

**T a r g e t  
c h a m b e r  
f o r  
l a s e r- p l a s m a  
i n t e r a c t i o n  
s t u d i e s.**



**The new canteen building.**

### **Chamber for laser-plasma interaction studies**

The workshop has designed and fabricated a target chamber for carrying out experiments to study laser produced plasma as well as laser plasma interaction, when a multibeam high power laser irradiates a microballoon target uniformly. The high vacuum chamber with an inner diameter of 600 mm and length 630 mm is made of SS304. It has 31 ports of different sizes for mounting various diagnostics and high power lasers. Twentyeight of these ports are positioned in such a way that their axes meet at a point (within a sphere of dia 0.1mm), where the microballoon target will be positioned. The chamber is leak tight to  $1 \times 10^{-8}$  std. cc/sec (helium) and an ultimate vacuum of  $5 \times 10^{-6}$  torr has been obtained during vacuum testing.

### **Construction activity**

The Laser R & D building - Blocks 'A', 'B' & 'C' and the Accelerator Support Technology building have now

been handed over. In these buildings, a separate return air duct was eliminated and the false ceiling itself served the purpose of return air duct.

The canteen is now functional in its new building. Construction of the buildings for Library, Computer centre, and Administration has commenced. Construction of the building for Medical Centre and the extension of School building is also in progress.

The construction group has taken up construction of all the houses of 8th plan except 2 'E' type houses. During 2 years of plan period, out of 188 sanctioned houses 78 houses (18 Type 'A', 54 nos. Type 'B', 6 Type 'C') have been handed over.

CAT was also entrusted with the job of construction of one dormitory and four flatlets at TIFR, Pachmarhi. It is noteworthy that despite the site being at a long distance and high altitude, the work has been completed in scheduled time.

## **Optical Nonlinearities in Quantum-confined Systems**

Nonlinear optics deals with interaction of light with light in a medium. For example, when two or more monochromatic waves interact in an optically nonlinear medium energy can be transferred from one to the other. Although every material is in principle nonlinear, to make a device one needs materials which have relatively large nonlinearities.

In the early days of nonlinear optics, the search for new materials was mainly aimed at frequency conversion devices based on second order effects. In recent years, however, this search is increasingly driven by the need to develop photonic computing devices based on third order

nonlinear effects particularly optical bistability and optical phase conjugation by degenerate four wave mixing. In degenerate four wave mixing (DFWM) there are 3 input beams — two oppositely directed pumps and third, a probe beam incident at an angle to one of the two pump beams. The signal beam is a beam which retraces the path of the probe beam irrespective of the angle. Thus, a phase conjugate mirror acts as if every ray is incident normally on it. The DFWM process can also be seen as real time holography. The interference of one pump with the probe creates an intensity pattern — which is converted into a refractive index pattern by the intensity dependence of the

refractive index. Diffraction of the second pump beam from this pattern creates the signal beam which has the same wave front as the probe beam but travelling in opposite direction. It is thus clear that DFWM can be used to perform all the image processing traditionally performed by holography. It can also be used to implement shadow casting logic of Yatagai. Optical bistability is the situation in which optical response of a system shows hysteresis. Typically, this occurs if a nonlinear medium, e.g. a medium with intensity dependent absorption or refractive index, is introduced in a Fabry-Perot resonator. Although known since 1969, the phenomena remained a curiosity till 1979 when it was shown that optical bistability can be observed at relatively low powers ( $\sim$  few milliwatts) with the incident laser frequency close to the band gap of semiconductors.

With these observations it became apparent that an optical computer can be made using optical bistable switches. The kind of nonlinearity involved in these experiments was quite different. While the frequency conversion devices use passive nonlinearities which depend on virtual excitations and relatively large intensities these new nonlinearities involved real transitions. In an excited semiconductor, electrons fill the bottom of the conduction band and holes the top of the valence band, thus blocking virtual transitions at the band edge. It can be easily seen that this gives rise to a large optical nonlinearity. However, since the medium is really excited, such nonlinearities are rather slow i.e. the time taken by the medium to return to ground state is large — typically recombination time for the electron-hole pair. The next breakthrough came with the discovery of self electro-optic effect devices (SEED). This exploits the fact that in two dimensional quantum wells when an electric field is applied normal to the plane of the layers, the exciton energies shift without ionizing the exciton. Coupled with the fact that exciton binding energy increases by a factor of 4 if the electron and hole are both confined in a plane, this kind of nonlinearity has provided relatively fast ( $\sim$ ns) and low power optical bistable room temperature devices. Using more or less the same technology as GaAs integrated circuits, these devices and high power semiconductor lasers are the key components of the recently demonstrated all-optical computer by AT&T Bell Laboratories. This success has fuelled the expectation that lower dimensional semiconductor nanostructures will provide faster and more effective nonlinear optical devices. Particular attention has been paid to quantum dots and quantum boxes — spherical or cuboidal nanostructures of semiconductors which are expected to concentrate the total oscillator strength of the band-to-band transitions into a finite number of Lorentzian peaks.

With such applications in mind, we have investigated the electronic structure and linear and nonlinear optical

properties of semiconductor quantum dots over the last decade. The fabrication of such quantum dots and quantum boxes with good quality requires advanced semiconductor technologies not yet widely available. Like many other groups, for our experimental work we have so far used commercially available sharp cut-off colour glass filters which contain semiconductor nanoparticles usually of  $\text{Cd-S}_x\text{Se}_{1-x}$  with typical diameter of 10 nm. The first theoretical work, by Rustagi and Flytzanis, on optical nonlinearities in these composite materials was done in the background of the surprising results of Jain and Lind that a glass containing less than 1% CdS has the same nonlinearity as a CdS single crystal. Generalizing the well known Maxwell-Garnet theory of dielectric response of composite materials to the nonlinear situation, they noted the possibility of resonant enhancement of such nonlinearities and also showed that a new mechanism of nonlinearity exists due to the intensity dependence of the field penetration. If the dielectric function ( $\epsilon$ ) of a quantum dot has a resonance which saturates like that in a two level atom, Chemla and Miller showed that such a system should show optical bistability. We, however, found that a two-level model is very restrictive because the quantum dot resonances are collective resonances and depend on the real part of dielectric function. The presence of nearby resonances, even several widths away modifies the absorption spectrum as well as bistability. Generalising this still further we looked at the optical response of spherical quantum dots composed of uniaxial crystals such as Wurtzite CdS using simple models. We showed that while the linear spectrum of such a composite medium with randomly oriented spherical anisotropic crystals will show only split Fröhlich resonances, at higher intensities, intensity dependent orientational broadening of Fröhlich resonances is expected. It is important to appreciate that the essential question being addressed is the saturation or high intensity behaviour of collective excitations like plasmons and all the answers available so far are incomplete.

Trying to understand the band edge absorption spectrum in semiconductor quantum dots, we performed a variational calculation to determine the electron-hole motion in a quantum dot. Our results for CuCl nanoparticles were in agreement with experimental data and showed that the earlier expectation that in this region the energy levels correspond to quantization of the centre of mass motion was not justified. For III-V and II-VI semiconductors, however, the structure of the topmost valence band is more complicated than for CuCl, and more realistic calculations of electronic structure are needed. A typical quantum dot consists of a few thousand atoms with the atomic structure nearly the same as in bulk crystal. This system is obviously too large to adopt methods used for molecules. On the

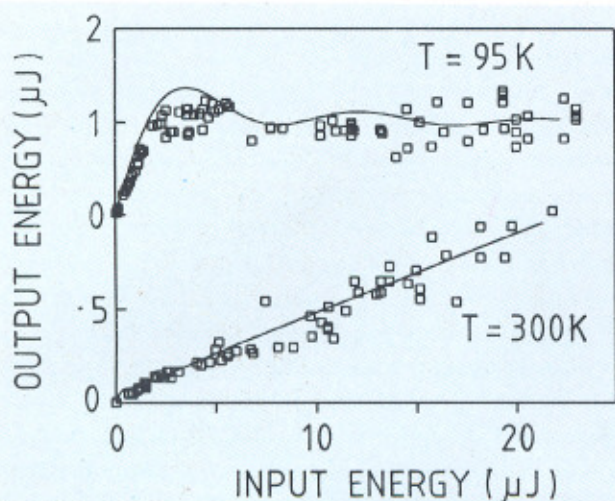


Fig. 1: Nonlinear transmission through OG530 filter at 1054 nm. Squares are experimental data and the line is a theoretical fit.

other hand because the typical de Broglie wavelengths are comparable to crystallite sizes, the simplification due to periodicity is no longer available.

We have used multiband effective mass approximation (EMA) and an effective bond orbital model (EBOM) to calculate the electronic structure of GaAs and CdSe quantum dots. By looking at wave functions in the two systems a correspondence was established between the two methods. The EMA is valid only close to the band edge while EBOM provides eigenvalues and eigenfunctions throughout the bands. Although far from complete, the calculated electronic structure was successfully used to explain the observed size dependent shift of the valence band edge as well as the structure seen in optical absorption and luminescence excitation spectra of narrow distribution quantum dots. These are two of the few examples where quantitative comparison between experimental and theoretical results has been possible. The main difficulty lies in the broad size distributions in the easily available semiconductor doped glasses (SDG) mentioned above. Already about 10 years ago it was realized that two filters made by different manufacturers can have dramatically different carrier relaxation times even though the crystallite size and linear absorption may be the same.

Therefore various nonlinear optical properties of such filters have been investigated in great detail specially in the absorbing region. Recently, however there has been some interest in the optical nonlinearities of SDG's when they are transparent to the incident laser. We have used a home made active-passive mode locked 6 ps Nd:Glass laser at 1054 nm and its second harmonic to investigate optical nonlinearities over a wide range of band gaps. We have shown that via two photon absorption one can generate a large density of electron-hole pairs in these nanoparticles.

We have also measured the intensity dependence of the refractive index by z-scan method wherein the transmis-

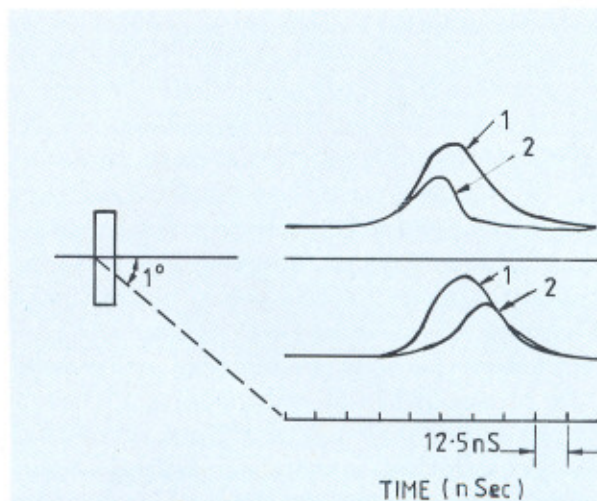


Fig. 2: Temporal profiles of laser pulse transmitted through  $C_{60}$  solution in Toluene. 1 & 2 refer to incident and transmitted pulses. Top set is on axis, and bottom is one degree off axis.

sion of a focussed laser beam through an aperture is monitored as a thin nonlinear optical sample is transported through the focal region. The nonlinear transmission changes dramatically as the laser wavelength scans the band edge region.

Since our laser source is of fixed wavelength, the scan over the band edge region was done by varying the sample temperature between liquid nitrogen temperature and room temperature (Fig. 1). The new data shows that transparent region of these samples may provide the more exciting nonlinearities although the nature of nonlinearity is not yet very clear.

Recently fullerenes have provided a novel class of quantum confined systems. Since the optical absorption spectrum of all known fullerenes is dominated by plasmon like peaks at  $\sim 6\text{eV}$  and  $\sim 18\text{eV}$ , we believe, it is appropriate to look at these molecules as quantum dots in which the  $\pi$ -electrons are confined in a rather thick shell while the  $\sigma$ -electrons are confined in a much narrower shell. We have developed an empirical pseudopotential model for the  $\pi$ -electrons in  $C_{60}$ . We have used this and a tight binding model to calculate linear and nonlinear optical response of several fullerenes  $C_{60}$ ,  $C_{70}$ ,  $C_{76}$ ,  $C_{59B}$  and  $C_{59N}$ .

We predict that less symmetric fullerenes like  $C_{59B}$ ,  $C_{59N}$ ,  $C_{70}$  and  $C_{76}$  will have much larger nonlinearity.  $C_{76}$  is particularly interesting because it is a chiral molecule and has substantial  $\beta$ , the lowest hyperpolarizability. Independent of these passive nonlinearities, another kind of nonlinear optical response of  $C_{60}$  has attracted attention. Last year Tutt and Kost observed that  $C_{60}$  solution in toluene shows a very strong optical limiting behaviour i.e. the transmission at low fluences is large and intensity inde-

pendent while at higher fluences the transmitted energy stays constant independent of the incident fluence. Using the second harmonic of a home-made Q-switched Nd:Glass laser oscillator-amplifier system we have investigated this phenomena in considerable detail. Angle and time resolved studies on various solutions of C<sub>60</sub> and C<sub>70</sub> by our group have shown that the thermally induced nonlinear scattering plays an important role (Fig. 2). One of the attractions of fullerenes as nonlinear material is that unlike other organics they do not have hydrogen and hence are expected to have relatively low residual absorption coefficient due to multiphoton absorption. Using a relatively large (1mm x 3mm) single crystal of C<sub>60</sub> grown at Anna University we found that the crystal had a linear absorption coefficient of  $\sim 7\text{cm}^{-1}$  compared to  $6\text{cm}^{-1}$  measured for thin films, a two-photon absorption coefficient of  $18\text{cm}^2/\text{GW}$  and a surface damage threshold  $\sim 40\text{GW}/\text{cm}^2$ . All these measurements were performed at CAT with a 6 ps laser at 1054 nm.

As mentioned above, the experimental facilities in the nonlinear optics group at CAT are centred around two home-made Nd:glass lasers — one Q-switched to give 40

ns pulses and the other active-passive mode locked to give 6 ps pulses. We also have a home made Ar-ion laser for Raman Spectroscopy. With the Nd:glass lasers, we can characterise nonlinear optical materials for frequency conversion at various stages — by electric field induced second harmonic generation at the molecular level, by powder second harmonic generation at powder level and finally by measurement of second harmonic efficiency when a cut and polished single crystal is available. These facilities are now being augmented with three new lasers. These are a Nd:YAG Q-switched laser obtained from the Production Unit of CAT, and two commercial lasers — a sub-picosecond Ti-Sapphire laser and an Ar-ion laser pumped CW dye laser.

To summarize, we have been investigating nonlinear optical response of a variety of quantum confined systems. With the availability of high repetition rate lasers in the near future, we expect to contribute significantly to the development of nonlinear optical materials and devices.

K C Rustagi

## CONFERENCES / WORKSHOPS

### National Laser Symposium

The annual National Laser Symposium was held at CAT this year from January 29 to February 1, 1994.

The large number of contributed research papers (totalling 158) covered almost all facets of Laser Science and Technology, and were of very high quality. Apart from these papers, there were eight "Thesis presentations" by young research scientists. A Proceedings for the symposium has been published in which these papers and the theses are presented in an extended abstract form.

The Symposium had twelve invited talks on topical subjects presented by leading laser scientists. Most of the research papers were covered in poster sessions, except the session "Laser Applications to Biology and Medicine". This was a special session and was well attended by Doctors and Researchers active in this upcoming field in India.

Another interesting feature of the Symposium was the exhibition. About twenty companies exhibited a large range of products from lasers to optical components and various testing and measuring instruments. A heartening feature was that many companies are now able to supply indigenously built He-Ne Lasers, optical components and optical mounts.

Apart from these activities, there was a session for visit to the laboratories at CAT and one evening devoted to General Body Meeting of the Indian Laser Association.

### National Workshop on Recent Advances in Quantum Optics, and National School on Modern Optics

A National Workshop on Recent Advances in Quantum Optics (RAQUO-94) was organised at CAT during March 7 - 10, 1994. It was attended by about seventyfive scientists. The Workshop covered a wide range of topics in quantum optics and related areas. There were about twenty invited talks, and about the same number of contributed papers. Amongst the invited speakers were Dr P D Gupta and KS Bindra from CAT. An open discussion on "Outlook of experiments in quantum optics in India" was held during the Workshop.

Preceding the Workshop, a "National School on Modern Optics", mainly targeted at post-graduate students, was held during March 3 - 5, 1994. The purpose of the School was to familiarise students with various advances in the field of modern optics, so that they could gain maximum out of RAQUO-94. The school had lectures on the following topics: Introduction to quantum optics, nonlinear optics, QED in cavities, and quantum chaos.