PICOSECOND ACOUSTICS WITH VERY HIGH LATERAL RESOLUTION

T. Bienville, L. Belliard, P. Siry, and B. Perrin
Laboratoire des Milieux Désordonnés et Hétérogènes, UMR 7603,Université Pierre et Marie Curie, Paris
lbelliar@ccr.jussieu.fr

Abstract.

We describe in this paper an experimental setup coupling a pump-probe technique with a near field optical microscope in order to reach a submicronic lateral resolution in picosecond acoustics measurements. In spite of the very weak light intensity which is detected, we proved that acoustic features like acoustic echoes or individual modes of vibration in nanostructures can be detected.

Introduction.

When a laser pulse is focused on a metallic surface, the light is absorbed within the absorption length (10-20nm in usual metals). This light absorption generates a sample heating which can be detected by the change of the sample reflectivity or the surface displacement. An additional phenomenon can be generated if subpicosecond laser pulses are used. Indeed, in 1984, the group of Humpfrey Maris [1] discovered that very short acoustic pulses are generated in that case. The duration of these pulses, which can reach a few picoseconds, is only limited by the physical properties of the materials and not by the laser pulse width. In this technique, so called picosecond acoustics, an ultra-short laser pulse (the pump) generates the acoustic waves which are detected by a less energetic laser pulse (the probe) which is time delayed with regard to the pump. The small spatial extension of these pulses, only a few nanometers, gives a very high axial resolution (along the propagation direction). This property has been intensively used to study thin films or multilayers systems [2,3].

Today, the race towards micro and nanotechnology implies to investigate artificial structures with submicrometric dimensions. Some advances have already been done in this direction in the field of picosecond acoustics. Nanoparticles in suspension in liquid or embedded in transparent glass matrices have been studied. Some individual modes of vibration have been observed in these systems [4,5,6]. In such experiments, the properties of individual nanoparticles are not directly extracted but averaged over a large number of excited objects. Even if the size dispersion can be reduced, other parameters like the crystallographic orientation are not uniform and measurements on a single object could be interesting. Collective modes like surface waves have also been generated and detected in patterned arrays or using a non homogeneous illumination across a transmission diffraction grating [7-8].

In general, if the optical illumination or/and detection are performed in the far field region, surface waves with frequency above a few GHz cannot be studied due to the optical diffraction limit. Moreover, the acoustic signature of individual objects is always mixed with the contribution of neighbors objects. In conclusion, the crucial problem to be solved in order to extend the investigation field of picosecond acoustics to nanotechnologies is to improve its lateral resolution.

Since the advent of the scanning tunneling microscope in 1982 [9], the scanning near field microscopies have been intensively developed in order to cover a wide field in surface science. Pohl was a pioneer in introducing the concept of tip sample interaction to extract some optical properties. The main objective was to illuminate a sample with a very small light source or to collect the reflected light with a very small aperture (smaller than the optical wavelength) [10]. Today a large variety of optical near field microscopes exists around the world and optical resolution around 50nm or even less are usually obtained. The natural following step was to couple ultrafast optical setups such as picosecond acoustics to an optical near field microscope. Recently, Vertikov [11] reported such acoustic measurements in gold stripes using a near field configuration. He used a two colors light method and found differences between measurements performed either in far field or near field, but vibrations of an individual gold stripe was not clearly detected.

In the present paper, we described the coupling between a picosecond acoustics setup with a near field optical microscope working in a reflection configuration called RSNOM [12]. The possibilities of such a system will be first described. Then acoustic features detected on a flat tungsten layer will be discussed. Finally, investigations on small nanostructured dots will be reported.

Experimental set-up.

The figure 1 represents the general set-up including a standard pump-probe scheme and a near field optical microscope. The laser source is a mode locked Ti:sapphire laser operating at 750nm and providing optical pulses of around 130fs. The repetition rate is 82MHz.

First, the laser beam is split into two arms, namely the pump beam which is the more energetic and the probe beam. The pump beam is modulated using an acousto-optic modulator working up to a few MHz. This light

modulation, coupled with a lock-in detection, is necessary due to the very small relative changes of reflectivity (10^{-5} to 10^{-7}) induced by the acoustic phenomena.

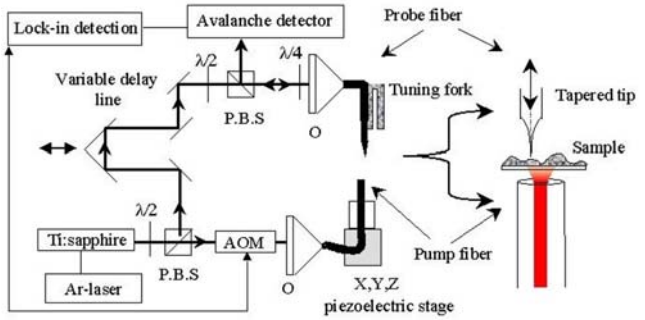

Figure 1: Experimental set-up

Moreover the light modulation frequency is an important parameter in photothermal experiments through its relation with the thermal diffusion length. The pump beam is injected in a single mode fiber using an objective lens with a numerical aperture close to 0.1. By this way, we succeeded to couple into the pump fiber more than 70% of the incident power. Finally, the pump fiber is approached to the back side of the sample to illuminate it through a transparent substrate.

The probe beam travels across a delay line, built with a corner cube mounted on a motorized translation stage. By moving the translation, we achieved a control of the time delay between pump and probe with a resolution under 20fs. A maximum time delay of 4.5ns can be achieved. Then the probe goes through a λ/2 plate to optimize the transmittance of a subsequent polarizing beam splitter. The linear polarization is converted into a circular one by a λ/4 plate and injected into an other single mode fiber (the probe fiber). This optical probe fiber is chemically etched in order to get an apex radius less then 50nm. The fiber is also glued on a quartz tuning fork. The fiber could be assumed to be rigid as soon as the fiber length without the jacket doesn't exceed 1mm. Using an electrical excitation and detection [13], we control the amplitude of vibration of this oscillator, located around 32KHz. When this oscillator interacts with the sample surface, the frequency and amplitude resonance are modified. A regulation of the probe-sample distance can be performed with a feedback loop, modifying the voltage applied to the piezoelectric stage on which the sample is mounted. The figure 2 shows a typical approach curve which exhibits a very fast decrease in the amplitude of vibration when the sample is very close to the end of the probe fiber.

Actually a regulation distance less than 10nm is routinely achieved. In such a shear force regulation system, this distance is not the only crucial parameter. The amplitude of the tip vibration in the sample plane can strongly affect the final lateral resolution. This amplitude of vibration has been measured between

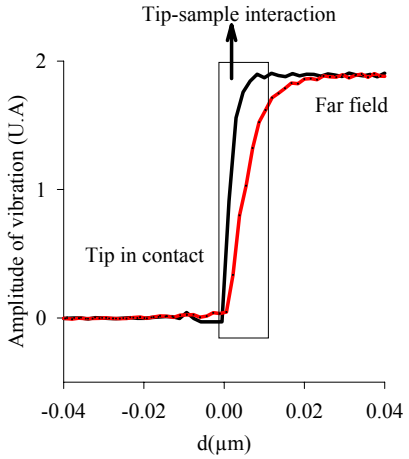

Figure 2: Approach curve showing a drastic change in the probe vibration in a range of 10nm.

5nm and 10nm by an interferometer ; these values are much smaller than the final expected resolution.

The light which emerges from the probe fiber is reflected by the sample surface and only a small amount of this light is coupled back into the same probe fiber and detected with an avalanche detector in order to perform lock-in detection or dc mapping of the sample reflectivity. Topographic images can be recorded simultaneously. An avalanche photodiode is required to detect the small intensity collected from the sample (~10μW). Figure 3 shows a mapping of the sample reflectivity obtained on an array of aluminum dots sputtered on a silicon substrate ; the dot diameter is 200nm.

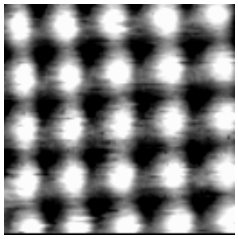

Figure 3: Reflectivity mapping of an aluminum dots array deposited on a silicon substrate. The period is 400nm and the dot diameter 200nm.

An optical lateral resolution around 100nm is routinely achieved. A better resolution could be obtained with a coated optical fiber but, in that case, it would be difficult to recollect the reflected light by the same coated probe fiber due to the small etched fiber transmittance (10^{-3} to 10^{-5}). Aperturless configuration is also a promising way to improved the resolution [14].

Acoustics echoes in tungsten layer.

Using the opportunity to move the sample with the piezoelectric stage, we can investigate the sample reflectivity change induced by the pump

excitation and record photothermal images. The figure 4 displays a photothermal image obtained on a 250nm thickness tungsten layer directly sputtered on the cleaved end of the pump fiber.

Figure 4: Photothermal image obtained with a modulation frequency of 1MHz. The image scale is 20.8µm.

When the modulation frequency is changed, the amplitude and the extension of the heating source are affected due to the change of the thermal diffusion length. Further photothermal mapping or transient photothermal studies have been done in this near field configuration but it is out of the scope of this paper. After having located the excited area, the probe location is chosen to perform acoustic measurements. A temporal scan recorded for a probe location at the center of the heating spot on a 600 ps range is shown on figure 5a.

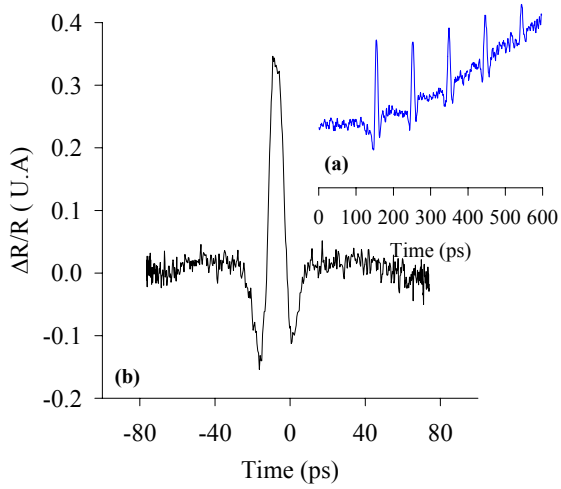

Figure 5: a) Five acoustic echoes spaced by 97.6ps are detected, in good agreement with the tungsten thickness. b) Experimental shape of the first echo.

As expected, we detected several echoes due to the large acoustic mismatch between the tungsten layer and the quartz substrate. These echoes are superimposed on a progressive heating due to the absorption of one laser pulse.
The variation of the echo amplitude can be studied in order to get information on the reflection coefficient at the interface with the glass substrate. In our tungsten layer, we find a coefficient equal to 0.68 instead of an expected value of 0.77 for a tungsten/glass interface. The discrepancy between the experimental and the expected reflection coefficient can be ascribed to the attenuation in the tungsten film. Moreover, we can record the echoes shape at different probe locations: a decrease of the echo amplitude has been observed when the probe location is farther from the pump probe center but no notable variations of the acoustical reflectivity were detected on the excited area. In such a way, adhesion properties could be mapped at a submicronic scale. For much larger distances from the acoustic pulse epicenter, the existence of a circular surface wave has also been evidenced [15].

Individual or collective vibration modes in nanostructures.

The first studied structure is made of aluminum dots, with a lateral size around 600nm and 90nm height, sputtered on a 90nm aluminum underlayer.

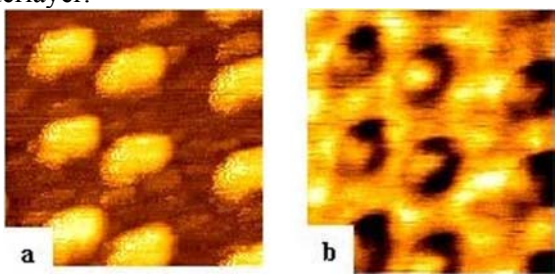

Figure 6: a) Topographic image. The image scale is 2.6µm. We can notice that the structure is not well defined in shape and in distance. The maximum dot size is around 700nm and the minimum one is 500nm. b) In phase photothermal mapping.

We recorded a large photothermal image to locate the excited area and we focused at the center. The figure 6 presents, topographic and photothermal (in phase) images measured simultaneously on this patterned array. The photothermal image presents submicronic features. Time-resolved measurements have been performed on and beside one aluminum dot. Results are displayed on figure 7.

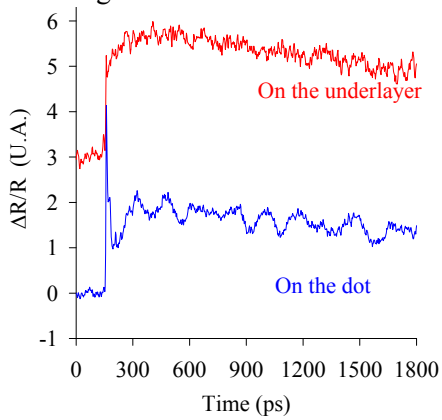

Figure 7: Temporal evolution over 1.8ns for two locations on the dot array.

The curve obtained on the aluminum underlayer shows an usual signal with only one acoustic echo

masked by the thermal step. In that case, all the acoustic energy is lost in the substrate and a large series of echoes is not expected at the free surface. In contrast, the curve obtained on the dot exhibits oscillations with a complex spectrum peaked around 6.3GHz. These additional oscillations can clearly be ascribed to individual vibrational modes of the aluminum dot. Nevertheless, we didn't try to assign these oscillations to specific dot modes since the dot shape is irregular. In the future, we will work on samples with well defined dot shapes and variable dot sizes in order to clearly identify these oscillations. The second studied structure was made of aluminum dots with a lateral size around 0.87 µm to 1.45µm and a 75nm height, sputtered on a 250nm tungsten underlayer.

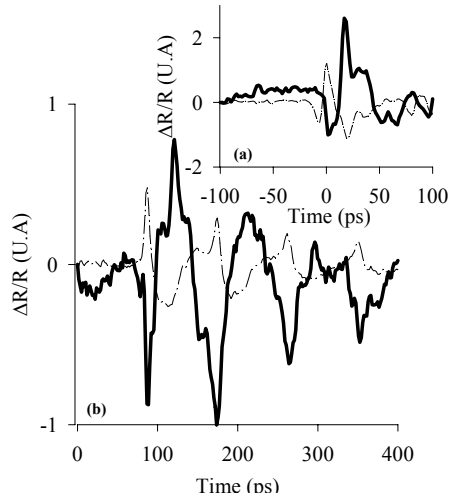

Figure 8: Change of reflectivity on the dot (bold line) or on the tungsten underlayer (normal line).

Preliminary results in figure 8a, show that the acoustic signal measured on the dot presents a time delay of around 10.8ps compared with the signal obtained on the tungsten underlayer. This value is close to 11.7ps, the time required for the acoustic waves to travel across the dot thickness.

On a larger temporal scan (figure 8b), a complex structure can be observed. The tungsten underlayer signal exhibits sharp peaks separated by 95ps in good agreement with the time needed to perform a round trip in the layer. Additional sharp peaks can be clearly seen on the signal recorded on the dot. Further experiments are under investigations to obtain a full understanding of these complex acoustic signals.

Conclusion.

We have demonstrated the possibility to perform picosecond acoustics measurements with a lateral resolution around 100nm using a near field optical microscope working with an uncoated tip and a shear force regulation. Acoustic echoes in a tungsten layer and individual vibrational modes in a submicronic aluminum dot have been detected. Further investigations are under progress to extend such preliminary results to still smaller size dots.

References.

[1] C. Thomsen, J. Strait, Z. Vardeny, H. J. Maris, J. Tauc, Phys. Rev. Lett. **53** (1984) 989.

[2] H. Grahn, H. J. Maris, J. Tauc, IEEE Journal of Quantum Electronics, **25** (1989) 2562-2569.

[3] B. Perrin, in « Systèmes Femtosecondes », P. Laporte and F. Salin (eds.), Publications de l'Université de Saint Etienne, 2001, 65-89.

[4] A. Devos, B. Perrin, B. Bonello, J.-C. Jeannet, Proceedings of the Tenth International Conference on Photoacoustic and Phtotothermal Phenomena - AIP Conference Proceedings **463** (1999) 445.

[5] J. H. Hodak, I. Martini, G. V. Hartland, J. Chem. Phys. **108** (1998) 9210.

[6] N. Del Fatti, C. Voisin, F. Chevy, F. Vallée, C. Flytzanis, J. Chem. Phys. **110** (1999) 11484.

[7] G. A. Antonelli, P. Zannitto, H. J. Maris, Physica **B 316** (2002) 377.

[8] B. Bonello, A. Ajinou, V. Richard, Ph. Djemia, S. M. Cherif, J. Acoust. Soc. Am. **110** (2001) 1943.

[9] G. Binnig, H. Rohrer, C. Gerber, E. Weibel, Phys. Rev. Lett **49** (1982) 57.

[10] D. W. Pohl, W. Denk, M. Lanz, Appl. Phys. Lett. **44** (1984) 651.

[11] A. Vertikov, M. Kuball, A. V. Nurmikko, H. J. Maris, Appl. Phys. Lett. **69** (1996) 2464.

[12] D. Courjon, J. M. Vigoureux, M. Spajer, K. Sarayeddine, S. Leblanc, Appl. Opt. **29** (1990) 3734.

[13] K. Karrai, R. D. Grober, Appl. Phys. Lett. **66**, 1842 (1995); Ultramicroscopy **61** (1995) 197.

[14] F. Zenhausern, M.P. O'Boyle, H.K. Wickramasinghe, Appl. Phys. Lett. **65** (1994) 1623

[15] P. Siry, L. Belliard. B. Perrin, Acta Austica United With Acustica **89** (2003) to be published.