J Indian Inst Sci., Sept–Oct 1996, 76, 577–587

# Molecular gases as Brillouin media for optical phase conjugation : An experimental investigation

B. RAGHAVENDRA PRASAD AND C. K. SUBRAMANIAN
Department of Physics, Indian Institute of Science, Bangalore 560 012, India.

Received on January 27, 1995, Revised on December 22, 1995.

## Abstract

Optical phase conjugation (OPC) studies in molecular gases by stimulated Brillouin scattering (SBS) have been carried out. In the first part, the role played by various material parameters in enhancing the Brillouin gains have been studied in four different molecular gases. The dependence of the material parameters on gas pressure and the overall gain of the media have been investigated, and the intensity dependence of OPC–SBS studied. In the second part, the dependence of efficiency and phase conjugate fidelity on interaction, coherence and acoustic lengths has been investigated.

**Keywords**: Phase conjugation, stimulated Brillouin scattering, molecular gases, time scales.

## 1. Introduction

Optical phase conjugation (OPC) through stimulated Brillouin scattering (SBS) is an inelastic scattering process. Spontaneous Brillouin scattering takes place due to the presence of propagating acoustic waves or isentropic pressure fluctuations generated by thermal fluctuations in the medium. At sufficiently high intensities the backscattered Brillouin signal can interfere with the forward-going pump beam. Since the frequency of the generated Stokes beam ($\omega_s$) is Doppler down shifted from the pump frequency ($\omega_l$), by an acoustic frequency $\omega_a = \omega_l - \omega_s = v_a/\lambda_a$, the interference pattern moves in the forward direction at the acoustic velocity, $v_a$, with an acoustic wavelength, $\lambda_a$. The presence of this time-varying electric field in a medium gives rise to a time-varying electrostrictive strain and is capable of driving acoustic waves in the medium. The presence of this acoustic wave modulates the optical dielectric constant and thus causes an exchange of energy between electromagnetic waves whose frequencies differ by an amount equal to the acoustic frequency $\omega_a$. If the energy contributed by the light waves to the acoustic wave within the period of this wave exceeds its losses due to hypersound attenuation, the acoustic wave will begin to expand in space. This will in turn cause further scattering of the pump beam by which more forward-going waves are generated at acoustic wavelength $\lambda_a$ due to electrostrictive forces. Because of these acoustic waves greater scattering of the pump radiation takes place, thus providing a gain factor to the medium and allowing the process to become 'stimulated' in the backward direction. The term stimulated emphasises the fact that the scattering oscillations in the medium are stimulated by the interfering fields themselves.

Practical realization of spontaneous wavefront reversal in SBS has made it essential to investigate the parameters of the nonlinear media that govern the efficiency of a medium, such as its electostrictive coefficient, frequency shift and the acoustic phonon lifetime. The basic physical properties that determine the Brillouin gain coefficient of the nonlinear media are its refractive index, density, viscosity, molecular weight, specific heats and thermal conductivity. Pressurised gases are very attractive for many applications of OPC–SBS due to their high transparency throughout the visible region, IR and UV regions of the spectrum. High gains can be achieved by increasing the pressure of the gas. The frequency shift of the SBS process in gases is relatively small compared to liquids and solids and this gives an added advantage for high-gain coefficients. The low-frequency shifts in gases make them suitable nonlinear media for correcting the dynamic aberrations[1,2].

## 2. Experimental layout

A high-pressure stainless steel gas cell was designed and fabricated to stand pressures up to 400 bars. The schematic of the experimental set-up is shown in Fig.1. A frequency-doubled Nd:YAG laser with a pulse duration of 10 ns was used as the pump beam source. The linewidth of the pump beam was fixed at 0.002 $cm^{-1}$. A phase distorter was used to introduce spatial inhomogeneities in the transverse intensity profile of the pump beam. The presence of strong inhomogeneities in the pump field is the necessary condition for high-quality phase conjugation in SBS. Pyroelectric detectors were used to measure the energies of pump and signal beams.

Five different molecular gases, (i) monofluorotrichloromethane ($CFCl_3$), (ii) difluoro dichloromethane ($CF_2Cl_2$), (iii) difluoromonochloromethane ($CHF_2Cl$), (iv) sulfur hexafluoride ($SF_6$), and (v) nitrogen ($N_2$) have been studied in detail. The first three gases

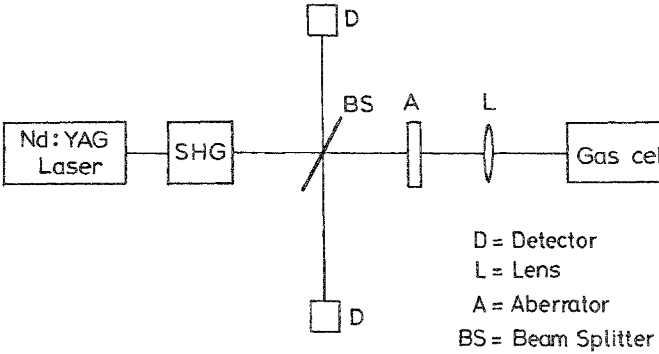

FIG. 1. Experimental layout for OPC–SBS.

are known as halogenated methanes or freons. These gases have useful characteristics such as high density, low viscosity and transparency from the UV to IR regions of the spectrum. They are highly stable even at high-power densities due to the absence of efficient overtone absorption by the high-frequency C–H vibrational modes. $SF_6$ and $N_2$ also show high stability and have been chosen for comparative study with halogenated methanes.

## 3. Dependence of OPC–SBS on material parameters

The steady-state Brillouin gain is given by[3]

$$g = \frac{\omega_s^2 \gamma_e^2}{2 n v_a \rho_0 c^3 \Gamma_B} \tag{1}$$

where $\gamma_e = \rho \frac{\partial \varepsilon}{\partial p}$ is the electrostrictive coefficient, $\omega_s = \omega_l - \omega_a$, the frequency of the scattered beam, $v_a$, the acoustic velocity, $n$, the refractive index, $\rho_0$, the density and $\tau^{-1} = \Gamma_B$ is the decay time of the acoustic grating. The acoustic frequency which determines the Brillouin shift is given by

$$\omega_a = \frac{2 n \omega_l v_a}{c} \tag{2}$$

where $\omega_l$ is the frequency of the pump beam. As can be seen from eqn (1), the gain of any medium depends on the electrostrictive coefficient, the acoustic velocity and the acoustic wave decay time. The magnitude of these quantities is determined by material parameters such as refractive index, density, viscosity, etc.

Electrostrictive effect, *i.e.*, creation of mechanical strain or change in the pressure of the media due to change in the electric field is a nonlinear phenomenon. It can be calculated by knowing the refractive index of the media through the Lorentz–Lorentz relation[4], *i.e.*, $\gamma_e = 1/3[(n^2-1)(n^2+2)]$. The gain of any medium is directly proportional to the electrostrictive coefficient. The most important parameter that determines the build up of an acoustic wave is the viscosity. The viscous damping of acoustic waves determines the gain of the media. For scattering of relatively short optical pulses, the acoustic decay time will determine the degree of transiency and efficiency of the gas for Brillouin scattering. The adiabatic density fluctuations comprising the acoustic wave result in a corresponding periodic temperature variation about the mean temperature of the medium. The thermal gradients produced thereby will be attenuated by the viscosity, $\eta$, and thermal conductivity, $\kappa$, of the medium. This type of decay is present in all media and is described by the acoustic absorption equation[5]:

$$\tau_B^{-1} = \Gamma_B = \frac{k_a^2}{\rho_0}\left[\frac{2\eta}{3} + \frac{\kappa}{2c_p}\left(\frac{c_p}{c_v} - 1\right)\right] \tag{3}$$

where $\Gamma_B$ and $\tau_B$ are the linewidth and decay time of the acoustic wave, respectively. $k_a$, the wave vector of the acoustic wave, $\rho_0$, the average density of the medium, $c_p$ and $c_v$

Table I
Comparison of theoretical and experimental gains

| Gas | *Theoretical gain* ($\times 10^{-11}$ *cm/W*) | *Experimental gain* ($\times 10^{-11}$ *cm/W*) |
|---|---|---|
| $CFCl_3$ | 1.98 | 1.62 |
| $CF_2Cl_2$ | 2.55 | 2.31 |
| $CHF_2Cl$ | 1.09 | 0.9 |
| $SF_6$ | 1.52 | 1.36 |
| $N_2$ | 0.007 | 0.004 |

are the principal specific heats at constant pressure and at constant volume. The acoustic waves that are generated in the Brillouin media move with a velocity that is characteristic of the media. Since the frequency of the generated Stokes wave is Doppler down shifted from the pump frequency $\omega_l$, by the acoustic frequency $\omega_a = \omega_l - \omega_s = v_a/\lambda_a$, the interference pattern moves in the forward direction (pump beam direction) at the acoustic velocity $v_a$ with an acoustic wavelength $\lambda_a$. The acoustic wave velocity is determined by using the gas equation.

Experimentally the gain is determined by measuring the absolute energies of the pump beam and the backscattered Brillouin signal. The backscattered signal is measured in the far-field zone with appropriate apertures to minimize the errors in energy measurements by reducing the contributions from pump uncorrelated or nonconjugate signals. The effective interaction length is determined by calculating the confocal parameter of the lens. Thus by knowing the threshold pump intensity and the interaction length, the gain is determined by making use of the *magic exponential* [$\exp(gIL) = \exp(30)$]. Table I lists the values of calculated and measured gains of the gases and are found to agree well with each other. The gas pressure of 1 bar is taken as (i) it offers a universal scale of comparison for gain in all the gases, (ii) all the available data in literature[6-8] are for 1 bar, and (iii) the maximum workable pressure that is available for $CFCl_3$ is 1 bar at a temperature of 300 K.

In ideal gases, the thermodynamic properties are independent of gas pressure, whereas in real gases they are highly pressure dependent. Change in the pressure leads to changes in its basic properties such as density, refractive index, viscosity, etc. The SBS gain as a function of gas pressure is determined by measuring the absolute intensities of the pump and backward-scattered Stokes signal. Figure 2 shows the dependence of the SBS threshold on gas pressure. There is an increase in the electrostrictive coefficient and the acoustic phonon lifetime with increase in pressure thus increasing the gain. The increase in the lifetime of the acoustic wave increases the amplitude of the acoustic wave and thus reduces the threshold by efficient scattering of the pump beam into the signal beam.

The threshold of the OPC–SBS process determines the lowest possible intensity that is required for obtaining the phase conjugate signal. This information is necessary for gain calculations—the lower the threshold the higher the gain. Systems with very low thresholds for OPC–SBS are useful for low-power applications of phase conjugation, but for high-power applications it is necessary to study the behaviour of the nonlinear media

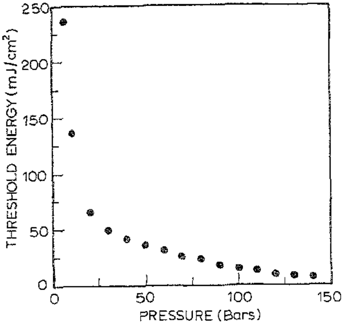

FIG. 2 Pressure dependence of SBS threshold in nitrogen.

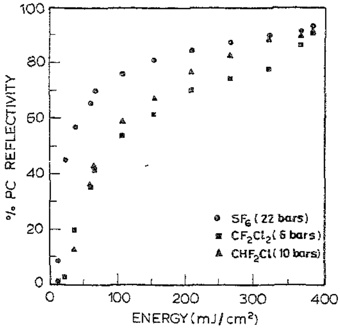

FIG. 3. Intensity dependence of phase-conjugate reflectivity in different gases.

under high-incident intensities. These studies include the determination of the saturation intensities and the dissociation energies of the nonlinear media. The phase-conjugate reflectivities as a function of pump beam intensities were determined for different pressures of gases (Fig. 3). No dissociation of gas molecules was observed even at focused power densities as high as 300 GW/cm$^2$. It can also be seen from the graphs that different gases have different saturation intensities. Low thresholds, high saturation and dissociation intensities can include these gases in the list of highly efficient nonlinear media for OPC–SBS[9-12].

## 4. Dependence of OPC–SBS gains on different time scales

SBS has been recognised as a highly efficient method for optical phase conjugation. Many applications of OPC–SBS involve the use of multimode pump lasers. For example, high-energy lasers must often be operated in multilongitudinal mode in order to extract all of the energy stored in an inhomogeneously broadened gain medium[1]. The extent to which the efficiency of the OPC–SBS process is degraded both in terms of gain and phase conjugate fidelity has to be clearly known.

There are three time/length scales involved in OPC–SBS. They are the effective interaction time or length ($L_i/c$ or $L_i$), the coherence time or length ($\Delta t$ or $L_c = c\Delta t$), and the Brillouin linewidth or the acoustic phonon lifetime$\left(\Gamma_{ph} = \tau_{ph}^{-1}\right)$ which is also known as the acoustic length ($c\tau_{ph}$), *i.e.*, the distance travelled by the scattered photon during the lifetime of the acoustic phonon. The understanding of these time/length scales is of utmost importance for gaining a complete insight into the process and to get the maximum gain under given experimental conditions for a given nonlinear medium[13-17].

Interaction length is the effective length of the medium in which the pump intensity is high enough to give rise to SBS, *i.e.*, the region in which the threshold conditions for SBS are met. Longer interaction length implies more scattering centres for the process. From the magic exponential $e^{(g/L)} = e^{30}$, for SBS process, it is evident that even if the pump intensity is low, one can still reach the threshold condition by increasing the interaction length. This implies that, the longer the interaction length, lower is the threshold pump intensity. For a focused beam geometry, the effective interaction length is set equal to the Rayleigh range or the confocal parameter of the focused Gaussian beam. The Rayleigh range is a measure of the length of the waist region of the focused Gaussian beam and alternately it is the minimum distance from the focused spot where the Gaussian beam intensity drops to half its original value. The confocal parameter which is generally used for experiments is exactly twice the Rayleigh range and is given by $L_i = \left(2\pi\omega_0^2 n\right)/\lambda$, where $\omega_0$ is the spot size and is related to the focusing optics by $(2\lambda f)/(\pi D)$, where $f$ is the focal length of the lens and $D$, the beam diameter.

The second parameter, the pump bandwidth $\Delta\nu$ determines the coherence length of the laser, and it is the length over which the frequency of the laser stays constant and is defined as $L_c = c/\Delta\nu$. When the linewidth of the pump beam is narrow, the acoustic gratings generated in the media have a better phase relationship and their scattering efficiency increases.

Brillouin linewidth ($\Gamma_{ph}$), which is a highly material-dependent parameter, determines the build up of the acoustic wave amplitude and thus is directly related to the Brillouin gain of the media. The inverse of the Brillouin linewidth is the decay time $\tau_{ph}$ of the acoustic wave. The larger the decay time, *i.e.*, the smaller the Brillouin linewidth, greater is the scattering of the pump beam into Stokes signal. The Brillouin linewidth is expressible in the spatial domain by the product $c\tau_{ph}$, which is the length travelled by a photon during the lifetime of an acoustic wave. The light scattered off a single acoustic wave maintains a constant phase over the length $c\tau_{ph}$ while the light scattered off a different acoustic wave would have a different relative phase. The time domain analogue of this is that for efficient SBS to occur temporal fluctuations of the pump must be slow compared to the phonon lifetime, *i.e.*, $(\Delta\nu)^{-1} > \tau_{ph}$, otherwise, acoustic waves do not have time to build up before the intensity of the pump drops suddenly.

Two distinct regimes of time scales, *broad-* and *narrow-band* regimes have been evolved by the combination of the time scales discussed above and their roles in affecting the gains and the phase conjugate fidelity have been investigated. In the broad-band regime, the coherence and the acoustic lengths are smaller than the interaction length and the narrow-band conditions prevail when $L_c > c\tau_{ph}$ and $c\tau_{ph} > L_c > L_i$. Experiments were carried out to study the roles of these two regimes of time scales on Brillouin gains and on the phase conjugate fidelity. $SF_6$ gas was used as the Brillouin media. The acoustic decay time or the acoustic length can be varied by changing the pressure of the gas. Lenses of different focal lengths were used for different interaction lengths. The $L_c$ of the laser can be varied from 5 to 500 cm using intra-cavity etalon and electronic line-narrowing facilities.

### 4.1. *Broad-band regime*

This regime is one in which either $L_c < L_i$ or $L_c < c\tau_{ph}$. Experiments were conducted using lenses of focal lengths 70 and 100 cm with effective interaction lengths of 13.5 and 27.5 mm, respectively. The $L_c$ of the laser was fixed at 1 cm. The gas pressure is chosen in such a way that the condition $c\tau_{ph} > L_c$ is satisfied.

The OPC–SBS reflectivities are measured as a function of the pump beam intensity for both the lenses (Fig. 4). It can be seen that longer the $L_i$ compared to $L_c$, lower is the overall phase conjugate efficiency. The reason for this is that even though the $L_i$ is long, the temporal fluctuation in the pump beam is so high that the acoustic waves do not have time to build up before the intensity of the pump drops dramatically, thus reducing the efficiency of the system. The further decrease in the conjugate efficiency with a lens of larger confocal parameter is due to the fact that the intensity distribution of the pump beam is over a larger region, thus reducing the overall available intensity to different scattering centres.

### 4.2. *Narrow-band regime*

This can be divided into two cases: one when $L_c > c\tau_{ph}$ and the second when $c\tau_{ph} > L_c > L_i$. Here the temporal fluctuations in the pump beam are low due to longer $L_c$ so that a well-defined grating is formed and there is a definite phase relationship among all the acoustic waves thus enhancing their scattering efficiency.

In the first case, for a gas pressure of 8 bars, the calculated $c\tau_{ph}$ value is found to be 24 cm. To satisfy the condition $L_c > c\tau_{ph}$, the $L_c$ of the pump beam was fixed at 50 cm. Phase conjugate reflectivity is measured as a function of pump beam intensity for different gain lengths. From the behavior of the curves in the plot given in Fig. 5, it is clear

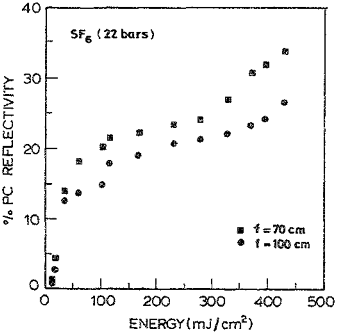

FIG. 4. Dependence of phase conjugate reflectivity on interaction length in broad-band regime.

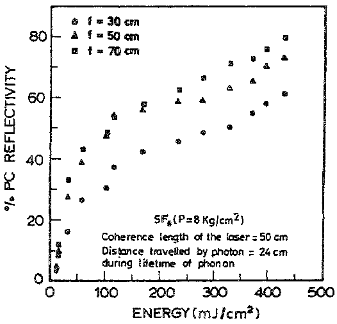

FIG. 5. Dependence of phase conjugate reflectivity on interaction length in narrow-band regime when $L_c > c\tau_{ph}$ ($L_c$ = 50 cm).

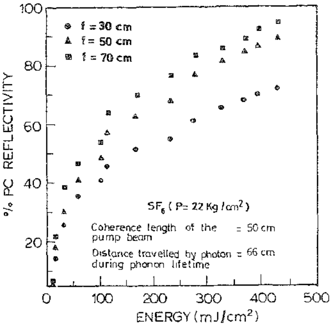

FIG. 6. Dependence of phase conjugate reflectivity on interaction length in narrow-band regime when $L_c < c\tau_{ph}$ ($L_c = 50$ cm).

that the efficiency of scattering is high for the condition $L_c > c\tau_{ph}$. It is also clear from Fig. 5 that the Brillouin gain coefficient increases with an increase in $L_c$.

In the second case, the phase of the intensity pattern formed by the forward-going pump with the backscattered signal wave is disrupted within the length $c\tau_{ph}$ by the short coherence length of the laser. That is, the pump beam loses its coherence over a distance that is short compared to the distance over which light scattered from the acoustic wave maintains its coherence. The calculated value of $c\tau_{ph}$ for $SF_6$ at a pressure of 22 bars is 66 cm and the $L_c$ of the pump beam was fixed at 50 cm. Figure 6 shows a plot of OPC reflectivity for different $L_i$ as a function of pump intensity. It is clear from Fig. 6 that the overall scattering efficiency is high for this combination due to a definite phase relationship between pump and scattered beams.

### 4.3. *Phase conjugate fidelity*

Phase conjugate fidelity is defined as the accuracy of the wavefront reversal process. The conjugation process in SBS is not perfect, *i.e.*, the backscattered signal is frequency shifted so that the phase-matching condition ($\Delta k = 0$) is not satisfied in the real sense. The extent of conjugation fidelity depends on parameters like frequency shift or $c\tau_{ph}$ and the $L_c$ of the laser beam. Experiments were conducted to study the dependence of phase conjugate fidelity on broad- and narrow-band regimes. A grid was used for creating a definite intensity pattern for the pump beam and a phase distorter was used to distort the transverse intensity profile of the pump thus distorting the image. The intensity distribution of the original pump beam, distorted pump beam and the signal beams were recorded photographically using a Linhof–Technica camera in the far field and are shown in Fig. 7(a-d). It can be seen from Fig. 7(c) and (d) that there is a dramatic change in fidelity from broad- to narrow-band regions. The reason is that the OPC–SBS process

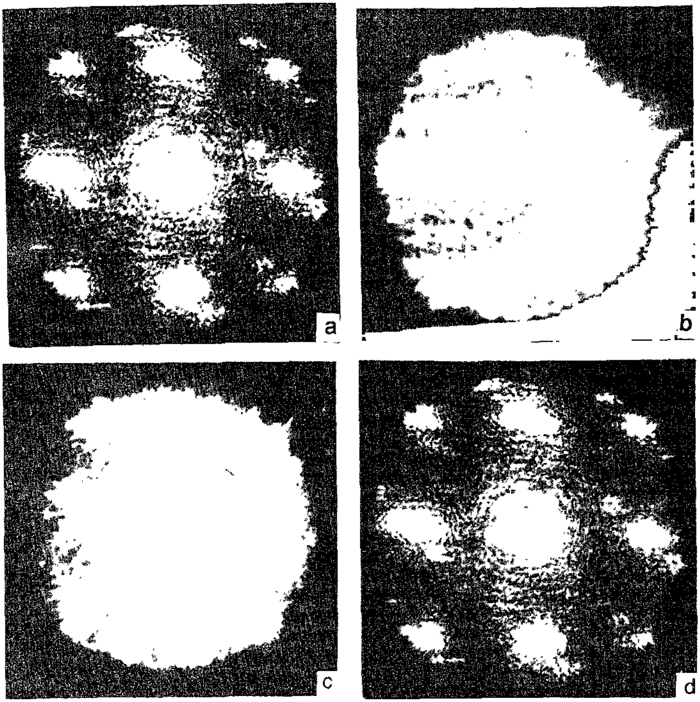

FIG 7. Distortion correction properties of OPC–SBS (a) Original beam, (b) Distorted beam, (c) Broad-band regime and (d) narrow-band regime

relies on the transformation of phase variations in the focused region, giving rise to phase- conjugated gain maze. If the region of maximum reflectivity is incoherent with the noise from the far field, the SBS will not be a phase conjugate of the input. This is the reason for the improvement in phase conjugate fidelity in the narrow-band regime.

## 5. Conclusions

The Brillouin gain is a highly material-dependent parameter, *i.e.*, it depends mainly on the refractive index, density and viscosity of the medium. The refractive index determines the electrostrictive coefficient and the gain is directly proportional to this. The density and viscosity determine the Brillouin linewidth or the acoustic wave decay time. The gain is high for systems with higher acoustic decay times. The Brillouin gain also depends on the acoustic velocities, and the lower the velocity, higher is the gain. From these studies it has been found that the halogenated methanes and sulfur hexafluoride are highly efficient systems compared to nitrogen. The increase in the gas pressure results in increase of the electrostrictive coefficient and the acoustic wave decay times thus enhancing the gain of the medium.

From the above studies it is clear that the efficiency of the process OPC–SBS can be improved by choosing an appropriate time/length scale. It is also found that the fidelity of the process improves remarkably in the narrow-band regime. It is possible to have a very high efficiency phase conjugator with very good fidelity even at low working pressure of gases by proper choice of pressure, coherence length of the pump beam and finally the focusing optics.

## Acknowledgements

The authors gratefully acknowledge the help and encouragement given by late Prof. P. S. Narayanan and dedicate this work to his memory. Funds were provided by the Defence Research and Development Organisation (DRDO), Ministry of Defence, Government of India, under its Research and Training programme.

## References

1. Zeldovich, B. Ya., Pilipetsky, N. F. and Shkunov, V. V. — *Principles of phase conjugation*, 1985, Springer.
2. Basov, N. G. (ed) — *Phase conjugation of laser emission*, 1988, Nova Science.
3. Kaiser, W. and Maier, M. — *Laser handbook*, Vol. 5 (F T. Arechi and S. DuBois, eds), 1972, pp. 1077–1150, North-Holland.
4. Erokhin, A. I., Kovalev, V. I. and Faizullov, F. S. — Determination of the parameters of a nonlinear response of liquids in an acoustic resonance region by the method of non-degenerate four-wave interaction, *Sov J. Quant. Electron.*, 1984, **14**, 209–214.
5. Damzen, M. J., Hutchinson, M. H. R. and Schroader, W. A. — Direct measurement of the acoustic decay times of hypersonic waves generated by SBS, *IEEE J.*, 1987, **QE-23**, 328–334.
6. Braker, W. and Mossman, A. L. — *Matheson gas data book*, 6th Edn, 1980, Lindhurst.
7. *Gas encyclopedia, l'air liquide*, 1976, Elsevier.
8. *Handbook of compressed gases*, 1966, Reinhold.
9. Rockwell, D. A. — A review of phase conjugate solid state lasers, *IEEE J*, 1988, **QE-24**, 1124–1140.
10. Kummrow, A. and Meng, H. — Pressure dependence of stimulated Brillouin backscattering in gases, *Opt. Commun*, 1991, **83**, 342–348.
11. Raghavendra Prasad, B., Wanare, H., Raghuveer, S., Subramanian, C. K. and Narayanan, P. S. — Optical phase conjugation by stimulated Brillouin scattering in halogenated methanes, *Curr. Sci.*, 1993, **65**, 979.

12. RAGHAVENDRA PRASAD, B., RAGHUVEER, S., SUBRAMANIAN, C. K. AND NARAYANAN, P. S. Optical phase conjugation studies in high pressure nitrogen gas by stimulated Brillouin scattering, *Proc. Natn Laser Symp.*, Feb. 1993, Indian Institute of Technology, Madras, pp. 213–214.

13. MULLEN, R. A., LIND, R. C. AND VALLEY, G. C. Observation of stimulated Brillouin scattering gain with dual spectral line pump, *Opt Commun.*, 1987, **63**, 123–128.

14. VALLEY, G.C. A review of stimulated Brillouin scattering excited with a broad band pump laser, *IEEE J*, 1986, **QE-22**, 704–712.

15. NARUM, P., SKELDON, M. D. AND BOYD, R. W. Effect of laser mode structure on stimulated Brillouin scattering, *IEEE J*, 1986, **QE-22**, 2161–2167.

16. SEIGMAN, A. E. *Lasers*, 1986, Oxford University Press.

17. RAGHAVENDRA PRASAD, B. *Optical phase conjugation studies in molecular gases by stimulated Brillouin scattering and in organic systems by four wave mixing*, Ph. D. Thesis, Department of Physics, Indian Institute of Science, Bangalore, India, 1993.