FEMTOSECOND REFLECTIVITY MEASUREMENT OF HIGHLY PHOTEXCITED SILICON

C. Hirlimann

To cite this version:

C. Hirlimann. FEMTOSECOND REFLECTIVITY MEASUREMENT OF HIGHLY PHOTEXCITED SILICON. Journal de Physique Colloques, 1983, 44 (C5), pp.C5-99-C5-105. <10.1051/jphyscol:1983516>. <jpa-00223096>

HAL Id: jpa-00223096
https://hal.archives-ouvertes.fr/jpa-00223096
Submitted on 1 Jan 1983

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
FEMTOSECOND REFLECTIVITY MEASUREMENT OF HIGHLY PHOTOEXCITED SILICON*

C. Hirlimann

Université P. et M. Curie, Laboratoire de Physique des solides**, 4, place Jussieu, 75250 Paris Cedex 05, France

Résumé - Nous décrivons dans une première partie les progrès récents qui ont permis de descendre la durée des impulsions laser dans le domaine femtoseconde. Nous montrons ensuite l'application de cette technique à la très forte photoexcitation du silicium et présentons des mesures de réflectivité résolues en temps, à diverses longueurs d'onde que nous interprétons à travers des modèles simples.

Abstract - Recent advances have taken place in optical pulse generation techniques pushing optical pulse widths down into the femtosecond domain. Time resolved reflectivity measurements at various wavelength are presented and interpreted in the form of very simple models.

I. Colliding Pulse Modelocked Dye Laser.

The development of the colliding pulse modelocked (C.P.M.) dye laser, in 1981, has allowed the generation of optical pulses in the subpicosecond regime /1,2/.

Figure 1 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 shorts the pulse with each pass around the cavity until the limiting pulse width is achieved. The basic limitation is group velocity dispersion inside the cavity.

*This work has been done at Bell Telephone Laboratories, Holmdel New-York (U.S.A. under the direction of C.V. Shank.

**Laboratoire associé au C.N.R.S., LA 154.

Fig. 1. Colliding pulse modelocked laser resonator configuration.

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jphyscol:1983516
The additional mechanism operative in the colliding pulse configuration is the interaction of the two counter propagating 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.

The shortest optical pulses, in the range 65 to 90 fs, are produced with a thin (10 μm) 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 counter propagating pulses.

Figure 2 shows the experimentally measured autocorrelation function of the produced pulses, using second harmonic generation (S.H.G.) in a thin (100μm) crystal of K.D.P.. The optical pulse width deduced from the autocorrelation function is 65 fs.

The operating wavelength of such a dye is 620 nm, with energy per pulse less than 1nJ at a repetition rate of 100 MHz, depending on the cavity length.

![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.](image)

2. Ultrashort Optical Pulses Amplification.

With such a low energy per pulse it is necessary to amplify these ultrashort optical pulses if one wants to highly photoexcite semiconductors. This is done using a four stages dye amplifier pumped by a frequency doubled Nd: YAG laser (10 Hz rep.rate). 70 femtoseconds optical pulses have been amplified to peak pulse powers of several gigawatts. /3/.

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 frequency components which destroys the linear frequency sweep in the pulse spectrum.

The nonlinear effects are limited by the use of a multistage amplifier (Figure 3), 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 pulses. The use of saturable absorbers also provides isolation of the different stages.

The output pulses are then only linearly chirped by the group velocity dispersion through the various transparent media of the amplifier.

Fig. 3. Schematic diagram of four stage amplifier and grating compressor.

3. Pulse Compression Technique.

In order to recover the original pulse width one has to recompress the broadened optical pulses at the output of the amplifier.

More than a decade ago Gires and Tournois/4/ and Giordamine, et al. /5/ proposed shortening of optical pulses using compression techniques analogous to those used at microwave frequencies. The actual design of an optical compressor is due to Treacy /6/ and is simply a parallel pair of gratings. Each wavelength passing through the pair is diffracted at a different angle by the first grating and recolmimated by the second one. The end result is a wavelength dependent optical path.

Such a grating pair is set-up at the output of the amplifier (Figure 3) and compensates for the group velocity linear chirp in the pulses.

In this way it is possible to currently achieve 90 fs optical pulses in the gigawatt power regime, with 10 Hz repetition rate, at a wavelength of 620 nm.


The C.P.M. dye laser and its amplifier are not tunable. For spectroscopic purposes this difficulty is overcome by the generation of white light continua. This is done by focusing gigawatt amplified 90 fs duration pulses at 620 nm into a thin (500 μm) jet of ethylene glycol acting as a non linear medium /7/. A completely reflective optics is used to minimize group velocity dispersion effects. In this way efficient production of white light continua extending from 1000 nm to 430 nm, with time sweeps as small as 10 fs/100 nm, is achieved.

Various non linear processes are expected to be responsible for the wide extension of the continuum, but due to the steep time variation of the exciting pulse envelope, self-phase modulation becomes a sizable effect in the femtosecond regime, even with very small non linear coefficient (n₂~10⁻¹³ e.s.u) and thin media /8/. Therefore self-phase modulation is expected to act as a seed for the various four-wave mixing processes involved in the continuum generation. Unlike in the picosecond regime /9/ these non linear processes do not need to start from the quantum noise and should therefore be more efficient leading to the very broad continua observed.
5. Highly Photoexcited Silicon.

In the experiments reported here, we utilize the previously described femtosecond optical pulse techniques to excite a dense electron-hole plasma in crystalline Silicon and to measure the induced reflectivity changes as a function of wavelength time and excitation energy /10/.

The experimental arrangement is shown on Figure 4. A 90 fs pulse from the amplified C.P.M. dye laser is split into two pulses, forming a pump and a probe pulse. The probe pulse is focused into a cell containing D$_2$O to generate a white light continuum pulse. The pump pulse is focused to about 150 µm in diameter and the central 10% of the probing area is imaged onto the input spectrometer slit. The (111) silicon wafer is moved in a raster pattern so that each laser pulse sees a fresh region.

![Experimental set-up used in the measure of time resolved reflectivity of optically excited silicon.](image)

The Figure 5 shows the log of the reflectivity change as a function of time and pump intensities at three different wavelengths: 100 nm, 678 nm and 440 nm. The pulse threshold $E_{th} = 0.1 J/cm^2$ is defined by the formation of a clearly visible amorphous layer.

The pump pulse directly excites an electron-hole plasma in a thin layer on the Si surface having a depth $d = \alpha^{-1} = 3 \mu m$ at 620 nm. In the simple Drude model for the refractive index of the plasma:

$$n_p = n_c \left(1 - \frac{\omega_p^2}{\omega^2}\right)^{1/2}, \quad \omega_p^2 = 4\pi N_p e^2/\epsilon_c m^*$$

where $n_c$ is the refractive index for the crystalline silicon, $N_p$ the plasma density, $m^*$ the effective mass, $\epsilon_c$ the dielectric constant of the crystal.

At an excitation level below the melting threshold $E_{th}$ (Figure 5a), the reflectivity decreases immediately following excitation, indicating that the plasma frequency $\omega_p$ is less than the probe frequency $\omega$. From the minimum of reflectivity one can deduce a plasma density of the order $N_p = 6 \times 10^{20} cm^{-3}$ assuming a reduced effective mass for the electron and holes $m^* = 0.12 m_0 /11/$. This estimation is not very accurate depending on the effective reduced mass used. Also based on results obtained in heavily doped silicon /12/ the electron effective mass has been shown to double for carrier concentration around $5 \times 10^{14} cm^{-3}$.

For excitation levels at $1 \mu m$, the reflectivity sharply increases after the pulse (Figure 5a). We expect under these conditions the plasma frequency to exceed the probing frequency giving a plasma metallic reflection. This is in contrast with the slower rise in reflectivity for higher frequency in Figure 5b, 5c (with the exception of the higher excitation level at 678 nm). Therefore, the plasma wavelength at $E_{th}$ lies between $1 \mu m$ and 678 nm, which is consistent with the estimated value for the plasma density.
Fig. 5. Transient reflectivity of silicon at three probe wavelengths following a 90 femtosecond excitation pulse at 620 nm.
It has been shown (COMBESCOT M., BOK J., Private Communication) that, for such plasma densities, Auger recombination accurately accounts for the slow recovery of the reflectivity after excitation below threshold at 1\,\mu m.

In reference 10 the free electron mass was used in estimating the plasma density yielding a value around $5 \times 10^{21}$ cm$^{-3}$. For such a high density Auger recombination cannot account for the reflectivity recovery. Further experiments remain to be done to study the recombination processes of a very dense plasma in silicon.

Thus far the reflectivity change has only been described in the first few hundred femtoseconds in terms of a dense solid state plasma. At later times for intensities near and above threshold considerable structure is observed in the reflectivity spectrum. The reflectivity change is observed to increase then change sign and then increase to a plateau value within the first picosecond and a half. This behaviour can be understood by considering a thin molten layer expanding from the surface into the bulk with a velocity $V$. Using the optical properties of molten Si /13/ and thin film optical formulas, we find that destructive interference between the wave reflected from the air melt interface and the melt solid state plasma interface lowers the intensity of the reflected beam. Since the Si melt is highly absorbing, the optical reflectivity becomes dominated by the optical properties of melted Silicon when the melt front has penetrated more than an absorption depth for the probing radiation.

The velocity of the melt front and the plateau reflectivity were left as free parameters of the fit to the data. We find that a single melt front velocity of $6.2 \times 10^5$ cm/s closely matches the reflectivity for all observed wavelengths for an excitation of $1.0 \, E_{th}$. This value is near the velocity of sound in crystalline Silicon. In a similar manner, we fit the data at $1.26 \, E_{th}$ and found a melt front velocity of $9 \times 10^5$ cm/s. To explain the rapidly using reflectivity at $2.5 \, E_{th}$ a velocity of $25 \times 10^5$ cm/s is necessary. Such high values of the melt front velocity might be due to local melting in the depth resulting in an effective velocity.

For all three probing wavelengths the final plateau value of the reflectivity matches the melted silicon reflectivity for an excitation energy $2.5 \times E_{th}$. A striking and not clearly understood feature is the dependence of the final plateau reflectivity on the exciting energy below $2.5 \times E_{th}$.

At the highest excitation level of $4.0 \, E_{th}$, the crystal is damaged and the reflectivity variation is probably due to a combination of plasma emission and scattering effects.

A unique aspect of these experiments is that by exciting the silicon surface with a short 90 fs optical pulse, we have been able to create a previously unobserved unstable form of highly excited Silicon which persists for a fraction of a picosecond. The properties of this material remain to be determined in detail. We observe apparent melting to take place after the excitation rather than during the exciting pulse.

Reference.

4. GIRES F. and TOURNOIS P. Comp. Rend. (Paris) 258, (1964) 6112


11. COMBESCOT M. and NOZIERES P. Solid State Comm. 10, (1972) 301
