

LASER RELATED RESEARCH AT COCHIN UNIVERSITY OF SCIENCE AND TECHNOLOGY - AN OVERVIEW

Prof. C P Girijavallabhan

International School of Photonics

Cochin University of Science & Technology

Cochin 682 022, INDIA

Email: vallabhan@cusat.ac.in

1. Introduction

In this article an attempt is made to summarize the research and development activities that were being carried out during the last five years at Cochin University of Science and Technology (CUSAT) in the area of lasers and optoelectronics. Obviously I will only be able to touch upon the salient features and a few important results due to constraints on space. Whenever possible, references to the original publications have been given so that further detailed information is available to the interested reader.

At CUSAT we have made a modest attempt to concentrate on certain selected topics for which the foundations were laid during the eighties. Prominent among these topics are time and space resolved studies on laser produced plasmas, photoacoustic and photothermal effects, nonlinear optics and optogalvanic effect. Development of fibre optic sensors is yet another area in which some notable efforts have been made during this period. A brief account of the work done in each of these areas is given in the succeeding sections.

2. Laser produced plasmas

Laser-matter interaction is a subject of considerable significance as well as practical importance. The evolutionary history of laser induced plasma (LIP) can be traced out best by making spectral studies in a time resolved manner. Spacewise mapping of spectroscopic quantities yields a great deal of information on the dynamical aspects such as distribution of particles like neutral atoms, ions and molecules as well as clusters and their velocities in the laser induced plasma. At CUSAT, investigations have been carried out using a variety of target materials like metals, insulators, semiconductors, polymers and high temperature superconductors. These studies have yielded many interesting results and provided a better understanding of the whole range of phenomena associated with LIP. Such measurements have assumed great practical significance in the context of laser deposition of thin films of various materials including diamond like thin films. Some of the most important of these results are outlined below.

In our experiments the surface of the target material which is kept in a chamber maintained at a low pressure ambient atmosphere of appropriate gases is irradiated with the output from a Q-switched Nd:YAG laser. The details of the experimental set up are given elsewhere (1). Earlier studies on the time variation of emission intensities from different atomic, ionic and molecular species revealed clearly measurable time delays which are characteristic of the species (2). These delays are similar to the time of flight measurement and consequently they yield fairly accurate information on the velocities of different species ablated from the surface of the target

due to laser irradiation. However, the temporal profile of emission from Yttrium atoms and YO molecules yielded a twin peak structure while this was absent in the case of Y^+ when YBCO ($YBa_2Cu_3O_7$) superconducting material was used as target (3). This was interpreted as due to the different formation mechanisms of the particular species. For example, excited YO molecules could be ablated directly from the surface of the target giving characteristic diatomic molecular emission spectrum from a certain section of the plasma plume. It is also possible that collision of Yttrium and Oxygen atoms can lead to formation of excited YO molecules in the region in front of the target and this could yield another YO emission peak. Quite recently we have observed a double as well as triple peak structure in the emission profile of C_2 molecules when graphite target is irradiated with Nd-YAG laser beam (4-9). Such studies have been carried out in an extensive manner by varying the laser energy under different ambient gas environments. The length of the plume in the LIP is maximum under a low pressure He ambient while it is considerably shortened in an Argon atmosphere (10).

Two vitally important parameters of the plasma are the electron temperature and electron density. Measurements of line widths of stark broadened emission lines and line intensity ratios directly give information on these two quantities. We have shown that the electron density varies as z^{-1} and t^{-2} while the electron temperature shows a $z^{-0.1}$ and t^{-2} type dependence where z is the distance from the target surface and t , the time after the termination of the laser pulse (11). Studies have also been carried out to determine the electron density and temperature in superconducting $YBa_2Cu_3O_7$ plasma (12,13). Laser interferometric techniques are also employed to measure electron densities and such measurements demonstrate plasma shielding at high laser intensities (14). The interaction of ablated species with ambient gas molecules can give rise to formation of molecules like nitrides and oxides. We have in this case studied the emission from diatomic CN from which the values of vibrational temperatures of the molecular species have been obtained in the expanding plasma generated by laser irradiation of graphite (1,15).

Another interesting result that we have been able to obtain in the case of LIP of YBCO in air at atmospheric pressure is the inversion phenomenon exhibited by Ba^+ resonant emission line. There is a striking dip at the line centre revealing absorption of the unshifted Ba^+ emission wavelength due to cooler outer layers in the LIP from YBCO at a laser intensity upto 1.6 GW cm^{-2} . When the incident laser intensity reaches 1.6 GW cm^{-2} a new peak begins to appear at the line centre which is well within the inversion dip. Such curious life profile is an indication of anisotropic resonance scattering taking place in the boundary layers of the LIP (16).

We have also succeeded in placing a Langmuir probe into the plasma chamber to look at the probe current variation as a function of time (17). We have noticed that the electron energy values range from 50 - 100eV in the case of prompt emission and obviously this must be due to the ponderomotive force of the electric and magnetic fields provided by the focused laser beam. It is noteworthy that these high energy electrons are able to ionize and vibrationally excite the nitrogen molecules in the low pressure ambient air and a clear signature of the molecular N_2^+ emission occurring within 50 ns after the laser pulse can be clearly recorded.

3. Studies on photoacoustic phenomena

Photoacoustic (PA) effect and its applications form a very important area of research in which the work carried out at CUSAT has become noteworthy at international level. A comprehensive review on photoacoustic instrumentation done in this context is now available in the literature (18). Only a brief survey of the major achievements in this area is given below.

3.1 PA set up using continuous wave laser as the excitation beam

A detailed description of the experimental set up is shown elsewhere (18). Resonant PA cell also has been constructed and used in certain experiments. They are especially useful for the study of phase transitions in solids (18).

Non-destructive testing of multilayer dielectric coatings ($\text{SiO}_2/\text{TiO}_2$ structure) has been carried out using the photoacoustic technique (19). This technique makes use of a 10mW He-Ne laser, a photoacoustic cell and a lock-in amplifier. In this case PA signals were measured as a function of the chopping frequency. Striking step-like variations are observed in the phase against frequency plot which clearly reveals the different layers present in the multilayer structure. The number of steps in the phase plot exactly matches the number of steps in the thin film structure. This work shows that the phase dependence of the chopping frequency of the photoacoustic signal is more sensitive than the dependence of PA signal amplitude on chopping frequency when a large number of layers are present in the thin film structure. It has been demonstrated that photoacoustics is an ideal technique to probe and analyse multilayer stacks non-destructively (20).

Bleaching of cresyl violet in polyvinyl alcohol due to irradiation by a He-Ne laser has been investigated using photoacoustic technique (21). Our studies show a decrease in PA signal amplitude with irradiation time. This information is very useful since cresyl violet can be used as a potential medium for holographic recording and information storage.

Another use of PA technique is the measurement of thermal diffusivity of various materials available in small quantities. We have used PA studies for the measurement of thermal diffusivity of some halogeno benzimidazole complexes of cobalt(II), copper (II) and copper (I), Phthalocyanines (22,23). Phthalocyanines are good organic semiconductor materials. The measured values of thermal diffusivity of phthalocyanines show that their amplitudes are comparable with those in semiconductors like Si and Ge and hence thermal conductivity of these materials are suitable for their use in various devices .

Another interesting result obtained in this connection is the observation of thermal anisotropy in crystals like KDP. The variations of thermal diffusivities with temperature in this crystal obtained along the a or b axis and that along c axis and this clearly reveals the anisotropy in thermal conductivity (24). Gamma alumina is yet another interesting material of considerable industrial importance. The thermal properties of these materials have been investigated using PA effect. The influence of hydroxyl groups on thermal diffusivity was studied by degassing the sample at different temperatures in these experiments (25).

3.2 Pulsed photoacoustic measurements

The PA cell is made of stainless steel with glass windows for the entry and exit of the laser beam. The acoustic transducer that detects the laser-induced PA signals is a PZT (Lead Zirconate Titanate) disc. The second harmonic output beam from a Q-switched Nd:YAG laser is focused into the PA cell containing the sample. The transducer output is observed on a digital storage oscilloscope. The detailed schematic of the set up is shown elsewhere (25).

A detailed description of the pulsed photoacoustic experimental set up is given elsewhere (26). Pulsed PA method has extremely high sensitivity and hence it can be used for probing very weak nonradiative processes. The information thus obtained is complementary to that from fluorescence measurements. We have carried out a series of pulsed PA experiments in selected laser dyes, organic liquids and fullerenes. In the case of dyes like rhodamine 6G, PA measurements yield absolute value of fluorescence quantum efficiency (26,27). It also yields information about multiphoton absorption (MPA) processes taking place in the dye solutions and processes like dimerization, aggregation, complexation etc. in laser dyes (28,29).

Recently we have carried out pulsed PA measurements in fullerenes. Though PA measurements do not directly reveal the occurrence of two photon absorption processes in C_{60} and C_{70} (30,31), evidences have been obtained to show the role of excited state absorption (ESA) in the process of optical limiting by C_{60} and C_{70} solutions in toluene. We have also proved that PA technique is an excellent tool for detecting photoinduced dissociation processes in organic liquids (32,33). In the case of carbon disulphide, four or five photons at 532 nm can be directly absorbed by a molecule leading to dissociation or photofragmentation (34).

4. Dual-beam thermal lens studies

Thermal lens effect is a very useful photothermal phenomenon which can be profitably exploited for the measurement of material properties. We have successfully set up dual-beam thermal lens experiment which can be used for the measurement of thermal and optical properties of weakly absorbing materials.

The experimental set up for the dual-beam thermal lens study is given elsewhere (35-37). Knowledge of the fluorescence quantum efficiency of laser dyes and their concentration dependence is essential for selecting efficient laser media. Thermo-optic techniques, such as thermal lens technique and photoacoustic techniques, have been adopted recently for this purpose. The thermal lens method offers significant advantages over traditional optical detection methods since (i) it does not require a standard and hence reduces uncertainties and (ii) the quantum yield can be determined absolutely and with high confidence. We have carried out double beam thermal lens studies in selected laser dyes and measured their fluorescence quantum yields (37). Quite recently we have measured the thermal lens effect in C_{60} and C_{70} solutions and the results clearly show the occurrence of two photon absorption processes in addition to ESA (38).

Thermal diffusivity is an important parameter which controls the rate at which heat may flow through the medium. Pulsed thermal lens technique can be successfully adopted for thermal diffusivity measurements. We measured values of thermal diffusivity of several liquids using the pulsed thermal lens technique (39). The measured values of thermal diffusivity are in good agreement with the literature values. However aqueous solutions gave results which were strongly dependent on concentration and nature of the dissolved material. This has yielded new understanding on the thermal diffusion processes in ionic solutions (40).

5. Photothermal beam deflection studies

Photothermal deflection (PTD) Technique is one of the sensitive laser based measurement methods which we have perfected in our laboratory. The technique is based essentially on the changes in the refractive index of the medium produced by the absorption of optical radiation (also called the pump beam). It is detected by sending a second, low power laser beam (probe) through the sample or through the adjacent coupling medium so that the probe beam will experience a change in optical path length causing it to deflect from the original direction. This phenomenon is also called mirage effect.

In our PTD experiments, the details of which have been published earlier, (41,42) the mirage effect is detected by means of an optical fibre based position sensitive detector. It must be noted here that being an extremely sensitive and non contact method of measurement, PTD set up is ideally suited for investigating thermal and optical properties of very delicate materials like liquid crystals and thin films. Some of the major results of these experiments are outlined below.

We have used the cw PTD set up to determine the thermal diffusivity of various samples available only in small quantities. The pump beam is a chopped 488nm radiation from an Ar ion laser at 40mW.

Striking variations in the PTD signal take place at the temperature at which phase transitions occur in the sample material. PTD studies have been made on phase transition in ferroelectric triglycine sulfate (TGS) and barium titanate (BaTiO_3) (43,44), liquid crystals viz. E8, M21 (44) and M24 and in some metal phthalocyanines (Mpc).

Laser induced damage and ablation thresholds in some polymers like nylon, teflon and perspex and in some high temperature superconducting samples (41) are also studied using PTD. Here the pump beam used is a 1.06micron radiation from a Nd-YAG laser operated in single shot mode. The polymer samples studied (nylon, teflon and perspex) (46) are increasingly being used for the manufacture of optical components.

When a high power laser beam is incident on a surface, there will be a mass transport in the surrounding gas medium directed away from the target surface (47). We have measured this mass transport velocity using PTD from different targets. The single beam PTD has also been used to carry out imaging in different samples with internal inhomogeneity and physical defect (42). Polypyrrole thin films doped with iodine at spatially selected locations and at different doping concentrations have also been examined using PTD technique to demonstrate its usefulness to study the presence of dopants and their spatial variation in concentration (48).

The laser beam deflection technique has been successfully employed to study the behaviour of stratified fluid media. This experiment also has led to the first direct observation of what is known as internal wave phenomenon in stratified fluids (49). The diffusion process occurring in such media has been studied using beam deflection technique (50).

6. Optogalvanic studies

Tunable lasers are excellent light sources to induce optogalvanic (OG) effect. When the wavelength of a laser beam passing through an electrical discharge is finely tuned, perceptible changes in the impedance takes place corresponding exactly to those wavelengths which are able to induce transitions between the energy levels of the various atomic, ionic or molecular species in the discharge. Thus one can get an optogalvanic (OG) spectrum by monitoring the discharge current as a function of wavelength. With the help of a tunable ring dye laser capable of operating in single mode with very narrow linewidth (<5 MHz), we have successfully carried out high resolution OG studies in various gas discharges. Notable among these is the high-resolution spectroscopic study of nitrogen discharge (51). The N_2 rotational spectrum of (11,7) band in the first positive system of N_2 molecule has been recorded using OG technique and assignment of 432 rotational levels belonging to the 27 branches of this band has been carried out.

Also pressure broadening effect has been studied by measuring the linewidth of Ne emission line using OG effect (52,53). In another experiment modification of spectral characteristics and OG response in Ne hollow cathode under laser excitation has been investigated (54). In what is called photoemission optogalvanic (POG) effect changes in discharge current take place when one of the electrode is irradiated with a laser beam. We have used pulsed laser beams to investigate POG effect in number of metals like copper, silver and gold by using these as electrode materials (55,56). Multiphoton induced POG effect has been found to take place when these materials are used as irradiated electrodes in a discharge tube. POG effect near the instability region of a hollow cathode discharge has also been investigated (57). Characteristic features of chaotic phenomena has been observed with laser irradiation of the discharge (58).

7. Design and Development of optical fibre sensors

Fibre optical technology forms an integral part of photonics. Optical fibre communication and the development of optical fibre sensors (OFS) are the two essential aspects of this novel and emerging branch of technology. At CUSAT we have been paying some attention to the latter area in the past few years. OFS possess some unique characteristics and advantages over the conventional electromechanical/electronic sensors. Immunity to electromagnetic interference, extreme sensitivity, compactness flexibility and their usefulness to work satisfactorily in hazardous environments are some of the features that make them superior to conventional sensors.

We have in the past few years made some serious attempts in the direction of the design and development of intensity modulated fibre optic sensors. The range of FOS developed at CUSAT includes those for measurement of temperature, pressure, viscosity and surface tension of liquids and concentration of chemicals and dyes in solutions.(59-64). A compact portable FOS developed for glucose level monitoring has been found to be very useful for medical applications (65). Our next attempt is to package this as a commercially viable product.

8. Changes in organisational set up

Laser related teaching and research at CUSAT were reorganised under International School of Photonics that was formed in March 1995 so as to consolidate the activities by increasing international co-operation in these ventures. Notable among these is the Dutch assistance under the MHO programme of the Government of the Netherlands. A bilateral MOU has been signed with Eindhoven University of Technology in Eindhoven, The Netherlands for strengthening the activities of the International School of Photonics. Strong academic links exist with institution in several other countries that include Sultan Kuboos University in Sultanate of Oman, Rochester Institute of Technology in New York, Fraunhofer Institute in Freiburg, Germany, to mention a few.

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