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NEW EXPERIMENTS IN FEMTOSECOND CONDENSED MATTER SPECTROSCOPY

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ABSTRACT

Recent advances have taken place in optical pulse generation techniques pushing optical pulsewidths down into the femtosecond domain. Optical pulses with a duration as short as 30 femtoseconds have recently been reported[1]. In this discussion we will describe these new advances and show how these techniques are giving new insights in the very fast processes in condensed matter physics.

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I. Colliding Pulse Modelocked Dye Laser

The development of the colliding pulse modelocked (CPM) dye laser in 1981 is the milestone on the way toward subpicosecond light pulses[2]. Figure 1a shows the ring configuration of the laser cavity which contains two elements : an optically pumped saturable gain medium (Rhd 6G) and a saturable absorber (DODCI). The mechanism for the optical pulse shaping is based on the preferential absorption of the leading edge of the pulse by the saturable absorber and preferential amplification on the leading edge of the optical pulse by the gain dye. This effectively shortens the pulse with each pass around the cavity until the limiting pulsewidth is achieved. The basic limitation is group velocity dispersion inside the cavity.

The additional mechanism operative in the colliding pulse configuration is the interaction of the two counterpropagating pulse streams present in the ring laser cavity. These two pulse streams "collide" in the saturable absorber. Synchronization of the two oppositely directed pulse streams occurs because the cavity losses are minimum when the pulses are superimposed in the saturable absorber. The pulses interfere and set up a standing-wave pattern in the absorber jet which further minimizes the losses.

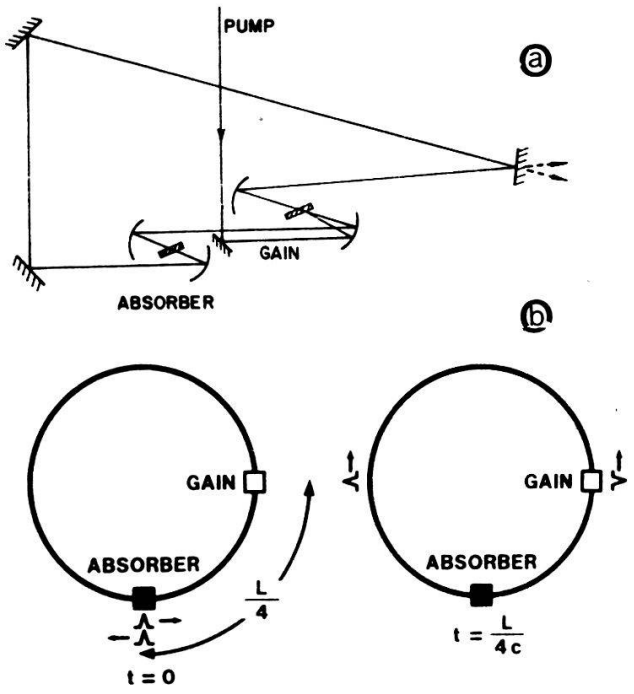

Figure 1 a) Colliding pulse modelocked laser resonator configuration.
b) Schematic of pulse timing in the cavity.

The shortest optical pulses are produced with a thin (10μm) saturable absorber medium which confines the standing wave field. The separation between the gain and saturable absorption media is chosen to be 1/4 of the total cavity length, which allows identical amplification for the two counterpropagating pulses (figure 1b). The amazing property of that laser configuration is to produce very short pulses using dyes whose characteristic recovery times are several hundreds of picoseconds.

Figure 2 shows the experimentally measured autocorrelation function of the produced pulses, using second harmonic generation (SHG) in a thin (100μm) crystal of KDP. The optical pulsewidth deduced from the autocorrelation function is 65 femtoseconds.

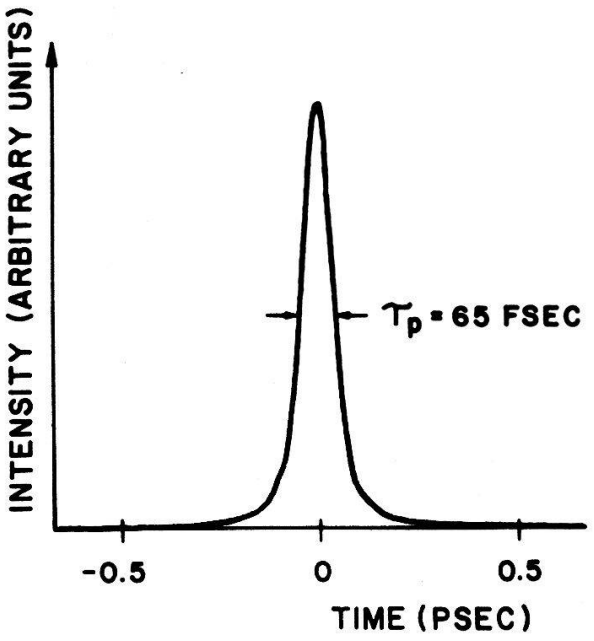

Figure 2 Autocorrelation function of the pulse from the colliding pulse modelocked dye laser. The full width at half maximum corresponds to a pulse duration of 65 fs.

II. Pulse Compression Techniques

More than a decade ago Gires and Tournois[3] and Giordmaine et al[4] proposed shortening of optical pulses using compression techniques analogous to those used at microwave frequencies. The basic idea is to create new frequencies in such a way that the pulse has a sweep in frequency and then to reassemble the pulse through a dispersive delay line.

When an intense optical pulse is passed through a nonlinear medium the refractive index is modified by the electric field[5] :

$$n = n_0 + n_2 \langle E^2 \rangle + \ldots \quad (1)$$

where n_2 is the nonlinear index of the material. The pulse then experiences a phase change :

$$\delta\psi = n_2 \langle E^2 \rangle \omega_0 \ell / c \quad (2)$$

where ω_0 is the central frequency and ℓ the distance traveled in the medium. The instantaneous frequency shift is

$$\delta\omega(t) = -(\omega_0 \ell n_2 / c) \frac{\partial}{\partial t} \langle E^2(t) \rangle \quad (3)$$

The frequency of the outcoming pulse varies almost linearly in time from the shorter frequencies to the high frequencies. One problem is to avoid any other nonlinear process which would destroy this linear chirp. Nakatsuka and Grischkowsky[6] showed it can be overcome by using a single mode optical fiber as a nonlinear medium.

The linear dispersive delay line needed to recompress the pulse has been designed by Treacy[7] as a simple parallel pair of gratings. Each wavelengh passing through the pair is diffracted at a different angle by the first grating and recollimated by the second grating. The end result is a wavelength dependent optical path.

Figure 3 shows the set-up for optical pulse compression which allowed the formation of 30 femtoseconds duration pulses[2]. Optical pulses of 90 femtoseconds duration were focused into a 15 cm long polarization preserving single mode optical fiber. With about 0.5μJ energy/pulse coupled into the fiber, the frequency spectrum was broadened by about a factor of 3 (from 60Å to 200Å). Optical compression was achieved by the grating pair.

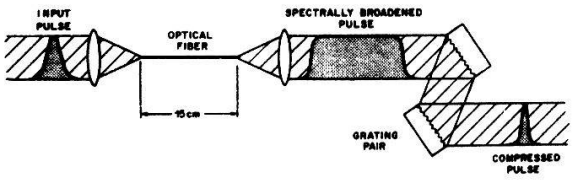

Figure 3 Experimental arrangement for compressing optical pulses.

III. Ultrashort Optical Pulses Amplification

Advances have also taken place in the amplification of ultrashort optical pulses. Using a four stage dye amplifier pumped by a frequency doubled Nd : YAG (10 Hz rep. rate) laser, 70 femtosecond optical pulses have been amplified to peak pulse powers of several gigawatts while retaining, or in some cases, slightly reducing the duration[8].

Short duration and high power of the amplified femtosecond pulses introduce new problems : group velocity dispersion in the dye solvent and amplifier optics causes significant temporal broadening ; the light pulse intensity causes nonlinear generation of new frequeny components which destroys the linear frequency sweep in the pulse spectrum.

The nonlinear effects are limited by the use of a multistage amplifier (figure 4) ; a careful choice of dyes and gain levels in the different stages and by the use of saturable absorbers after each stage, to avoid preferential amplification of the leading edge of the pulse. The use of saturable

absorber jets also provides isolation of the different stages. The output pulses are then only linearly chirped by the group velocity dispersion and need only to be compressed down to their original duration by a grating pair compressor.

That kind of amplificator currently produces 70 femtoseconds optical pulses in the gigawatt power regime with 10 Hz repetition rate.

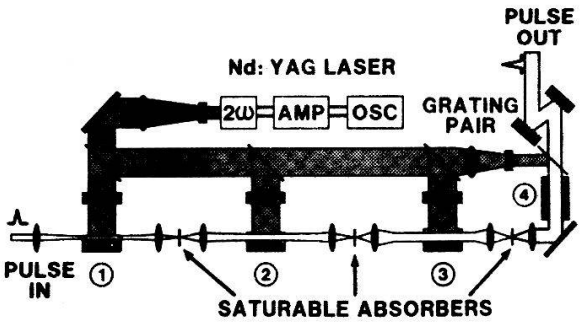

Figure 4 Schematic diagram of four stage amplifier and grating compressor.

IV. White Light Generation

The development of powerful femtoseconds optical pulses has enabled the efficient production of white light continua extending from 0.19μm to 1.6μm and with time sweeps as small as 10 fs/100nm. This is done by focusing gigawatt amplified 80 fs duration pulses at 620 nm into a thin (500μm) jet of ethylene glycol acting as a nonlinear medium[9].

Various nonlinear processes are expected to be responsible for the wide extension of the continuum, but due to the steep time variation of the exciting pulse, the time derivative term, in expression (3), is important. Therefore, self-phase modulation is a sizable effect in the femtosecond regime, even with very small nonlinear coefficients ($n_2 \sim 10^{-13}$ e.s.u) and thin media.

This phase modulation effect has been evidenced through a pump-probe experiment in H_2O in which the weak probe experiences the index change (1) induced by the strong pump[10] (figure 5). A record of the frequency shift of the probe versus the time delay between the pump and the probe is shown on figure 6. This time dependence is proportional to the negative of the time derivative of the optical pulse envelope (see expression (3)). It is remarkable that on a time scale on the order of 150 fs it is not possible to resolve any decay time. This is a good indication to the fact that electronic hyperpolarizability is a really very fast process as deduced from frequency domain measurements[11].

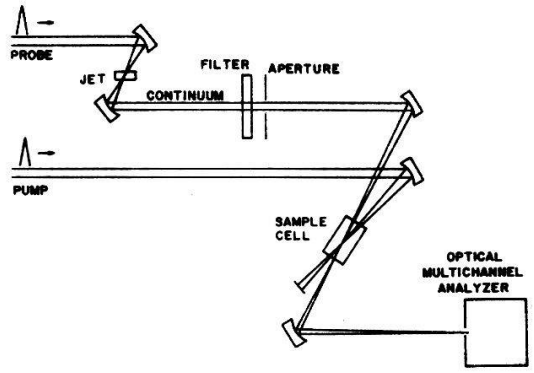

Figure 5 Experimental arrangement for measuring optically induced pulse variation in water.

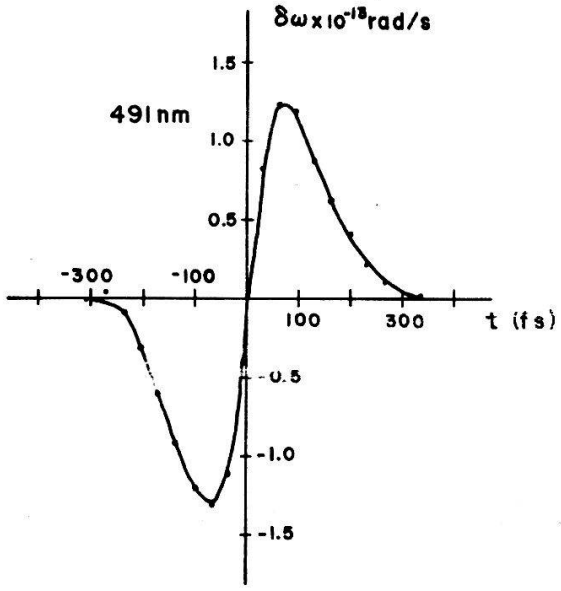

Figure 6 Relative frequency shift versus time delay experienced by a 491 nm probe in water during excitation by the amplified pulse.

Self-phase modulation should act as a seed for the various four-wave mixing processes involved in the continuum generation. Unlike in the picosecond regime[12] these nonlinear processes do not need to start from the quantum noise and should therefore be more efficient, leading to the very broad continua observed.

V. Femtosecond Nonlinear Optical Properties in CS_2

With fully one order of magnitude increase in time resolution over most previous picosecond studies, qualitatively new results bearing on ultrafast relaxation mechanisms in matter are expected. As a matter of fact, several groups[13,14,15] have made measurement of transient refractive index changes in CS_2 revealing a novel subpicosecond response time.

In their original experiment T.M. Halbout and C.L. Tang measured and time resolved the phase shift induced by a strong pump beam on a weak beam which probes an actively stabilized white light Mach-Zender interferometer where a CS_2 sample cell has been placed in one of the arms. The two beams were derived from the two out puts of a CPM dye laser with 70 fs duration pulses.

This experiment clearly showed two distinct exponential decay times, with time constants 330 fs and 2 ps. The"long" 2 ps relaxation time is in good agreement with previous estimations of the rotational motion relaxation in CS_2. The detailed nature of the fast contribution is not yet certain. This

contribution, however, shows that the predominant contribution to the third order nonlinearity in CS_2 is of nuclear motion origin down to the subpicosecond regime.

By this time the presence of femtosecond relaxation times in CS_2 has been confirmed by other groups[14,15] using optically induced Kerr effect techniques (figure 7).

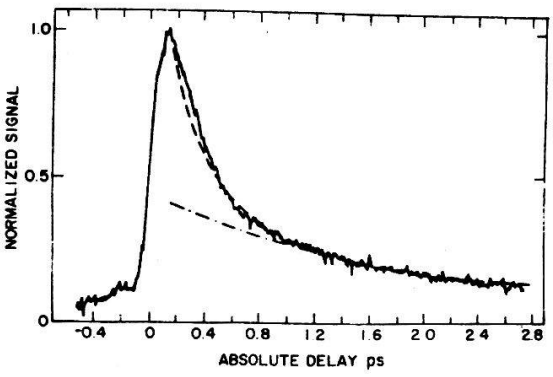

Figure 7 Measured relaxation of induced birefringence in CS_2 showing a 240 fs decay time (reference 14).

VI. Plasma Response of Optically Excited Silicon

The development of femtosecond white light generators opens the way to time resolved optical spectroscopy with a spectral resolution of the order of 7 nm.

We recently performed an experiment in which we measured the reflectivity change of crystalline silicon following the optical creation of high density electron plasma[16]. In this experiment a 90 fs pulse from an amplified CPM dye laser is split into two beams. One of the beams acts as a pump exciting the sample, the other one goes through a heavy water cell generating white light pulses. The two beams are both focused onto the silicon sample and the change in reflectivity at selected wavelengths is measured as a function of the variable delay time between the two beams (figure 8).

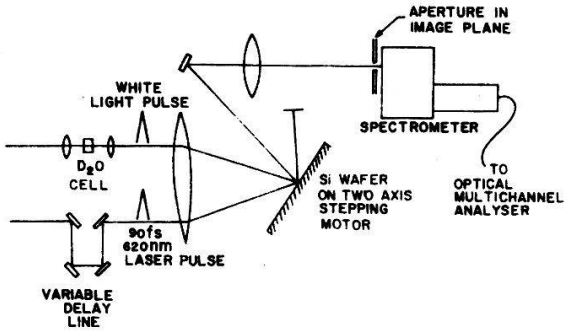

Figure 8 Experimental set-up used in the measure of time resolved reflectivity of optically excited silicon.

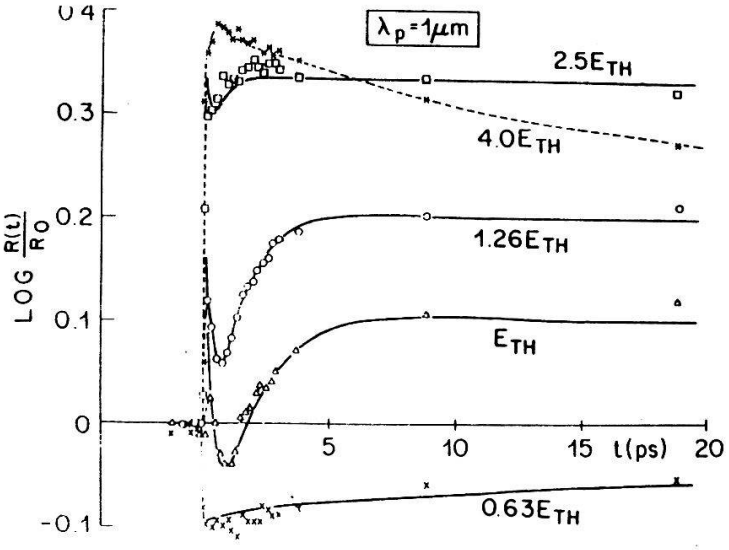

Figure 9 Transient reflectivity data in silicon at probe wavelength 1μm and pump wavelength 620 nm at various incident energies.

The figure 9 shows the results we obtained for a 1μm probing wavelength, for various exciting energies around the amorphization threshold energy E_{th}. For energies below threshold the reflectivity first decreases as one can expect from a simple Drude model for a photo carrier density $n = 5 \times 10^{21}$ cm^{-3}. It then increases again due to carrier diffusion away from the excitation region.

As the pump energy is increased it becomes possible for the plasma frequency to exceed the pumping frequency, the dielectric function becomes imaginary under such condition and the reflectivity increases within the first 100 fs following excitation. Thus far the reflectivity is described in terms of a dense solid state plasma.

At later times the reflectivity change above E_{th} has an oscillation like behavior which can be explained by considering a thin molten layer at the surface which expends in depth into the bulk with a velocity v. Using the optical properties of molten silicon and well known optical formulas, it is shown that interference takes place between the wave reflected from the air-melt surface and the melt-solid-state plasma interface.

Because of its high time resolution capabilities this experiment has permitted the observation of the transition between a solid state plasma and a molten material.

Conclusion

Time resolved spectroscopy with resolution better than 100 fs is now currently available. High power pulses in the gigawatt regime are also available, opening the way for the generation of very broad white light continua, which one can use for frequency spectroscopy. These high power pulses should lead to new information on the properties of highly excited condensed

matter, just below destruction, with a time resolution two orders of magnitude better than previously achieved.

New progress in the field is expected which should lead to higher repetition rates in the KHz regime for the amplified pulses. On the 100 fs time scale, light scattering experiments as well as photoluminescence cannot be directly achieved due to the lack of streak cameras which such a high resolution ; there is a need for developing an appropriate optical gate.

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