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Sub-Picosecond Optical Pulses

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The generation of ultrashort light pulses in mode locked C.W. dye laser systems has made possible a wide range of new experirnents in which the interaction of radiation with matter is studied. Accurate measurements ot' the optical pulse width are essential for a quantitative interpretatiori of the experimental results. A discussion ofultrashort pulse generation and measurements is presented, together with a section on dynamic spectroscopy.

A geração de pulsos luminosos ultracurtos em *Mode Locked* C. W *dye Zasers* tornou possível uma nova variedade de experimentos onde se estuda a interação de radiação com a matéria. Para interpretar quantitativamente os resultados experimentais, torna-se necessária uma medida precisa da largura dos pulsos luminosos. A geração e técnicas de medida de pulsos ultracurtos são apresentadas, juntamente com uma secção que trata de espectroscopia dinâmica.

1. INTRODUCTION

Scientific developments are almost always related to advances in measurement techniques. The recent developments in time measiirements, in the pisosecond range[1a], will certainly lead to a better understanding of non-equilibrium processes in Physics, Chemistry and Biology. This developments tias been made possible by the availability of ultrashort pulses of light from mode locked lasers. It is possible now to investigate, on the picosecond time scale, the interaction of light pulses with molecules, atoms and solids.

From an engineering point of view, ultrashort pulses offer possibilities for pulse-code modulation at very high rate and multichan-

nel communication systems. Picosecond light pulses can also be converted into electrical pulses which are much shorter in duration (typically 20 ps of duration) than those generated by conventional electronic techniques.[1b]

Phase dependent phenomena in the propagation of light pulses through matter, such as coherent photon response, self-induced transparency and photon echoes can be also studied with these pulses. More recently picosecond pulses have been used to monitor plasma compression studies.

For the quantitative interpretation of the results of experiments using ultrashort pulse techniques, an accurate measurement of the duration of the exciting pulse is essential. This paper will concentrate on general principles and simple physical pictures, leaving the detailed model and calculation to be provided by the references.

The paper is organized in the following way: Section 2 discusses modes in optical resonators and mode locking of dye lasers. Section 3 treats pulse measurements techniques. Section 4 describes in detail the experimental arrangement used. Finally, section 5 presents applications to the various fields. This paper is intended as a tutorial and a review paper in ultrashort pulse generation in C.W. dye laser measurement techniques and applications. It is intended to be of use as an introduction to the subject for the general reader as well as an overview for the specialist in the field.

2. MODE LOCKING

A typical laser consists of an optical resonator formed by mirrors, and some laser gain medium within the resonator. The frequency band $\Delta\nu$ of the possible laser oscillation is determined by the gain medium, while the properties of the resonator define more precisely the laser frequency. Usually, there are many modes of the optical resonator which fall within this oscillation band, and the laser output consists of radiation at a number of periodically spaced frequencies. The modes of an open resonator formed by a pair of coaxial mirrors may have different field distribution normal to the resonator axis (transverse modes). For

each of these transverse modes there is an infinite set of modes which have different axial distributions corresponding to different numbers of half wavelengths of ligh along the resonator axis. These longitudinal modes are spaced in frequency by $c/2$ L, where c is the velocity of light and L is the optical path length between the mirrors. Usually it is possible to discriminate against all transverse modes except the lowest order, by aperturing down the resonator.

For this lowest-order transverse mode, the energy distribution in a plane transverse to the resonator axes, has a simple Gaussian profile. For a long enough resonator ($L >> c/2\Delta\nu$) and no highly selective frequency discrimination, a typical laser output spectrum is illustrated in Fig. Ic.

Each mode is largely uncorrelated with its neighbors and the thermal pattern presents the features of a random distribution even though the average power remains relatively constant.

If, somehow the relative phase an the amplitude ofthese modes is fixed, this uniformity in the energy distribution is destroyed and it is possible to concentrate the enerygy in short pulses within the laser cavity. This process is called mode locking, and it can produce an amplitude output consisting of a regularly spaced train of pulses[2]. It is also possible to obtain a constant amplitude frequency - modulated output signal. This process is called mode locking, because in the frequency domain it corresponds to correlating the spectral amplitudes and phases. When the correlation of the modes is complete, the energy is confined to a single pulse with a bandwidth Δt which is approximately equal tothe reciprocal of the total mode locked bandwidth $\Delta\nu$ (fig.1d).

Such a mode locked laser output is shown in Fig.1. Mode locking is achieved experimentally by placing, inside the laser cavity, either an externally driven modulator or a passive device which exhibits saturable absorption. The mechanism of pulse generation depends on the dynamics of the particular type of laser. For the solid state laser (Nd: glass and ruby, typical pulse width $\geq$ 10 ps) the growth of the laser pulse relies on the discrirnination by the saturable absorber and the pulse width is essentially determined by the saturable absorber recovery time.

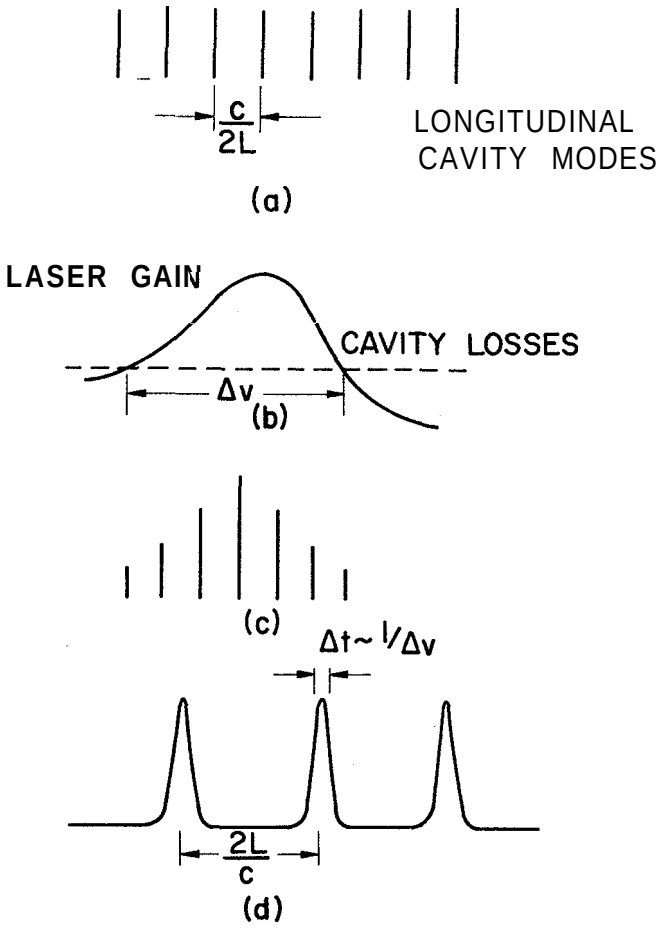

Fig.1a. - Longitudinal modes of a laser resonator operating on a single transverse mode. 1b. Lascr gain versus frequency, for a region where the gain exceeds the cavity losses. 1c. Laser output in the frequency domain 1d. - Temporal output of the mode locked laser.

In dye lasers, however, ultrashort pulses are generated by the combined action of amplifier and absorber saturation[3]. In this system the pulse width is not limited by the recovery time of the saturable absorber. An explanation of this factwaspresented in a recent paper in which the relaxation times of both the active and passive dyes were included[4]. The shaperning of the leading edge occurs because the absorption cross section of the passive dye is larger than the emission cross section of the active dye. The pulse experiences non-linear gain in the amplifier,

non-linear loss in the saturable absorber and linear cavity losses, which results in a net gain region onlu in a small part of the round trip cavity time[5]. The compression of trailing edge is due to the saturation of the gain medium.

A new scheme for obtaining ultrashort pulses was recently proposed[6]. Assume that the saturable absorber is a laser dye. Due to its absorption at the lasing frequency, there is a population inversion at the trailing edge of the laser pulse. The resulting stimulated emission leads to the formation of a second pulse. This pulse causes a rapid restoration of the losses for the first pulse, effectively shortening the recovery lifetime, resulting in a much shorter primary pulse followed closely in time by an additional ultrashort pulse at a longerwavelength. This technique can be useful in a variety of experiments.

Another approach is mode locking dye lasers by synchronous pumping, which consists of pumping a dye laser with an actively mode locked argon laser with matched cavity lengths[7]. The advantage of this scheme is that the dye laser can be tuned anywhere within its wavelenght range, This allows generation of pulses at wavelengths where passively mode locked systems are not available.

A disadvantage of this scheme is that the typical generated pulses are 10 to 50 ps, much longer than those produced within saturable absorber mode locking.

3. PULSE MEASUREMENT TECHNIQUES

Before and picosecond pulse system can be used, the pulses must be characterized a accurately. The simplest method of recording the temporal iritensity profile of a light pulse is provided by a combination of photodiode and oscilloscope. Photodiodes with rise time of the order of 100 ps are available commercially and the fastest real time oscilloscopes have band-widths of 5 GHz. Thus direct pulse measurement down to a resolution of around 100 picosencond can be achieved. However a time resolution as short as 1 picosencod is required for a precise characterization of the light pulses.

Recent developments in ultrafast streak camera technology enable the intensity $I(t)$ to be recorded directly on a picosecond time scale, with a time resolution of ~ 5 picosecond[8], which may be adequate in some cases.

A common experimental method for studying the temporal structure of picosecond pulses involves the recording of the second-order correlation function of the intensity $I(t)$. The most commonly used methods for obtaining such second-order correlation functions are second harmonic generation (SHG) in a non linear crystal and two photon fluorescence (TPF) in a material which is transparent to the fundamental laser radiation but can absorb by the two-photon process and then fluoresce. In the SHG method a Michelson interferometer, shown in Fig. 2, divides the pulse into two beams which then travel colinearly in the nonlinear crystal with a adjustable time delay between them. The second harmonic output intensity provides information about $I(t)$ through the second-order correlation function

$$I_{SHG}(\tau) = \int_{-\infty}^{+\infty} I(t+\tau)\, I(t)\, dt$$

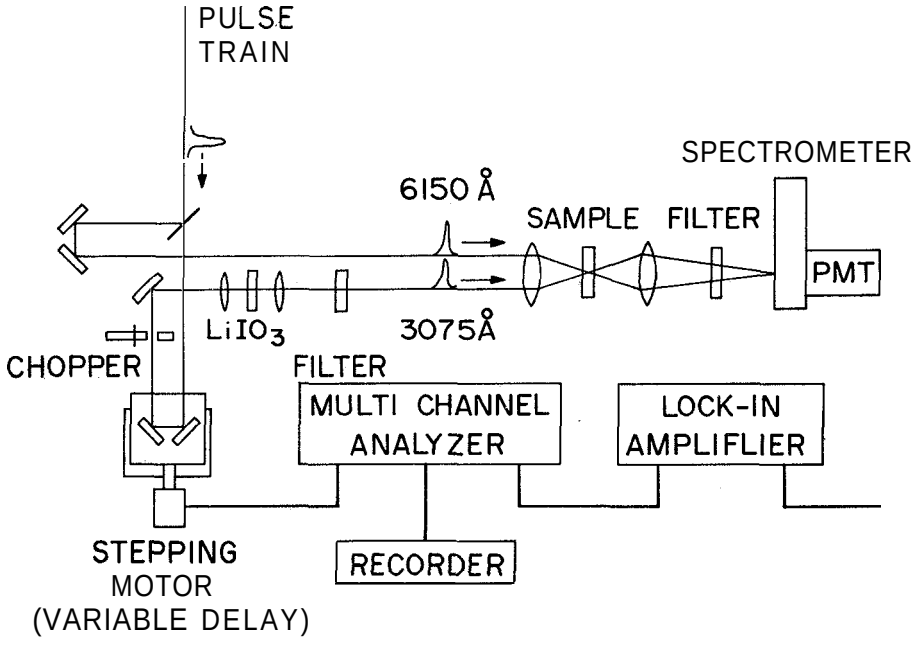

Fig. 2 - Experimental arrangement for the measurement of pulse autocorrelation functions.

Resolutions of .1 picosecond are achieved by this method[9]. In a TPF measurement the laser pulse is divided into two components which travel opposite directions through the TPF medium. Fluorescence is observed and, generally, photographed from the side of the medium. Because TPF is anon linear prooess the most intense fluorescence occurs from theregion where the two pulses overlap. The fluorescence, along the direction of propagation is proportional to the second order correlation function.

These are the most common techniques for pulse measurement however there is a number of others which are described in details in ref. 8.

4. EXPERIMENTAL ARRANGEMENT

The passively mode locked C.W. dye laser used in our experiments in shown schematically in Fig.3 (Ref.9). This is the only subpicosecond system to our knoledge that has been applied systematically with success to studies of ultrafast processes. The gain medium Rhodamine 6G in a free-flowing stream of ethylene glycol is excited with a C.W. argon laser. The second free-flowing stream of ethylene glycol contains the saturable absorber dyes for mode-locking DODCI, one of the saturable absorber dyes produces short pulses an operation which occurs near laser thre threshold and requires careful adjustment. Addition of the dye malachite green to the DODCI solution permits short pulse operation well above threshold. The best pulses are obtained near λ = 6510 Å.

There is a cavity dumping arrangement at one end of the resonator. The set up offers several advantages over taking the pulse output from a partially transmitting end mirror. Higher peak powers can be obtained with an adjustable repetition rate. The energy per pulse is about 5×10^{-9} joule and the repetition rate can exceed 10^5 hertz. The pulse measurements are made by a correlation technique using phase-matched sum frequency generation in KDP; the arrangement is shown in Fig. 2. The beam from the dye laser is split in a modified Michelson interferometer arrangement. The two beams travel different paths and the time delay ($\Delta t = \Delta \ell / c$) between the two is varied with a stepping motor and translation stage in one arm of the interferometer. The stepping motor drive also increments a multichannel analyzer which allows accumulation and signal averagingover

a time period from minutes to hours. The two beams are then focused into a .1 mm thick crystal of KDP.

Sum frequency generation, proportional only to the product of the two beam intensities, is detected at an angle bisecting the angle between the two fundamental beams, which allows a background-free measurement of the correlaction function (Fig. 3). A typical autocorrelation trace of the pulses in shown in Fig. 4. The important point to note is

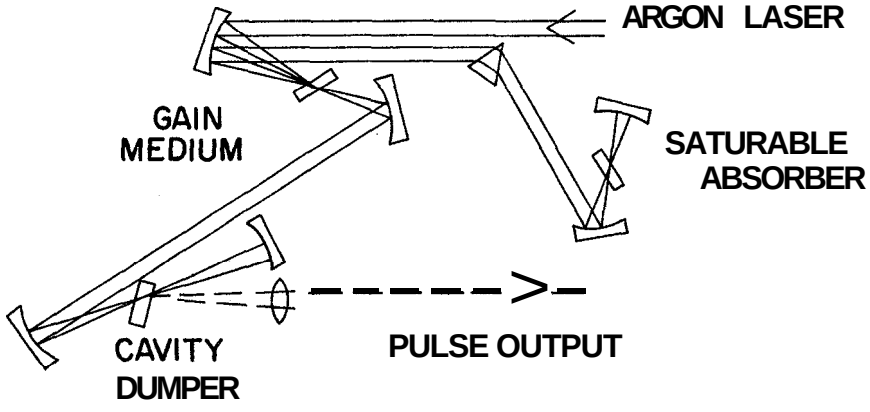

Fig. 3 - Passively mode locked C.W. dye laser diagram.

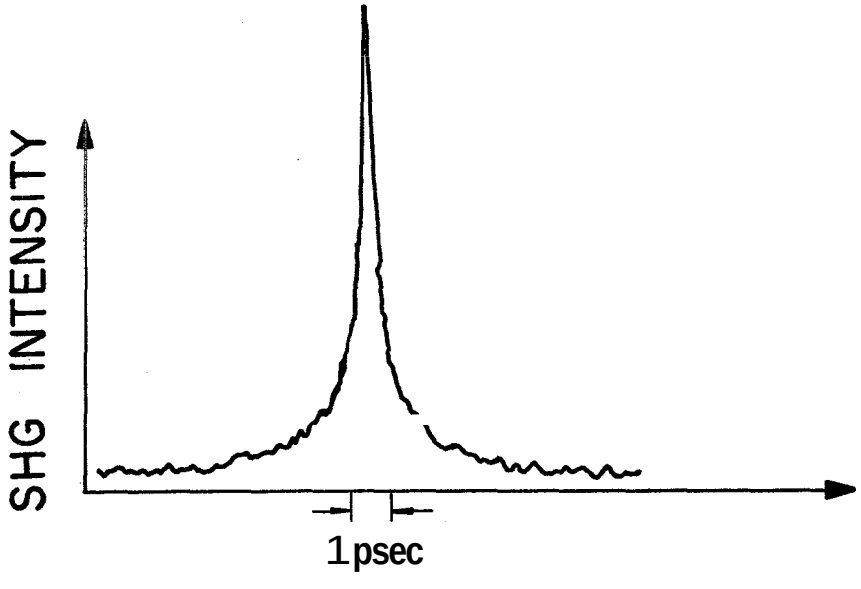

Fig. 4 - Autocorrelation measurement of the laser output pulses.

the reproducibility of the subpicosecond pulses which, combined with the high repetition rate, makes possible their application to the study of ultrafast processes.

The pulses generated by the laser are thus well characterized and the application of this system to ultrafast time resolved processes is now possible.

5. APPLICATIONS

A common scheme used is pump-probe configuration where the detector monitors directly the change in transmission induced by the pump beam. This is shown in Fig. 5. The visible optical pulse train is split in a modified Michelson interferometer into a purnp and probe beam. The pump beam passes through the lithium iodade crystal and second harmonic is generated with an approximately 15% efficiency. The modified Michelson interferometer also provides a variable delay between them. The ge-

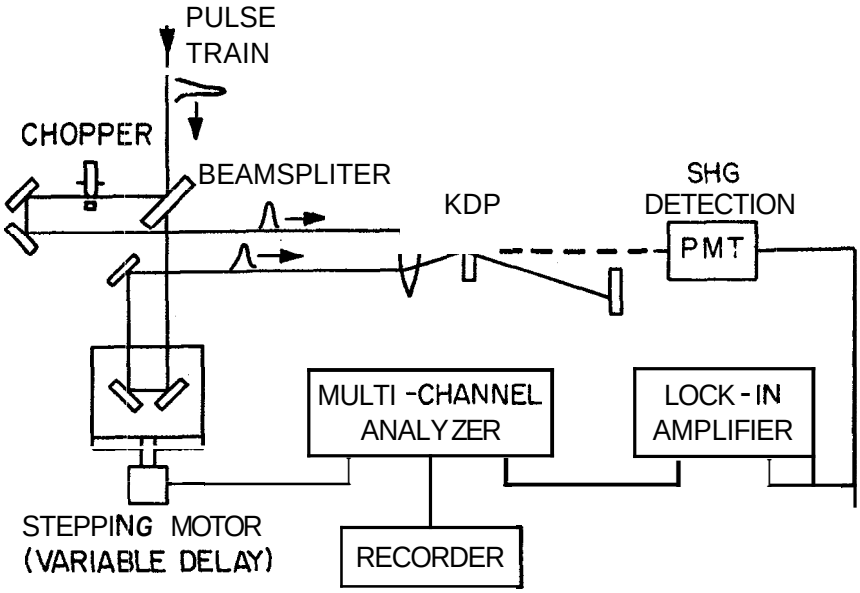

Fig. 5 - Experimental arrangement for the measurement of time resolved absorption or reflection.

nerated beam is combined with the weaker visible beam by means of a dichroic beam splitter. The ultraviolet beam was chopped and modulation on the visible beam was detected using a photomultiplierandphase-locked detection. To accurately determine the position of zero time delay we use a solution of the molecule cresyl violet which exhibits a strong ground state absorption for both ω_2 = 6150 Å and ω_1 = 3075 Å. When the ultraviolet pulse at ω_1 removes a molecule from the ground state S_0 an absorption decrease is immediately sensed by the weaker pulse at ω_2. This arrangement can be utilized to investigate radiationless relaxation of large orfanic molecules in solution.

The measurement of the characteristic time in which thermal equilibrium is established within the manifoid or vibrational levels of an excited electronic state is performed for Rhodamine B and Rhodamine 6G (Ref. 10). The pumping and probing intensities are sufficiently low that the induced population S_1 remains small and decays with its normal fluorescence lifetime in the manosecond range.

A short wavelength optical pulse ω_1, at 3075 Å excites the molecule from its ground electronic state S_0 to a highly excited electronic and vibrational level.

The molecule relaxes to the bottom of the S_1 state, which is probed by measuring optical gain for a second pulse at ω_2. The delay between the rise of optical gain at frequency ω_2 and the exciting pulse at ω_1 gives a measurement of the relaxation time.

In Fig. 6 we plot the experimentally measured response for cresyl violet on the top of gain response curves Rhodamine B in both methanol and glycerol. We can say that these curves all agree with each other within 2×10^{-13} seconds indicating that any delay in the gain risetime is less than 2×10^{-13} seconds. Relaxation on this rapid time scale suggests that the thermalization process is intramolecular and does not require an interaction with the solvent.

In an additional experiment the dynamics of the intramolecular motion os stilbene is investigated.

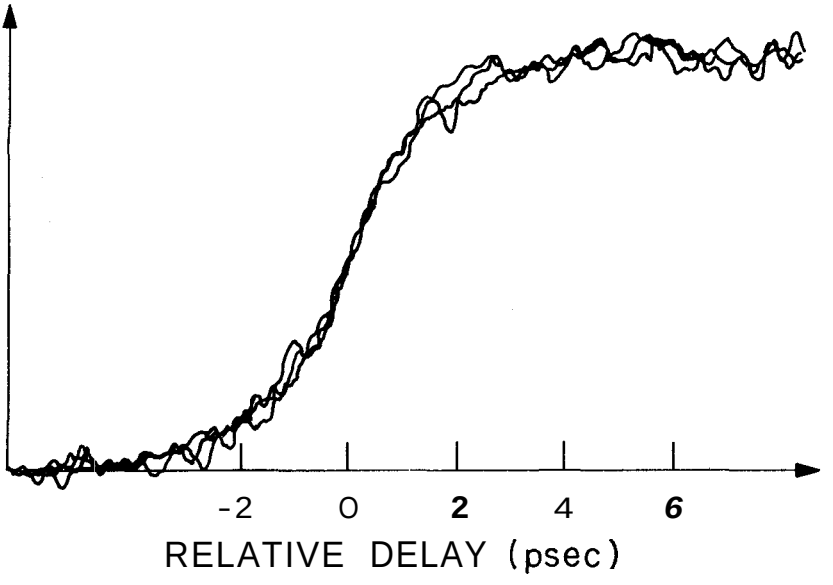

Fig. 6 - Subpicosecond rise of gain for solutions rhodamine B methanol and rliodamine 6G rnethanol compared with an instantaneous response reference.

Photoisomerization of the stilbene molecule is knows to occur in both the singlet and triplet states. Changes in the viscosity of the solvent make large differences in quantum yields of fluorescence, suggesting important geometry changes, which take place in the excited state. A direct observation of the excited state process is possible, by monitoring the: time dependence of the excited state absorption in the picosecond range (Fig.7) .

The intermolecular rotation lifetime measured was 52 picoseconds for trans-stilbene in hexane, while excited cis-stilbene has a lifetime of 7 picoseconds (Fig. 7)[11]. It also was shown that this lifetime is not viscosity-dependent, for low-viscosity solvent ($\eta \leq 2$ centipoises). These data combined with fluorescence quantum yield measurements provide information leading to a complete understanding of inter-convention among the stilbene excited singlet state isomers.

Ultrashort optical pulses offer a unique opportunity to study the dynamics of nonequilibrium carriers in semiconductors. The subpicosecond optical pulses allow us for the first time to directly time-re-

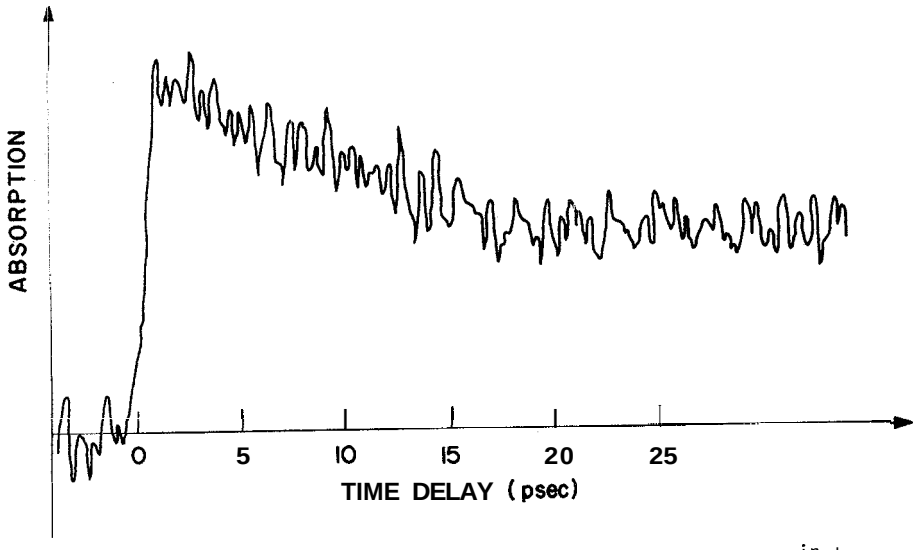

Fig. 7 - Time resolved excited state absorption for cis-stilbene in hexane.

solve interband energy relaxation processes in a semiconductor. Subpicosecond optical pulses (h = 3075 Å) will excite hot carriers, and a second probing pulse (h = 6150 Å) with a variable time delay will time-resolve the reflectivity change.

The energy of the excitation pulse is gr-eater than the bandgap and initially the mean kinetic energy of the carriers will exceed the thermal energy of the lattice. The carriers will then relax to the band extrema by various mechanisms until they are in thermal equilibrium with the lattice-, after which they will recombine.

Figure (8) shows the time evolution of the incremental change in reflectivity of GaAs (Ref.12). These results can be interpreted by considering the effect of the nonequilibrium carriers on the real and imaginary parts of the optical dielectric function $\varepsilon(\omega)$. For short time delays, the carrier distribution is very hot, various high energy conduction band states will be occupied and corresponding valence band states will be vacant. Consequently the imaginary part of $\varepsilon(\omega)$ will be reduced at high frequencies compared to the probe. The real part of $\varepsilon(\omega)$ at the probing frequency is reduced and consequently the reflectivity will de-

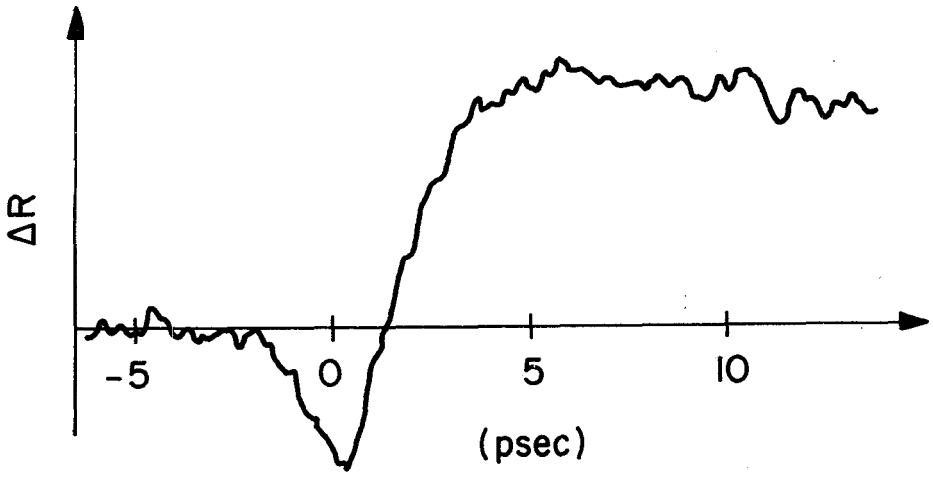

Fig. 8 - Transient reflectivity from carriers in GaAs after excitation at 4 ev with a 2 ev probing wavelength.

crease. When the carrier distribution has relaxed to the band edge, the imaginary part of $\varepsilon(\omega)$ will be decreased at frequencies near the band edge and hence the real part of $\varepsilon(\omega)$ will increase at the probe frequency causing the reflectivity to increase. The zero crossing timeis a measure of ths rate of energy loss of the hot electron distribution. The estimated ensrgy loss rates are 0.4 ev/picosecond for electrons and holes having mean energy the range of 1 to 2 ev.

Recently ultrashort optical pulses have been applied to the study of photolysis of hemoglobin. The fundamental unsolved problem of hemoglobin is the mechanism of its cooperative binding of oxygen. The three systems which have been investigated are hemoglobin, and twoof its complexes oxyhemoglobin and carboxyhemoglobin. The molecules were excited with picosecond light pulses and the rapid relaxation and photodissociation were observed.

The result showed that the rise of the induced absorption appears in less than 0.5 picosecond, for carboxyhemoglobin and has a slow time decay. For oxyhemoglobin there is a rapid increase in the induced absorption but in this case the response decays rapidly with a time constant of about 2.5 picoseconds. A discussion of the experimental details and results may be found in Ref. 13.

CONCLUDING REMARKS

We have attempted to present a tutorial review of the subject of picosecond pulse generation and applications in the various fields so that the interested reader will realise the potential of such light pulses, which provide the highest temporal resolution for investigating the optical properties of matter.

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