for the other argon-ion laser lines at 457.9 nm, 476.5 nm, 488.0 nm, 496.5 nm, and 501.7 nm. The same experimental setup is used to generate nonlinear signals also from $10^{-3}$ M Fluorescein-doped boric acid glass sample.

Optical phase-conjugation in dye-doped glasses has also been studied extensively [21]. We adopt the backward DFWM geometry described in Section 1.3. To generate a phase-conjugate signal, it is essential that the pathlengths of pump beams $f$ and $b$ and probe beam $p$ at the point of mixing should be within the coherence length of the laser. For a laser of coherence length $\leq 10$ cm, an experimental geometry like the one shown in Fig. 8 of Ref. [21] is used so as to match the pathlengths of $f$, $b$ and $p$ at the sample. We devise a simpler configuration shown in Fig. III.3 for the narrow-band laser mentioned above. Long coherence length of the laser made it possible for us to produce a phase-conjugate signal with very few optical elements. The angle between the forward pump $f$ and probe $p$ is kept at about $8^\circ$. The back mirror $M_3$ is fixed as close to the sample as possible. In our case, the path difference between $f$ and $b$ beams at the sample is about 12 cm, which is well inside the coherence length of the beam. The sample and various mirrors are kept slightly tilted so that backward laser beam and other reflections are not fed back into the laser. (This can also be achieved by putting a Faraday rotator and a half-wave plate at the output of the laser). A beam-splitter $BS_2$ with 5% transmittance is put in the path of the probe beam so that the pump intensity
Fig. III.3 Experimental set-up for optical phase-conjugation. f is the forward pump, b is the backward pump, p is the probe, and s is the phase-conjugate signal. M₁, M₂, M₃ are mirrors, BS₁ is a 50/50 beam-splitter, and BS₂ is a 5/95 beam-splitter. The path-difference between f and p is about 11 cm and that between b and f is about 12 cm.
is about 20 times that of probe. The conjugate signal $s$ is generated in the direction opposite to the probe beam and is extracted from the beam-splitter $\text{BS}_2$ as shown in Fig. III.3. The phase-conjugate signals are observed with $10^{-4}\text{M}$ Rhodamine 6G-doped boric acid glass at a wavelength of 514.5 nm and with $10^{-3}\text{M}$ Fluorescein-doped boric acid glass at a wavelength of 476.6 nm.

3.3. Transient behavior of the nonlinear signal

In this section, we consider the electromagnetic wave-mixing interactions inside our sample giving rise to modulations of macroscopic optical properties to create a laser-induced grating (LIG). As seen in Section 2.1, the polarization in the medium is given by the off-diagonal elements of the density operator of atoms. From Eqs. (III.2.4) and (III.2.6), it is evident that the decay of the density operator when pump beams are turned off depends on the relaxation times $T_1$ and $T_2$ of the medium. The rise time of the density matrix to its steady-state value also depends on medium relaxation times besides various pump parameters. The transient behavior of the polarization in the medium can be monitored by measuring the decay/rise of the intensity of the signal diffracted from the medium. Here we report the measurements on the transient buildup and decay of the diffracted signal from our samples, carried out at the Indian Institute of Technology, Kanpur. First we study the buildup of the self-diffracted signal. The experimental setup is shown in Fig. III.4. A 15 W CW argon ion laser (Coherent, Innova 100) provides the TEM$_{00}$ beam of 2.3 mm diameter at 476.6 nm. The laser beam is chopped using an acousto-optic modulator (AOM) (Newport, N23080 with N21080-1SAS driver). The output from the AOM is split with a 50/50 beam-splitter (BS) to get two pump beams which are made to intersect in the dye-doped glass sample symmetrically, at an angle of
Fig. III.4 Experimental set-up for transient build-up studies of the self-diffracted signal. M₁ to M₆ are mirrors. A₁ to A₃ are apertures. AOM is an acousto-optic modulator. BS is a 50/50 beam-splitter. and PD is a photodiode.
$1.65^\circ$ to the normal. The path difference between the two beams is kept at less than 1 cm and is within the coherence length of the laser. The pump beam powers are measured with a digital power meter (Newport, 815). The entire optical setup, excluding the laser, was placed on a vibration isolation table. The first-order ($n=1$) diffracted signal from the sample is collected by a photodiode (PD), or a PMT for weak signals, and fed into a chart recorder (HIOKI, 8801) along with the step-function output from a function generator (Systronics, 1012) used to trigger the AOM. The typical relaxation times $T_1$ and $T_2$ for the dye-doped solids (e.g., Rhodamine 110-doped boric acid glass) are of the order of $10^{-3}$ s and $10^{-15}$ s, respectively [23]. The chopping frequency of the pump beams is kept sufficiently low to about 10 Hz to allow the diffracted signal to reach the steady-state when the pump beam from the AOM is on. We record the buildup of the diffracted signal with time for different input pump powers (see Fig. III.5). The estimated rise time $\tau_r$ of the first-order diffracted signal for different pump powers $I_1$ and $I_2$ are given in Table III.1(a).

In the self-diffraction geometry, the pump beams diffract from the grating created by them and hence put a serious limitation in the study of the transient decay of the LIG. It is then convenient to study the decay of an uncorrelated probe beam diffracted from a decaying grating, when the pump beams are switched off. For this study, we take a weak probe beam from an independent laser (Coherent, Innova 70/4) at the same wavelength of 476.6 nm and make it incident at an arbitrary angle on the dynamic grating formed by strong, coherent pumps in the sample. A small frequency difference between the two lasers, even for the same transition, is negligible compared to the broad
Fig. III.5 Record of (a) the AOM trigger and (b) the build-up of self-diffracted signal (inverted) with $I_1 = 65$ mW, $I_2 = 62$ mW (see Table III.1(a), Sl. No. 3).
absorption spectrum [22] of the sample. Figure III.6 shows the experimental setup. We chop the pump beams by the AOM so as to study the decay of LIG formed by the superposition of the pump beams. We record (see Fig. III.7) the buildup and decay of the diffracted probe beam ($I_p$) on the chart recorder and measure the grating rise and decay times ($\tau_r$ and $\tau$, respectively). $\tau_r$ is the build-up time of the saturation and $\tau$ gives the effective time in which the saturable absorber recovers from saturation when the exciting fields are removed. The estimated grating rise times $\tau_r$ for different pump powers $I_1$ and $I_2$ are given in Table III.1(b). Grating decay times are simultaneously measured (see Fig. III.7) at a few pump powers in the range of 2 mW to 32 mW and with a probe power $I_p$ of about 8.0 mW. Though all the decays are found to be nonexponential, the grating decay curves are approximately fitted using a single exponential function, and the decay time $\tau$ is found to be 21.5 ± 5.6 ms. This is of the same order as the relaxation time $T_1$.

Table III.1. Measured grating rise times $\tau_r$ of the first-order diffracted signal from R6G–boric acid glass at 476.6 nm for various pump powers.

<table>
<thead>
<tr>
<th>Set No.</th>
<th>Sl. No.</th>
<th>$I_1$ mW</th>
<th>$I_2$ mW</th>
<th>$I_p$ mW</th>
<th>$\tau_r$ ms</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a) Self-diffraction geometry (Fig III.4)</td>
<td>1.</td>
<td>37 ± 1</td>
<td>36 ± 1</td>
<td>-</td>
<td>13.3 ± 2.2</td>
</tr>
<tr>
<td></td>
<td>2.</td>
<td>54 ± 1</td>
<td>51 ± 1</td>
<td>-</td>
<td>10.0 ± 2.2</td>
</tr>
<tr>
<td></td>
<td>3.</td>
<td>65 ± 1</td>
<td>62 ± 1</td>
<td>-</td>
<td>8.0 ± 2.2</td>
</tr>
<tr>
<td></td>
<td>4.</td>
<td>74 ± 1</td>
<td>71 ± 1</td>
<td>-</td>
<td>7.3 ± 2.2</td>
</tr>
<tr>
<td>(b) Three beam geometry (Fig III.6)</td>
<td>5.</td>
<td>1.5 ± 0.1</td>
<td>2.0 ± 0.1</td>
<td>8.3 ± 0.1</td>
<td>62.0 ± 2.2</td>
</tr>
<tr>
<td></td>
<td>6.</td>
<td>15 ± 1</td>
<td>16 ± 1</td>
<td>8.0 ± 0.1</td>
<td>28.9 ± 5.6</td>
</tr>
<tr>
<td></td>
<td>7.</td>
<td>26 ± 1</td>
<td>27 ± 1</td>
<td>8.0 ± 0.1</td>
<td>25.0 ± 5.6</td>
</tr>
<tr>
<td></td>
<td>8.</td>
<td>32 ± 1</td>
<td>34 ± 1</td>
<td>8.0 ± 0.1</td>
<td>16.7 ± 5.6</td>
</tr>
</tbody>
</table>
Fig. III.6 Experimental set-up for transient decay/build-up studies of the diffracted probe beam. $M_1$ to $M_8$ are mirrors, $A_1$, $A_2$, $A_3$ are apertures, AOM is an acousto-optic modulator, BS is a 50/50 beam-splitter, and PD is a photodiode.
Fig. III.7 Record of (a) the AOM trigger and (b) the build-up and decay of diffracted probe beam (inverted signal) with $I_1 = 32$ mW, $I_2 = 34$ mW and $I_p = 8.0$ mW (see Table III.1(b), Sl. No. 8).
By applying the rate equation formalism to a three-level saturable absorber, it can be easily shown that the recovery time $\tau$ is given by the product of the triplet state life-time $T_1$ and the quantum yield of the triplet state [21]. The saturation intensity $I_s$ is inversely proportional to $\tau$. The build-up time $\tau_r$ of the saturation depends on the incident intensity $I_1 \approx I_2 \approx I$, and for homogeneous broadening, $\tau_r \propto \tau/(1+1/I_s)$. However, our measured values of $\tau_r$ and $\tau$ do not follow this simple relation for a fixed value of $I_s$. In the three-beam geometry, $\tau_r$ at low values of the pumps is found to be longer than $\tau$. A detailed analysis allowing site to site variations and energy transport processes in the glass is necessary to account for the observed behavior.

3.4. Dependence on the relative strength of pump beams

The intensity of the diffracted signal depends on the intensities of two pump beams which form the LIG. In the next set of experiments, we use the self diffraction geometry to generate the nonlinear signal (Figure III.4 without the AOM) and vary the pump power $I_1$ of one pump beam (wavevector $k_1$), keeping the power $I_2$ of the other pump beam (wavevector $k_2$) almost fixed. We measure the power $S$ of diffracted signal of order $n = -1$ in the direction $k_4 = 2k_1 - k_2$. The pump powers $I_1$ and $I_2$ are recorded by a digital power meter (Newport, 815) and signal power $S$ is recorded by an analog power meter (Newport, 820) which is calibrated with respect to the digital power meter. The background power level is recorded for each measurement by blocking the pump beams before the sample. Variation of the self-diffracted signal $S$ with pump power $I_1$ below saturation is presented in Table III.2. Two photons from the $I_1$ beam and one photon from the $I_2$ beam are involved in the generation of the $n = -1$
diffracted beam, and hence the diffracted power in this order obeys the relation: \( \gamma \propto I_1^2 I_2 \) [16]. Thus a log-log plot of \( \gamma \) vs \( I_1 \), keeping \( I_2 \) constant, should give a slope of 2. We plot the variation of \( \log \gamma \) with \( \log I_1 \) in Fig. III.8 and the slope of the curve for \( I_1 < I_2 \) is found to be 1.94. However, for \( I_1 > I_2 \), the slope reduces to about 0.6 due to onset of saturation [20].

Theoretical treatments of multiwave mixing which are based on perturbative expansions are not valid under strong and resonant excitations in a dye-doped glass. According to a nonperturbative model by Fragnito et al. [20] for self-diffraction by nonsinusoidal population gratings in saturable absorbers, the diffracted power \( \gamma \) in the first order, for the case when \( I_1 \neq I_2 \), and when pump depletion in the medium is negligible, is proportional to \( |C_1 E_1 + C_2 E_2|^2 \), where \( |E_1|^2 = I_1 \), \( |E_2|^2 = I_2 \), and \( C_1, C_2 \) are the first two Fourier coefficients in the expansion of the saturation parameter describing the spatial distribution of excited molecules. These coefficients are given as

\[
C_1 = \frac{\left\{ (1+S_0)^2 - S_1^2 \right\}^{1/2}}{S_1 \left\{ (1+S_0)^2 - S_1^2 \right\}^{1/2}} - 1 - S_0
\]

\[
C_2 = \frac{\left[ \left\{ (1+S_0)^2 - S_1^2 \right\}^{1/2} - 1 - S_0 \right]^2}{S_1^2 \left\{ (1+S_0)^2 - S_1^2 \right\}^{1/2}}
\]

where \( S_0 = (I_2 + I_2)/I_s \), and \( S_1 = 2(I_1 I_2)^{1/2}/I_s \), \( I_s \) being the saturation intensity.
Fig. III.8  Log-log plot of self-diffracted signal $\Psi$ with the pump power $I_1$. $I_2$ is kept constant at about 9.4 mW (see Table III.2). The solid curve is the theoretical fit.
Our experimental results of Table III.2 are fitted to the theory by adjusting the values of $I_s$ and the proportionality constant. In Fig. III.8, the solid curve is the best fit to the data with $I_s = 231$ mW.

### Table III.2. Experimental Data:

Variation of self-diffracted signal with pump power.

<table>
<thead>
<tr>
<th>Pump power</th>
<th>I_1 (mW)</th>
<th>I_2 (mW)</th>
<th>Background level blocking I_2 (nW)</th>
<th>Background level blocking I_1 (nW)</th>
<th>Total background (nW)</th>
<th>Measured signal (μW)</th>
<th>S - B (μW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1 ± 0.1</td>
<td>9.4 ± 0.1</td>
<td>18 ± 1</td>
<td>18 ± 1</td>
<td>36 ± 2</td>
<td>0.70 ± 0.001</td>
<td>0.034 ± 0.003</td>
<td></td>
</tr>
<tr>
<td>4.8 ± 0.1</td>
<td>9.4 ± 0.1</td>
<td>22 ± 1</td>
<td>20 ± 1</td>
<td>42 ± 2</td>
<td>0.14 ± 0.01</td>
<td>0.098 ± 0.012</td>
<td></td>
</tr>
<tr>
<td>6.6 ± 0.1</td>
<td>9.3 ± 0.1</td>
<td>20 ± 1</td>
<td>18 ± 1</td>
<td>38 ± 2</td>
<td>0.20 ± 0.01</td>
<td>0.162 ± 0.012</td>
<td></td>
</tr>
<tr>
<td>9.0 ± 0.1</td>
<td>9.3 ± 0.1</td>
<td>26 ± 1</td>
<td>20 ± 1</td>
<td>46 ± 2</td>
<td>0.32 ± 0.01</td>
<td>0.274 ± 0.012</td>
<td></td>
</tr>
<tr>
<td>17 ± 1</td>
<td>9.4 ± 0.1</td>
<td>100 ± 10</td>
<td>60 ± 1</td>
<td>160 ± 11</td>
<td>0.70 ± 0.01</td>
<td>0.540 ± 0.021</td>
<td></td>
</tr>
<tr>
<td>43 ± 1</td>
<td>9.4 ± 0.1</td>
<td>160 ± 10</td>
<td>60 ± 1</td>
<td>220 ± 11</td>
<td>1.2 ± 0.1</td>
<td>0.98 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>66 ± 1</td>
<td>9.4 ± 0.1</td>
<td>190 ± 10</td>
<td>60 ± 1</td>
<td>250 ± 11</td>
<td>1.5 ± 0.1</td>
<td>1.25 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>80 ± 1</td>
<td>9.4 ± 0.1</td>
<td>240 ± 10</td>
<td>60 ± 1</td>
<td>300 ± 11</td>
<td>1.6 ± 0.1</td>
<td>1.30 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>97 ± 1</td>
<td>9.4 ± 0.1</td>
<td>260 ± 10</td>
<td>60 ± 1</td>
<td>320 ± 11</td>
<td>1.9 ± 0.1</td>
<td>1.58 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>118 ± 1</td>
<td>9.4 ± 0.1</td>
<td>370 ± 10</td>
<td>60 ± 1</td>
<td>430 ± 11</td>
<td>2.3 ± 0.1</td>
<td>1.87 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>186 ± 1</td>
<td>9.4 ± 0.1</td>
<td>420 ± 10</td>
<td>50 ± 1</td>
<td>470 ± 11</td>
<td>2.6 ± 0.1</td>
<td>2.13 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>289 ± 1</td>
<td>9.3 ± 0.1</td>
<td>400 ± 10</td>
<td>50 ± 1</td>
<td>450 ± 11</td>
<td>3.4 ± 0.1</td>
<td>2.95 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>412 ± 1</td>
<td>9.4 ± 0.1</td>
<td>580 ± 10</td>
<td>50 ± 1</td>
<td>630 ± 11</td>
<td>4.2 ± 0.1</td>
<td>3.57 ± 0.11</td>
<td></td>
</tr>
<tr>
<td>415 ± 1</td>
<td>9.4 ± 0.1</td>
<td>570 ± 10</td>
<td>50 ± 1</td>
<td>620 ± 11</td>
<td>4.3 ± 0.1</td>
<td>3.68 ± 0.11</td>
<td></td>
</tr>
</tbody>
</table>

3.5. Dependence on the pump wavelength

Lastly, we study the variation of the diffracted power $S$ with respect to the wavelength of the pump beam, in an experimental setup similar to above. The pump powers $I_1$ and $I_2$ are kept nearly equal at 45 mW in one set and 70 mW in another set of measurements for different pump wavelengths. The intensities are chosen keeping in mind the optimum intensities needed for generating reasonably strong signals at all wavelengths and avoiding any damage to
samples. The variation of the self-diffracted signal with pump wavelengths is recorded in Table III.3, and Fig. III.9 gives the plot of the variation of the signal $\gamma$ with wavelength $\lambda$ of the pump beam. The $S_0$ (ground-state singlet) to $S_1$ (first excited singlet) absorption spectrum of the sample has a peak at around 474 nm [22]. The spectral response (Fig. III.9) shows a nearly N-shaped dependence on detuning with a minimum at about 488 nm. The signal for wavelengths larger than 514.5 nm could not be measured for the lack of a strong laser line in that region. It is expected that the self-diffracted signal will decrease beyond 514.5 nm and the spectral dependence of self-diffracted signal will yield an M-shaped curve as is seen for phase-conjugate reflectivity [21, 22]. The peak on the longer wavelength side is clearly the more pronounced one.

For a system with two sharp singlet levels, the zero of the phase-grating contribution should occur at the absorption line centre of 474 nm. Kabanov and Rubanov (K-R) [24] considered the Stokes shift $\delta$ of the minimum of the phase grating from the absorption peak, resulting from the band nature of the singlet states. The K-R model predicts that the phase-grating has a zero at a red-shifted detuning (of excitation wavelength) of $-\delta/2$, and has two peaks on either side of this detuning, the peak at higher detuning being the stronger one, as is seen in Fig. III.9. However, according to a very recent study [25], the phase-grating contribution is expected to be masked by the amplitude-grating contribution in the wavelength region between 445 and 480 nm; and it dominates below 445 nm and beyond 500 nm.
III.9 The spectral dependence of the self-diffracted signal $\gamma$ at two pump powers (a) $I_1 \approx I_2 \approx 45$ mW (see Table III.3, Set I), and (b) $I_1 \approx I_2 \approx 70$ mW (see Table III.3, Set II). The errors in the experimental points are of the size of the points drawn. The broken curves are drawn smoothly through the experimental points.
Table III.3. Experimental Data:

Variation of self-diffracted signal with pump wavelength.

<table>
<thead>
<tr>
<th>Set No.</th>
<th>Pump wavelength λ (nm)</th>
<th>Pump power I₁ (mW)</th>
<th>Pump power I₂ (mW)</th>
<th>Back-ground level blocking I₂ (nW)</th>
<th>Back-ground level blocking I₁ (nW)</th>
<th>Total background B (nW)</th>
<th>Measured signal S (μW)</th>
<th>Net signal J' = S - B (μW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>457.9</td>
<td>46 ± 1</td>
<td>45 ± 1</td>
<td>70 ± 1</td>
<td>26 ± 1</td>
<td>96 ± 2</td>
<td>2.4 ± 0.1</td>
<td>2.30 ± 0.1</td>
<td></td>
</tr>
<tr>
<td>476.5</td>
<td>47 ± 1</td>
<td>45 ± 1</td>
<td>45 ± 1</td>
<td>18 ± 1</td>
<td>63 ± 2</td>
<td>4.2 ± 0.1</td>
<td>4.14 ± 0.1</td>
<td></td>
</tr>
<tr>
<td>488.0</td>
<td>46 ± 1</td>
<td>45 ± 1</td>
<td>66 ± 1</td>
<td>21 ± 1</td>
<td>87 ± 2</td>
<td>2.1 ± 0.1</td>
<td>2.01 ± 0.1</td>
<td></td>
</tr>
<tr>
<td>496.5</td>
<td>46 ± 1</td>
<td>45 ± 1</td>
<td>62 ± 1</td>
<td>19 ± 1</td>
<td>81 ± 2</td>
<td>3.0 ± 0.1</td>
<td>2.91 ± 0.1</td>
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4. SUMMARY

Nonlinear optical processes, such as degenerate four-wave mixing, optical phase conjugation, two-beam coupling have enormous potential for application in dynamic holography, wave front reversal, optical computing and squeezed light generation. In this chapter, we have examined the behavior of the output light generated in the DFWM process in a resonant material with chaotic light fields. The output light produced in a self-diffraction geometry is calculated nonperturbatively by the numerical solution of three coupled Langevin equations with multiplicative noise due to pump fluctuations.

We have considered a DFWM interaction for which a beam is divided into two beams, and the split-beams along \( k_1 \) and \( k_2 \) are then mixed in a thin
resonant material consisting of two-level atoms. The output light field in the mixing process is proportional to the induced polarization in the medium. The variation of the output intensity with the pump intensity for resonant pump fields is studied. Below saturation, the output intensity is seen to increase almost linearly with the pump intensity indicating a dominant presence of the zeroth-order diffraction in the output. Also it is seen that more pump power is needed to reach saturation when the bandwidth of the incident light is larger. One has to pump much harder to attain saturation with off-resonant excitation. The output is also studied as a function of detuning, for various pump bandwidths. The qualitative behavior of the total output is found to be the same as the earlier reported behavior of the FWM signal calculated using a perturbation expansion of optical Bloch equations for two-level atoms [13,15]. In order to extract information about the FWM signal along $2k_2-k_1$, Vemuri et al. [13] assumed one of the pump beams to be weak and they calculated the FWM signal to first order in the weak field. However, in this method, the basic symmetry of the FWM signals generated along $2k_1-k_2$ and $2k_2-k_1$ in a typical experimental configuration with two equally strong pumps is not recoverable. We have made an attempt to formulate the problem along this nonperturbative line of approach.

Later in this chapter, we have reported the characteristics of the nonlinear signal generated in the DFWM process in resonant samples of organic dyes doped in solid environments. These materials are useful for basic studies as they are easy to prepare, and have a high resonant third-order susceptibility arising from a long upper-state life-time of the dye. The samples of Rhodamine 6G-doped boric acid glass were prepared in the laboratory and we have reported the generation of nonlinear signal in forward DFWM (self-
diffraction) geometry as well as in backward DFWM (optical phase-conjugation) geometry using a low-power narrow-band laser. We have considered the self-diffraction geometry of DFWM process in which two equally strong pump beams create a laser-induced dynamic grating and get themselves diffracted, generating signals in new directions. The time-resolved rise of the first-order nonlinear signal has been studied using appropriate modulation of the pump laser. For the transient decay of the signal, the decay of an uncorrelated probe beam is studied as it gets diffracted from the decaying grating, when the pump beams are switched off. We have also studied the dependence of the diffracted signal on the relative strength of pump powers, and on the pump beam wavelength. We find that the spectral response is similar to what is seen for phase-conjugate reflectivity in similar samples.

In the following chapter, we use the nonlinear process of four-wave mixing studied here to generate quantum states of light exhibiting squeezing, and investigate the effect of such long life-times of states, as in our dye-samples, on the squeezing properties.
REFERENCES


