Pramāṇa – J. Phys., Vol. 34, No. 6, June 1990, pp. 461–471. © Printed in India.

Wavelength dependence of phase conjugate reflectivity in absorbing media and thermal grating studies by four wave mixing

R KRISHNA MOHAN, B RAGHAVENDRA PRASAD,
C K SUBRAMANIAN and P S NARAYANAN
Department of Physics, Indian Institute of Science, Bangalore 560 012, India

MS received 26 May 1989; revised 21 November 1989

Abstract. The development of optical phase conjugation using thermally formed gratings in absorbing media, especially the dye solutions, has besides the high efficiencies, many other characteristics which are worthy of study. Here the wavelength dependence of phase conjugate reflectivity in relation to absorption spectrum of rhodamine-6G and rhodamine-B in water, methanol and ethanol is reported. Further the Bragg condition has been verified for the dominant diffracted beam which shows that the thermal grating behaves like an optically thick grating.

Keywords. Optical phase conjugation; absorbing media; thermal grating; Bragg condition.

PACS No. 42·65

1. Introduction

Optical phase conjugation (OPC) has been achieved in several ways. These methods of generating phase conjugated wavefronts, their efficiencies, utilities, shortcomings, and applications have been dealt with in many articles (Fisher 1983; Zel'dovich *et al* 1985). We have investigated optical phase conjugation using degenerate and non-degenerate four wave mixing (FWM) in absorbing media with particular reference to rhodamine-6G (Rh6G) and rhodamine-B (RhB) in various solvents. The effects of concentration, angle of interaction and the pump-probe intensities on phase conjugate reflectivity have been dealt with earlier (Prasad *et al* 1988). The present paper deals with (i) the effect of changing the wavelength used for writing the grating on conjugate reflectivity and (ii) characterization of the thermal grating (Shen 1986).

2. Principle

The underlying principle in the technique of OPC-FWM in absorbing media is the creation of a thermal grating in the medium using high power laser beams. The dye molecules absorb the incident light energy and undergo transitions to higher energy states. Subsequent radiationless relaxation of some of the optically excited molecules, and transfer of energy to the solvent molecules causes local heating or thermalization of the solvent giving rise to periodic temperature or entropy fluctuations. These result in spatial modulation of the refractive index of the solvent which then acts as a phase grating in the scattering of incident light and is termed as stimulated thermal Rayleigh scattering (STRS).

Most of the experiments on OPC-FWM in dyes show that pure thermal effects play a major role in the recording of the interference pattern, particularly if the absorbed energy relaxes into heat immediately. The dyes were chosen as absorbing media for these investigations because of their high absorption coefficient at the operating wavelengths. This plays a major role in yielding high phase conjugate reflectivities as will be seen later. The present investigations are also aimed at studying the grating characteristics especially to study whether a thermal grating in absorbing media behaves like an ordinary grating obeying the Bragg's law of diffraction.

3. Theory

The theoretical model as presented by Hoffman (1986a, b) was taken as the working model for our investigations. From her model, one obtains an expression for the phase conjugate reflectivity R given by,

$$R = f^2 Q^2 \exp(-\alpha L/\cos\theta)\{1-\exp(-\alpha L)\}^2 I_1 I_2 G(t_D) \tag{1}$$

where I_1, I_2 are the intensities of the forward propagating pump and the backward propagating read beams respectively, α is the absorption coefficient, L is the interaction length and θ is the angle of interaction or the angle between the forward propagating pump and probe beams. f is a phenomenological factor related to quantum yield of fluorescence of the dye which gives an estimate of the fraction of absorbed light energy converted into heat. Further,

$$Q = \frac{2n(\mathrm{d}n/\mathrm{d}T)}{\lambda \rho_0 C_p}$$

is a factor which is characteristic of the solvent and contains all the solvent dependent parameters viz. ρ_0 the density, C_p the specific heat at constant pressure, $\mathrm{d}n/\mathrm{d}T$ the change in refractive index of the solvent with temperature etc.

The details of the thermal grating build up and decay along with various time scales involved in the process are contained in the factor $G(t_D)$ of (1). Since our experiment was carried out in a near zero time delay condition, the $G(t_D)$ factor for small time delay regions can be written as (Hoffman 1986b)

$$G(t_D) = \frac{t_p'^2}{3\tau_{\mathrm{eff}}^2 \Gamma_\mu^2}\left[1 + \frac{3t_D(t_D + t_p')}{t_p'^2}\right] \tag{2}$$

where

$$\tau_{\mathrm{eff}}\Gamma_u = 1 + \tau_{\mathrm{eff}} D q^2$$

where t_p' is the conjugate pulse width which can be smaller or equal to the laser pulse width t_p, t_D is the time delay of the backward propagating 'read' beam with respect to the forward propagating beams, τ_{eff} is the effective thermalization time of the solute, $1/\Gamma_u$ is the thermalization time due to noninstantaneous and nonlocal solute-solvent energy transfer, also called the energy transfer time, q, defined as $q = 2k \sin(\theta/2)$, is the grating vector and D the diffusion constant.

3.1 *Wavelength dependence*

The above expression (1) for reflectivity can be rewritten, to suit our measurements, in terms of two factors; one containing the information about the dye solvent combination, the various time-scales involved in the process, the geometrical arrangement and the intensities of the beams while the other pertaining to the explicit and implicit dependence of reflectivity (R) on the wavelength. The expression can be rewritten as

$$R = A\exp(-\alpha L/\cos\theta)\{1-\exp(-\alpha L)\}^2/\lambda^2$$
$$A = \frac{4n^2 f^2 (\mathrm{d}n/\mathrm{d}T)^2}{\rho_0^2 C_p^2} I_1 I_2 G(t_D) \tag{3}$$

While the explicit dependence is λ^{-2}, the implicit dependence of reflectivity of the process on wavelength comes due to the fact that R depends on α which in turn is a function of wavelength and any dispersion effect in ($\mathrm{d}n/\mathrm{d}T$). f is assumed to be constant. The variation of the intensities I_1 and I_2 with wavelength is not considered in (3) because, experimentally, the dye laser output, which governs these intensities, can be maintained at the same value of intensity irrespective of the wavelength of the output throughout the operating wavelength range by adjusting the Nd:YAG laser output, which pumps the dye laser. The theoretical plot of normalized reflectivity against wavelength superimposed on the plot of normalized α against wavelength for Rh6G in water is given in figure 1. The normalization is done with respect to the corresponding peak values. The figure shows that the variation of R with wavelength follows the absorption spectrum of the dye-solvent combination in the spectral region 550–580 nm.

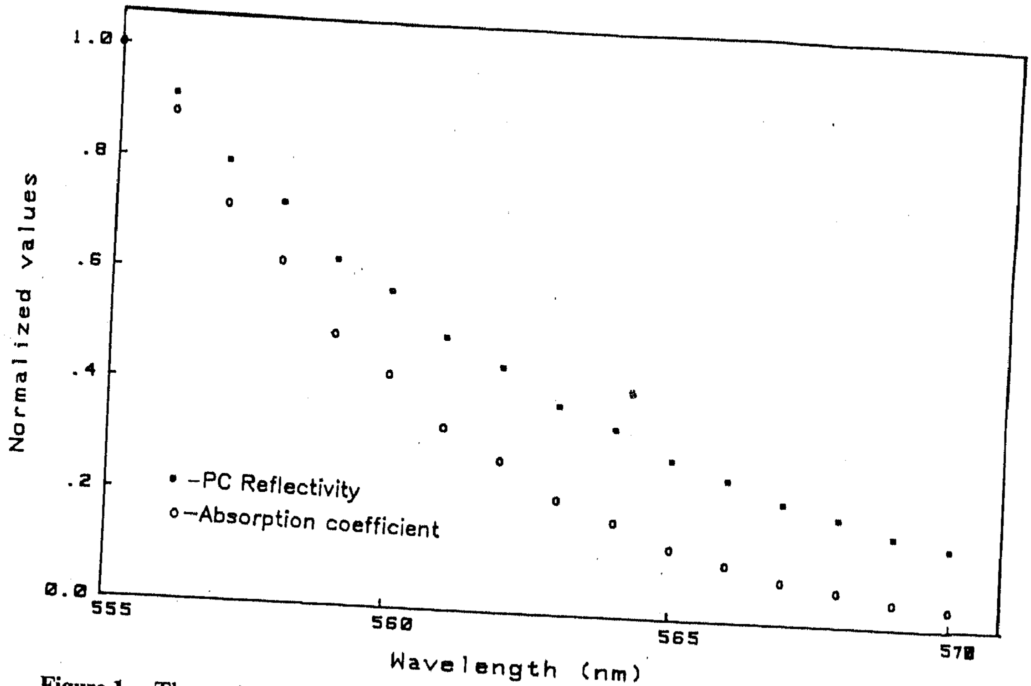

Figure 1. Theoretical plot of normalized reflectivity against wavelength superimposed on the plot of normalized absorption coefficient against wavelength for Rh6G in water.

3.2 *Thermal grating*

To study the thermal grating characteristics, a slightly modified technique termed as non-degenerate four wave mixing (NFWM) was employed. As the name suggests one of the three beams interacting in the sample cell should be of a different wavelength. An important feature of this technique is a broad acceptance angle for phase matching. In other words the process is phase matched for the 'read' beam wavelength over a range of hundreds of wave numbers for fixed pump and probe beams. It is assumed that at the power levels used, only the pump and the 'probe' beams couple to form the grating and read beam was just diffracted off the transient grating.

4. Experiment

As mentioned earlier, dyes Rh6G and RhB were chosen as the absorbing media because of their high absorption at this range of wavelengths i.e., 530 to 580 nm and their properties are well known. The experimental investigation was carried out in the wavelength interval from 555 to 575 nm as the output intensity of the dye laser falls off very rapidly beyond this interval and is not sufficient to yield an easily detectable signal.

4.1 *Absorption dependence*

Since the investigations were aimed at studying the effect of wavelength on reflectivity, all other parameters regarding the geometrical arrangement like the read beam time delay and the interaction angle etc. the medium parameters like the concentration, the interaction length etc. were fixed. The absorption spectra of different dye-solvent systems were taken by a Hitachi (model 150–20) spectrophotometer.

The experimental investigations were conducted using the output of the dye laser (PDL-II) which is pumped by frequency-doubled Nd:YAG laser (DCR-2A) with filled-in beam mode. The tunable wavelength range was 550–580 nm with the pulse duration between 6 and 8 ns and the power density used for the experiments was about $2\,\mathrm{MW/cm^2}$ per pulse.

The experimental set-up used for the investigations is shown in figure 2. The incident laser beam is split up into two vertically (or s-) polarized beams by the 10% reflecting beam-splitter BS1. The transmitted beam is then split into two beams of equal intensity by the 50% beam splitter BS2. The beam transmitted by BS2 is reflected on to the sample cell SC by a mirror M1. This beam is the backward propagating read beam. The beam reflected by BS2 is in turn reflected on to the sample cell by a mirror M2. This beam is referred to as the forward propagating pump beam or write beam. These two beams are made collinear and counter-propagating by careful adjustments of the mirrors M1 and M2. The reflected beam from the beam-splitter BS1 is further split by a 30% reflecting beam-splitter BS3 to maintain a high pump-probe ratio. The beam reflected by BS3 referred to as the forward propagating 'probe' beam, is made to interact with the 'pump' beam in the sample cell to form the thermal grating. It is mandatory to satisfy the coherence requirements for the two beams forming the grating by adjusting the path lengths of both the beams. The angle of interaction or the angle between the pump and the probe beam was maintained at 10°. The concentration of each of the dye-solvent systems was chosen to be 5×10^{-5} mol/l.

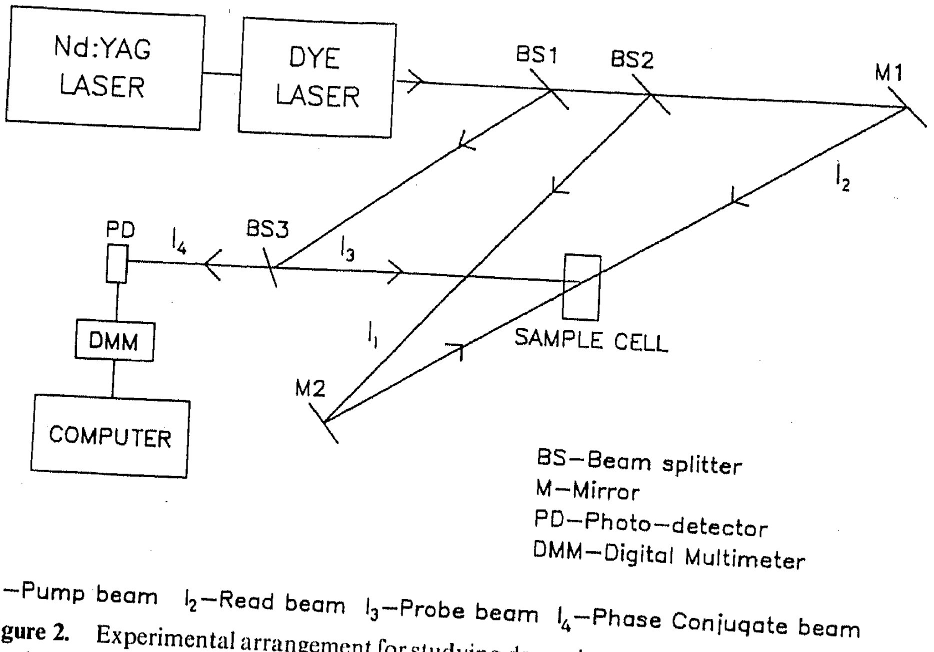

Figure 2. Experimental arrangement for studying dependence of phase conjugate reflectivity on absorption spectrum by DFWM.

The beam intensities at various stages in the set up, were monitored by photodetectors PD.

The experimental results shown in the figures reveal an interesting pattern which agrees very well with the theoretical predictions. Figures 3a to 3c show the variation of the α with wavelength for Rh6G in water, methanol and ethanol. The plots also show the variation of phase conjugate reflectivity for the same system. It is clear that the variation of reflectivity follows that of the absorption coefficient. This behaviour is identical for RhB in various solvents as shown in figures 4a–4c. In figures 5 and 6 the percentage reflectivities as function of wavelength are plotted for Rh6G and RhB in various solvents under identical experimental conditions. The highest reflectivity is observed in the case of Rh6G in ethanol, and water gives the least reflectivity. In figure 6, the situation is completely reversed with RhB in water giving the highest reflectivity which is much more than the peak reflectivity of Rh6G. RhB in ethanol gives consistently lower reflectivity than RhB in water. This dramatic change can be explained by the fact that the reflectivity is dependent on α which in turn is dependent on wavelength. At the initial operating wavelength of the dye laser i.e., at about 555 nm, the α of Rh6G in ethanol is much higher than that of Rh6G in water. Thus ethanol is a better solvent for Rh6G. But in the case of RhB, in any solvent the absorption peak is centered around 554 nm. Consequently absorption is much higher than that of Rh6G in any solvent in this range of 550–580 nm. Thus the overall higher reflectivity for RhB over Rh6G can be explained. For RhB in water the α is much higher compared to that of RhB in ethanol throughout the wavelength range and hence the reflectivity is correspondingly higher.

4.2 *Grating studies*

The experimental layout for studying the grating characteristics is given in figure 7. Non-degeneracy was introduced in the backward propagating read beam. The forward propagating pump and probe beams were kept at the same wavelength obtained from

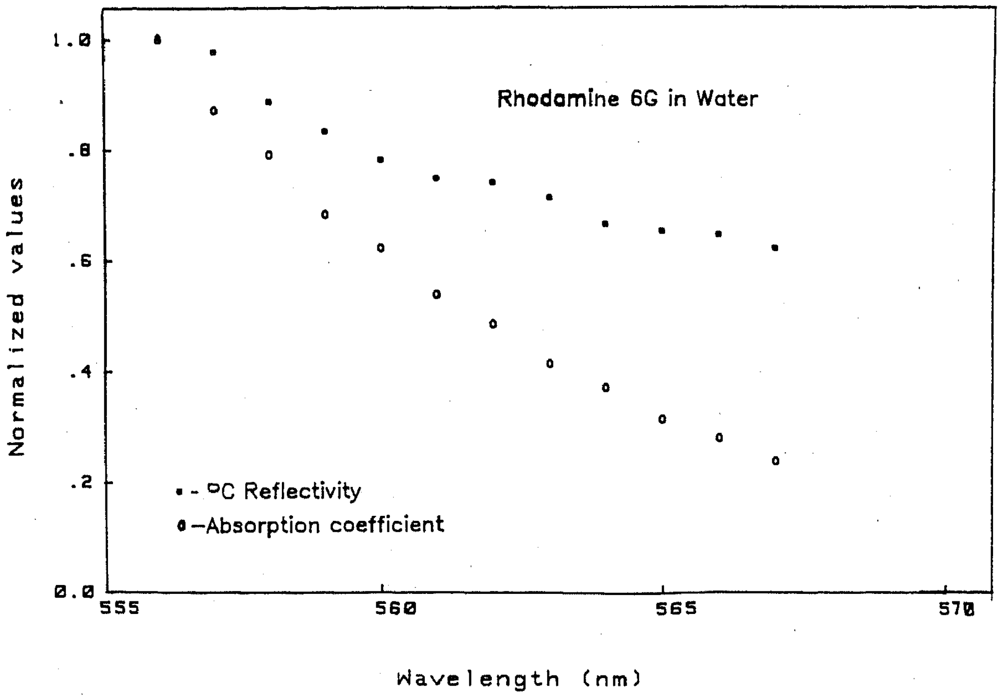

Figure 3a.

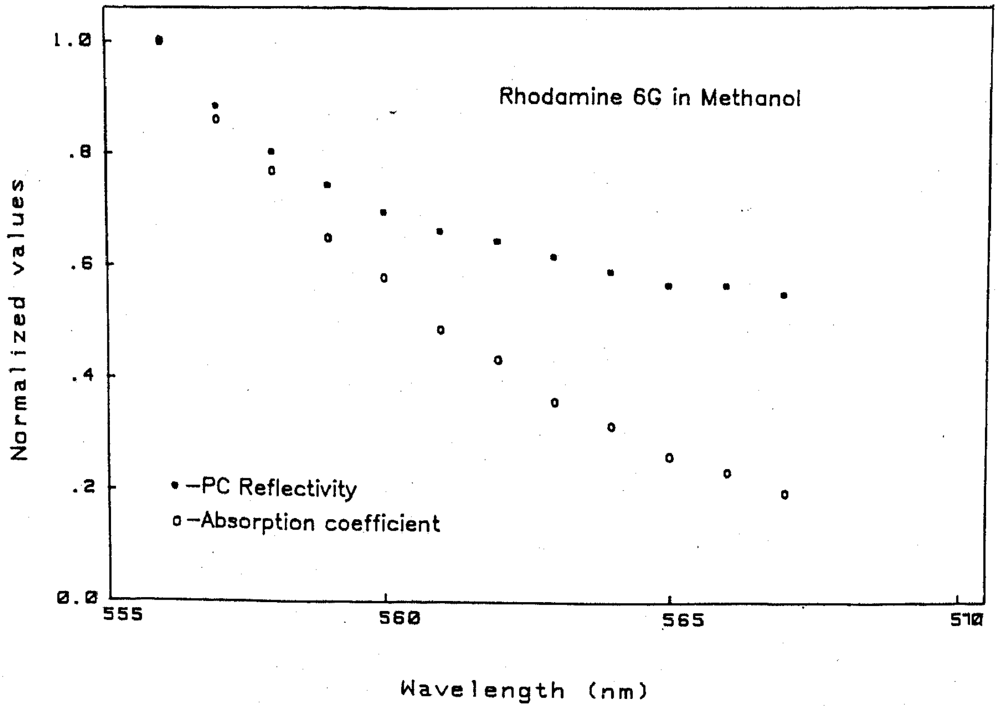

Figure 3b.

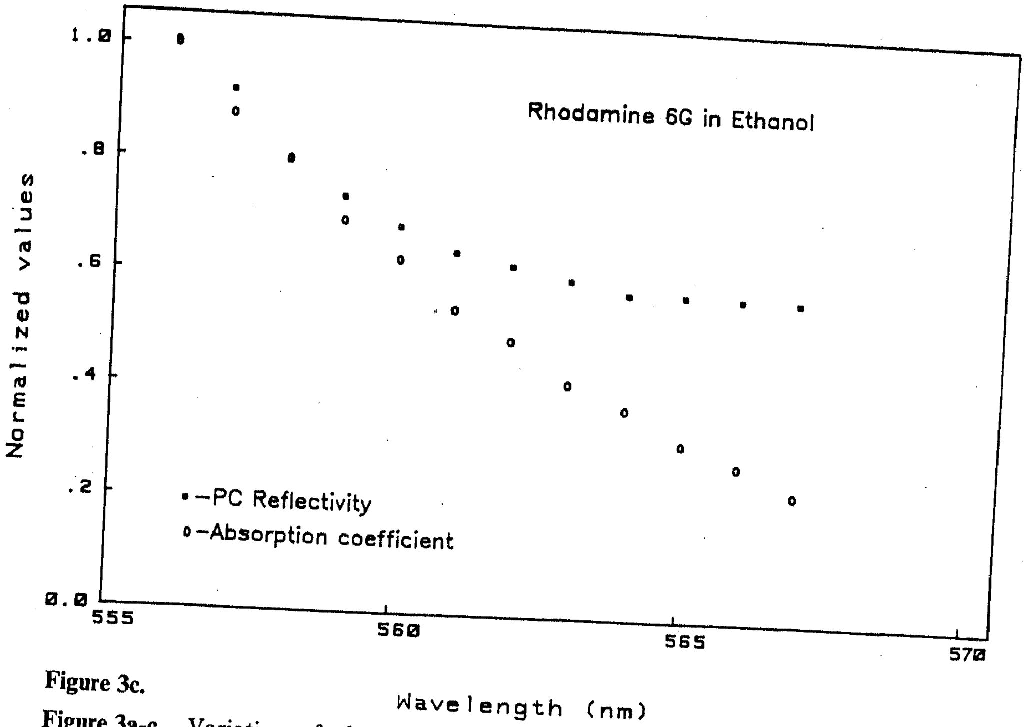

Figure 3c.

Figure 3a-c. Variation of absorption coefficient and phase conjugate reflectivity with wavelength for Rh6G in water, methanol and ethanol.

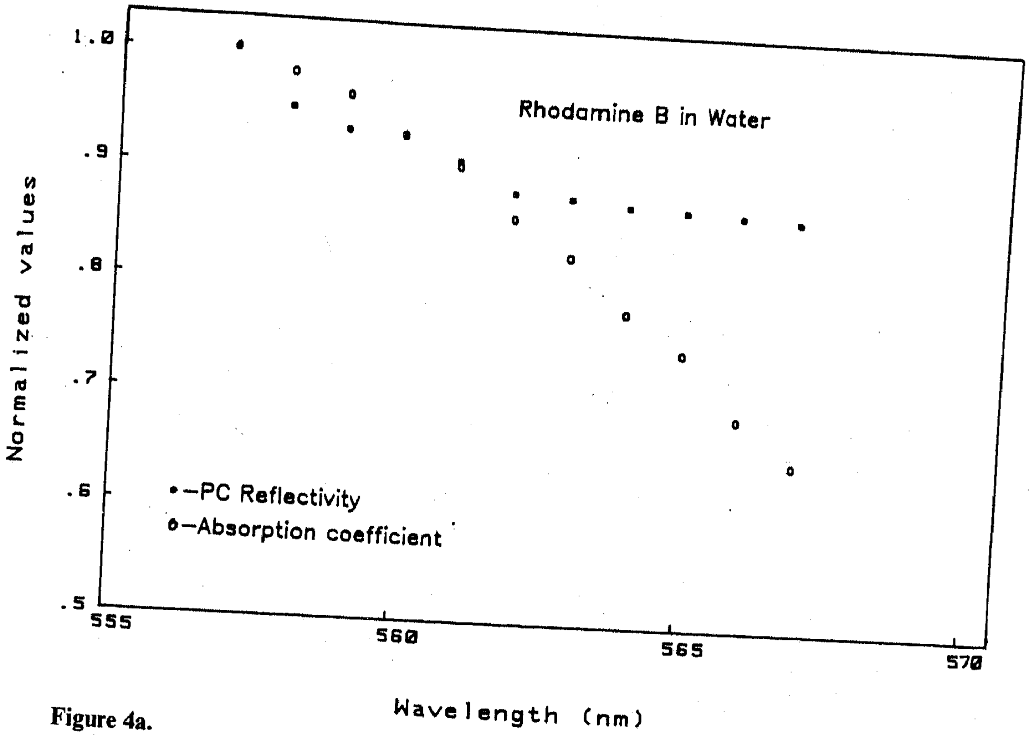

Figure 4a.

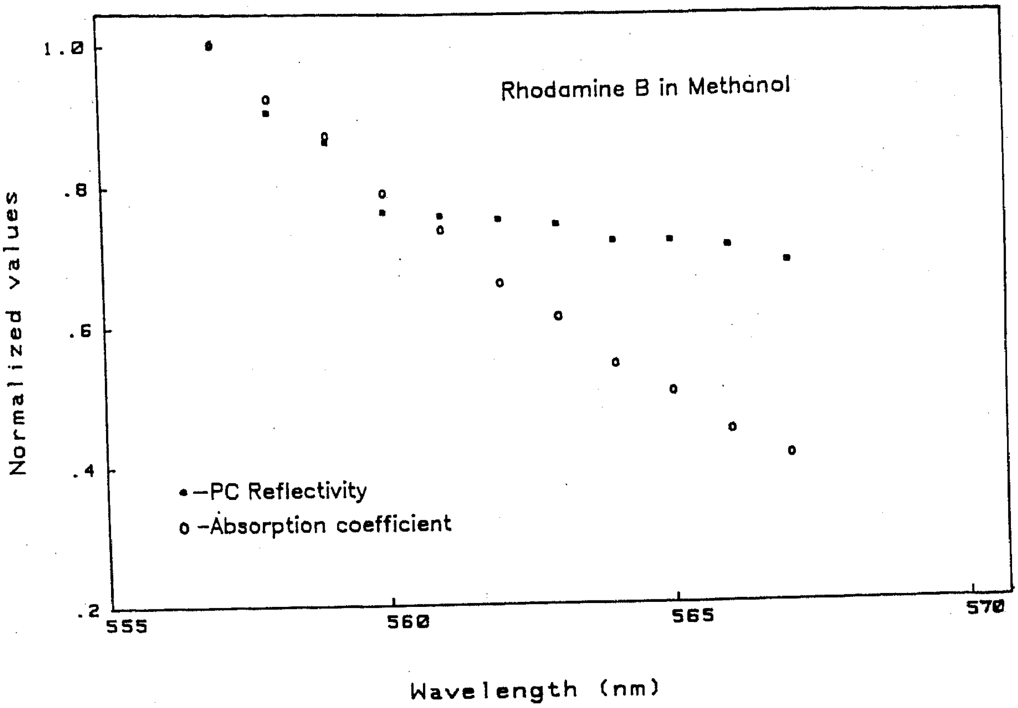

Figure 4b.

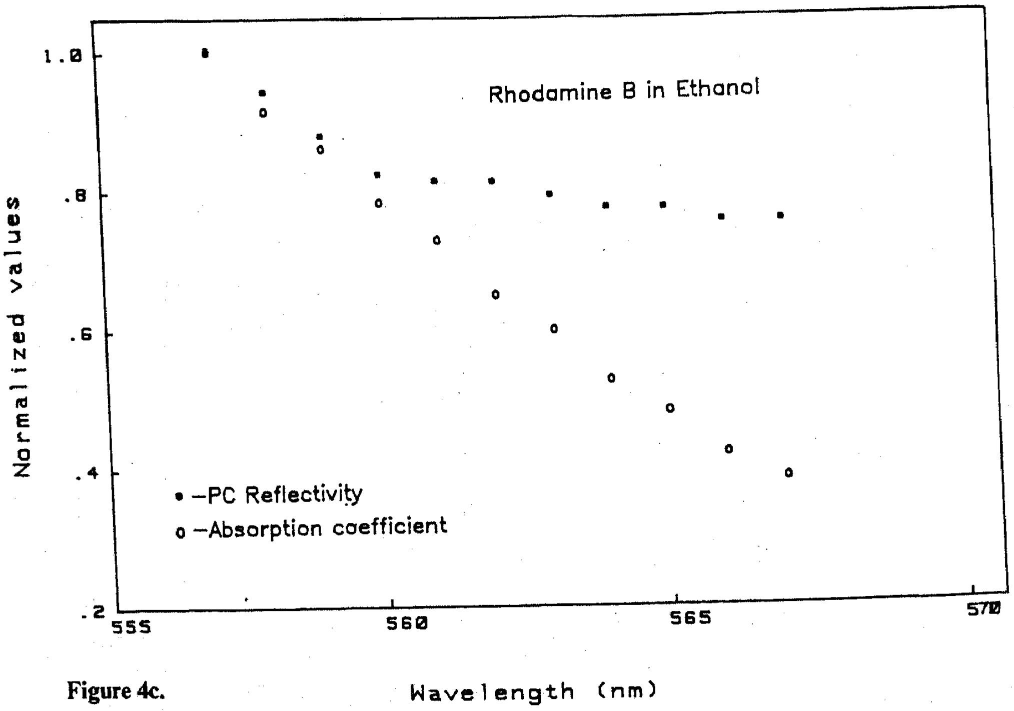

Figure 4c.

Figure 4a-c. Variation of absorption coefficient and phase conjugate reflectivity with wavelength for RhB in water, methanol and ethanol.

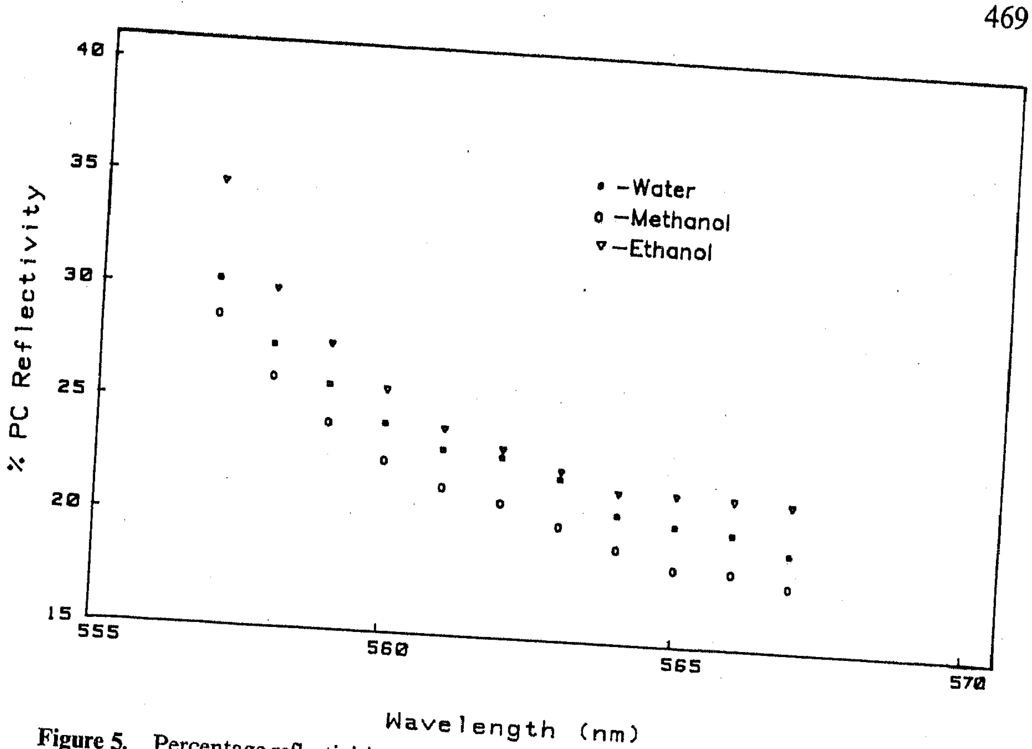

Figure 5. Percentage reflectivities as a function of wavelength for Rh6G in different solvents.

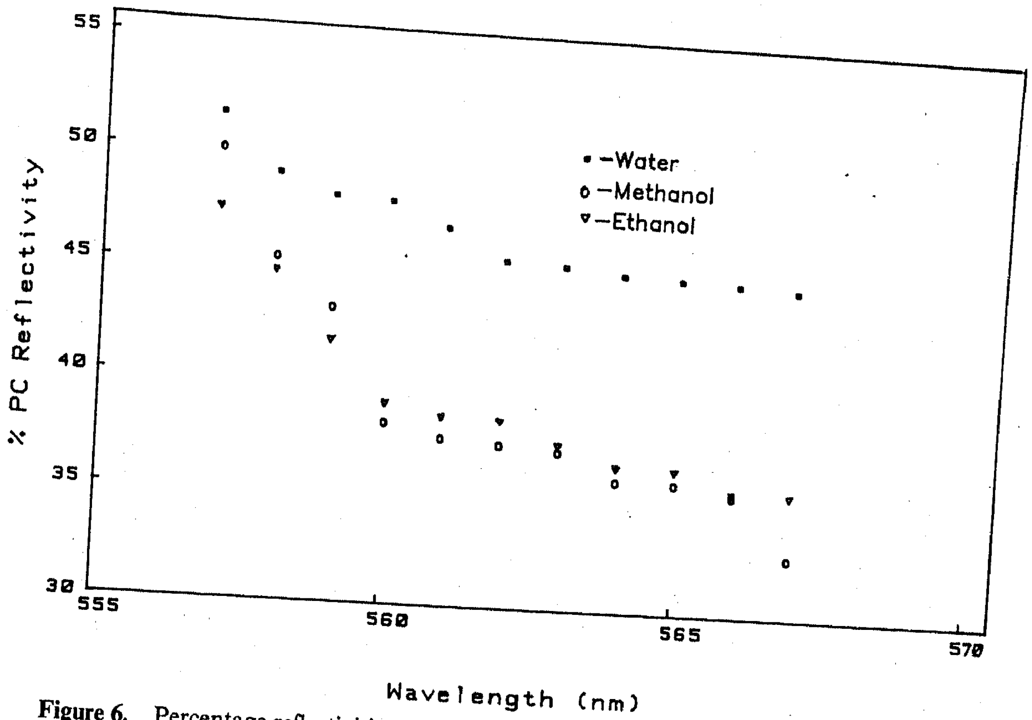

Figure 6. Percentage reflectivities as a function of wavelength for RhB in different solvents.

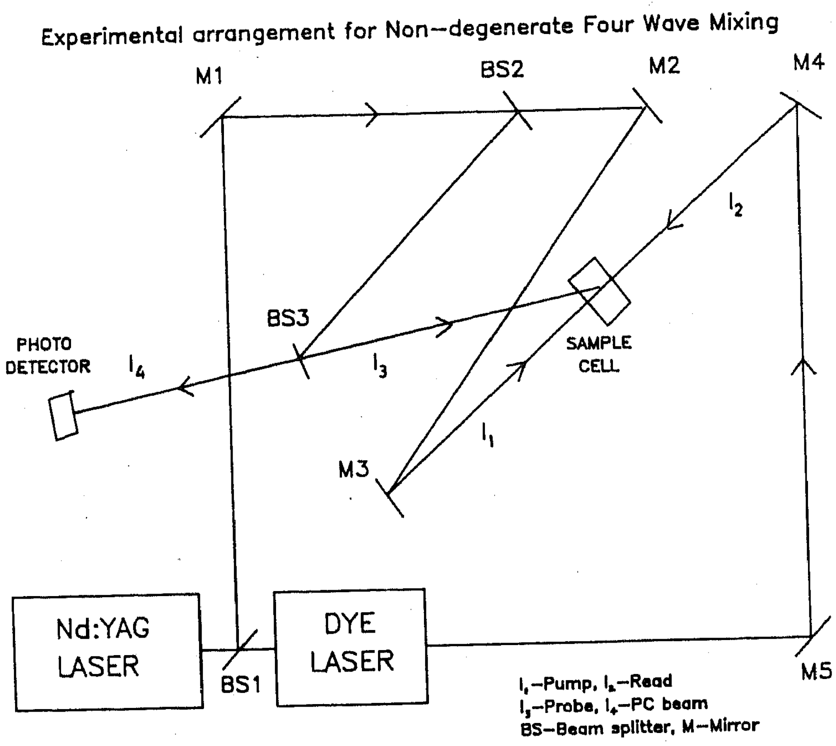

Figure 7. Experimental arrangement to study the thermal grating characteristics using NFWM.

Table 1. Theoretical calculations and experimental measurement of angular shift of read beam with wavelength in NFWM.

Read beam λ(nm)	$\theta=\sin^{-1}(\lambda/2d)$	$\Delta\theta$ (Theor)	$\Delta\theta$ (Expt.)
550	5·174		
		0·0472	0·047
555	5·2211		
		0·0472	0·047
560	5·2683		
		0·0472	0·047
565	5·3155		
		0·0472	0·047
570	5·3627		
		0·0472	0·047
575	5·4098		
		0·0472	0·047
580	5·457		

Grating forming wavelength = 532 nm, $L=332$ cm., Interaction angle = 5·0 deg., Grating spacing, $d=$ 3049·4 nm

the frequency-doubled Nd:YAG output at 532 nm. The read beam was taken from the output of the tunable dye laser PDL-II pumped by the 532 nm of Nd:YAG laser. The tunable range of the dye laser is 550–580 nm. The power density used in the experiment was about 4 MW/cm^2 per pulse.

Various dye-solvent combinations were used as the absorbing media. The concentration of the solution was kept at 3×10^{-5} mol/l to get the optimum reflectivity. The diffracted read beam was obtained as the phase conjugate signal. The read beam was tuned from 550–580 nm in steps of 5 nm. The phase conjugate beam at each of these wavelengths was detected by a photodetector mounted on a precision XY-mount placed about 3 m away from the sample cell. In all cases of the dye-solvent combination it was found that the shift in the position ΔL was equal for equal shifts in the wavelength. The angular shift was calculated from ΔL and L, where L is the distance of the detector from the sample cell using the formula,

$$\theta = \tan^{-1}(\Delta L/L). \qquad (4)$$

The value of the angular shift was also calculated using the Bragg diffraction condition. The experimental and theoretical values of the angular shift with the read beam wavelength were found to be in excellent agreement with each other.

5. Conclusion

The investigations show that the phase conjugate reflectivity of absorbing media depends to a very large extent on the absorption coefficient of the medium. The variation of the reflectivity with wavelength closely follows that of the absorption coefficient with all other experimental parameters being fixed. Further the thermal grating formed in these absorbing media behaves like a Bragg grating or an optically thick grating.

Acknowledgements

The work was carried out with the financial support from the Defence Research and Development Organization (Research and Training scheme). We also thank the Chairman, Department of Physics, Indian Institute of Science, Bangalore, for encouraging us to carry out these investigations.

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