GIANT OSCILLATIONS IN THE PICOSECOND ULTRASONICS RESPONSE OF CRYSTALLINE SILICON : CONNECTION WITH THE ELECTRONIC STRUCTURE

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Abstract

We present the results of picosecond ultrasonic experiments performed at different laser wavelengths in the vicinity of the direct band gap of silicon which show important oscillations in the reflectivity curves. We explain the existence of these oscillations by the fact that in this case, the strain pulse is detected by the probe directly inside the sample, without the use of the transducer. We propose then that the high amplitude of these oscillations comes from the vicinity of the direct band gap where the piezo-optic couplings are expected to change much with the probe wavelength. These results demonstrate that in semiconductors too, interband transitions may strongly influence picosecond ultrasonics studies.

Introduction

Thomsen and al. have first shown that ultrashort optical pulses can generate picosecond acoustic pulses which lead to study acoustic or thermal properties of thin films [1][2]. With this so-called picosecond ultrasonic technique, it is possible to generate acoustic waves whose frequency is so high (up to several hundred GHz) that they suit very well thin films characterization. It has been used to access to thicknesses, sound velocities, attenuation measurements using ultrasonics in a frequency range that is inaccessible by conventional techniques [3]. We have recently suggested that picosecond ultrasonics can also play a role in the study of electronic interband transitions in thin metallic films [4][5].

Here we present experimental results obtained in a silicon substrate. From these results we deduce that in semiconductors too, interband transitions may strongly influence picosecond ultrasonics studies.

The paper is organized as follow : in the first part we come back on picosecond ultrasonics and its connection with electronic structure. Then we describe the experimental set up. In a third part we present the experimental results. Then we propose an interpretation of these results which involves a well-known interband transition of silicon. That let us conclude about the general effect of interband transitions in semiconductors on picosecond ultrasonics.

Picosecond ultrasonics and electronic structure

In a picosecond ultrasonic experiment, a first optical pulse (the pump pulse) is incident at the sample surface where it is absorbed and the resulting dilatation generates a strain pulse the extension of which is related to the absorption length. In the particular case of a metal, absorption can be very strong giving a length of a few nanometers. Such a mechanical pulse propagates into the sample at the longitudinal sound velocity (typically a few nanometers per picosecond), which explains how absorption can generate picosecond acoustic pulses. Each pulse is reflected onto the film-substrate interface and the resulting echo comes back to the surface and modifies this way the dielectric constant of the film. This change can be detected by another optical pulse (the probe pulse) whose reflection or transmission is affected by the presence of the strain wave. By adjusting the delay between pump and probe pulses it becomes possible to monitor the successive echoes due to the strain generated by the pump pulse. Usually, these echoes are used to measure thicknesses of thin films, sound velocities, attenuation and so on.

The detection mechanism of such an experiment is based on the modification of the optical properties of materials when they are strained. This modification affects the reflection of the probe beam. A detected echo is not the strain pulse itself but instead its impact on the optical properties of the film. The interaction between the strain wave and the optical index is measured by the piezo-optic couplings. We have shown that they strongly influence the qualitative shape of echoes [4]. As they are very sensitive to interband transitions in the electronic structure. One may detect interband transitions by studying the shape of these echoes.

In the case of a layer which is partially transparent for the probe beam, the acousto-optic interaction acts in the whole layer and the short acoustic pulse is replaced by a decaying oscillation [6]. The period of these so-called Brillouin's oscillations may be written as :

$$T = \frac{\lambda}{2nv\cos\Theta} \qquad (1)$$

where λ is the probe wavelength, n is the index of refraction, v is the sound velocity and Θ is the angle of incidence. One should notice that the amplitude of these oscillations also reflects the value of the piezo-optic couplings. Following the previous study, these oscillations should strongly vary in the vicinity of an interband transition. That is what is demonstrated here in silicon.

Experimental details

The experiment is based on a conventional pump and probe setup associated with a tunable titanium:sapphire oscillator which produces 120 fs optical pulses with a repetition rate of 76 MHz centered at a wavelength tunable between 700 nm and 990 nm. The laser output is split to provide the pump and probe beams with crossed polarizations. The probe pulse can be delayed with respect to the pump pulse by an optical delay line based on a translation stage. Both beams are focused on the same point of the sample by a 60 mm lens. To improve the signal-to-noise ratio, the pump beam is chopped using an acousto-optic modulator and the output of the photodiode, which monitors the reflected probe, is amplified through a lock-in scheme.

Fluctuations in the probe intensity were normalized by splitting the probe into signal and reference beams and monitoring the difference between the signals detected in identical photodiodes.

Complementary experiments were also performed with a blue probe obtained by focusing the laser beam into a BBO crystal to generate the initial second harmonic.

The results we are presenting here concern a sample that was composed of an amorphous silicon dioxide (SiO_2) layer of thickness of 4400 Å grown on a (100) silicon substrate using plasma-enhanced chemical-vapor deposition (PECVD). A 120 Å thick film of polycrystalline aluminum has been evaporated on the silica layer in order to generate and detect acoustic waves with the laser.

Results

A basic experiment

Figure 1 reproduces the transient reflectivity signal measured in the sample. Both pump and probe pulses are centered at 810 nm. The signal is first composed of a jump at zero time that is the electronic contribution to the pump-probe signal. Few picoseconds later, the pump energy is converted into heat, which diffuses on a larger time scale. This leads to a slow decrease of reflectivity. Figure 1 also shows several acoustic echoes. The first one appears near 150 ps and corresponds to one round-trip of the acoustic pulse in the SiO_2 film : we retrieve this value by assuming a sound velocity of 5.9 nm/ps [7] in the 4400 Å SiO_2 layer. The second echo near 300 ps is hard to detect due to the low acoustic contrast between SiO_2 and Si.

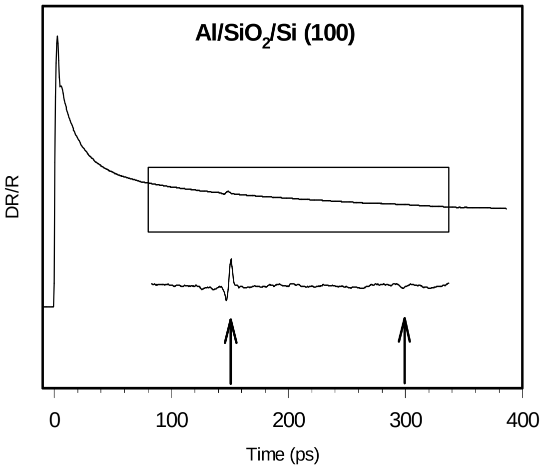

Figure 1. Transient reflectivity for the Al/SiO2/Si sample (λ probe : ~800 nm). The first arrow points the first acoustic echo, the second arrow points the second echo.

Giant oscillations

Figure 2 reproduces the signal obtained in the same sample using a blue probe and an infrared pump. First the signal has greatly changed : electronic, thermal and acoustic contributions have been strongly affected by the modification of the probe wavelength. Concerning the acoustic part one observe first an echo near 150 ps, which is the only common point in both experiments.

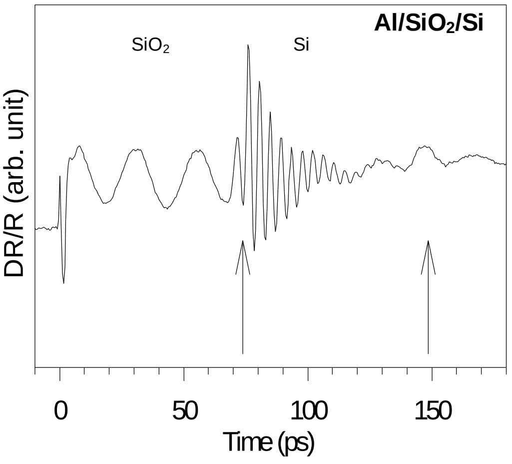

Figure 2. Transient reflectivity for the Al/SiO2/Si sample (λ probe : ~400 nm). The first arrow points the time when the strain pulse enters the Si layer, the second one points the echo detected at the surface in the Al layer.

Figure 2 also reveals strong oscillations : the first starts at t=0 and has a low frequency (T~24 ps). The second starts near 70 ps and has a much higher frequency (T~5ps). It should be noticed that these last oscillations appear at the precise time of arrival of the strain pulse into the substrate. As explained before the oscillations may result of an acousto-optic interaction respectively in the SiO_2 layer and in the Si substrate.

For the high frequency oscillations, the ratio $\Delta R/R$ reaches nearly 10^{-3}, which is very high compared with the ratio 10^{-5} generally observed in picosecond ultrasonics.

The wavelength dependence of the oscillations period

To confirm this interpretation we have explored different laser wavelengths in order to check the expected dependence of the Brillouin's oscillations. Figure 3 presents the evolution of the experimental period with the probe wavelength and a theoretical curve obtained by using the equation (1) and literature values of the index of refraction of silicon [7].

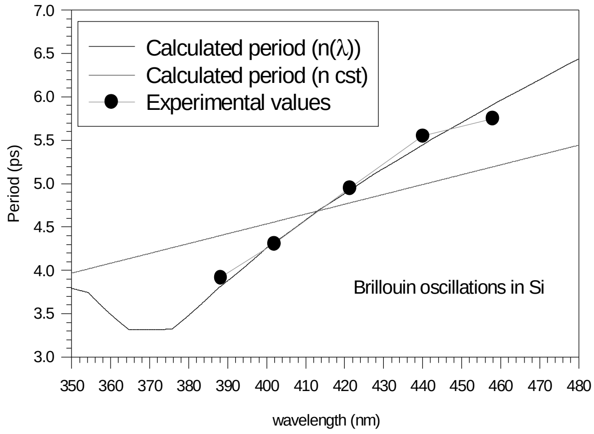

Figure 3. Brillouin period versus probe wavelength. Black points : experiments. Black line : theoretical curve. Red line : theoretical curve if n was constant versus λ.

One should notice the excellent agreement we have obtained.

These results finish to demonstrate that the oscillations well correspond to Brillouin's oscillations detected in the silicon substrate.

Discussion

The experimental results presented here have let us conclude that using a blue probe we detect a strong contribution of the silicon substrate which displays an oscillatory contribution due to the small absorption of the semiconductor. In a similar experiment, but with very different samples –except the silicon substrate– Bosco and al. [9] observed similar oscillations. This confirms the key role of the substrate in the amplitude of the oscillations One has also performed experiments with a red-infrared probe but in that case no such signal was detected. One may argue that in that last case all the probe beam is absorbed in the aluminum layer. Taking literature values for the index of refraction of aluminum one finds that the absorption length is shorter in the blue case than in the red one.

One way of understanding such results is to involve a change in the value of the detection coefficient. The acoustic pulse is the same since the pump beams are identical in both cases. The relative probe intensities inside the silicon substrate are smaller in the blue case whereas a higher contribution is observed. We conclude that the acousto-optic parameter should be strongly dependent with the probe wavelength. More precisely this parameter should increase in the blue region.

Here again the detection mechanism involved in picosecond ultrasonics is found to be strongly dependent with the laser wavelength. As in the previous studies we can rely this to an interband transition in the electronic structure of the studied material. Due to the nature of the detection mechanism, picosecond ultrasonics is sensitive to the derivatives of optical properties of materials : the derivatives of the real and imaginary parts of the index of refraction with respect to the strain. An interband transition produces a small irregularity on the optical index of a solid but a discontinuity of the first-derivative response, which could thus affect the detection mechanism in picosecond ultrasonics.

In the particular case of silicon, one has to remember that it is an indirect gap semiconductor. A particular interband transition known as the direct band gap is localized near 3.4 eV which corresponds to a photon wavelength of 365 nm. This transition is a vertical transition between the top of the valence band and conduction band in Γ. It should be noticed that it precisely falls in the wavelength range where we have made our observation. When we come close to this transition, the damping of the oscillations becomes so important that we can't measure their period anymore. As expected, the silicon is more and more opaque as the photons energy get closer to the direct gap. Nevertheless, the amplitude of the oscillation remains very important as shown on the Figure 4, and the curve's shape then looks like half an echo localized at the time when the strain pulse enters the silicon layer, say at half the time of a round-trip.

The effect of an interband transition, seen here in a single crystal, may be all the more important as the material is almost perfect. Moreover, the data available about the energy gaps of silicon substrates are very accurate. There is thus no doubt that the probe wavelength is close to the direct gap. Our previous experiments made on polycrystalline metals [4] didn't display such important changes in the amplitude of the probe response.

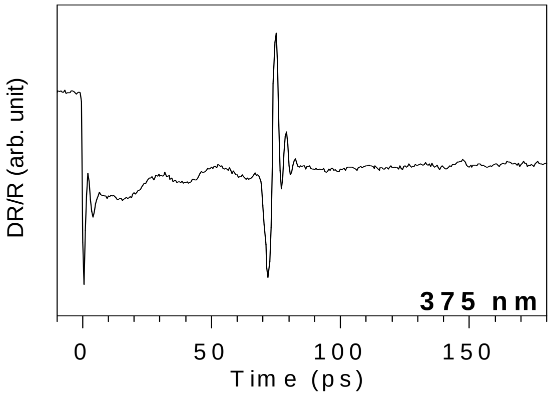

Figure 4. Transient reflectivity for the Al/SiO_2/Si sample (λ probe : 375 nm). Since λ is close to the gap, the amplitude of the oscillations remains high but the damping is important.

Conclusion

We have presented experimental results that confirm the connection that exists between picosecond ultrasonic experiments and electronic structure. We started from the observation of strong oscillations in the transient reflectivity for the Al/SiO_2/Si sample. Complementary experiments have demonstrated that these oscillations come from a strong change in the piezo-optic coupling of silicon near 370 nm. We have then relied the high amplitude of these oscillations to the direct band gap of silicon which falls in the same wavelength range.

Similar experiments could be performed in other semi-conductors whose electronic band structure is the same as silicon. In particular we expect similar effects in germanium for which the direct gap falls in the near infrared and in SiGe alloys.

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