PICOSECOND ULTRASONICS

Applying ultrasound in the Megahertz range for the investigation of the human body is a well-known medical practice. But using ultrasound to study nanometer structures requires ultrahigh sound frequencies in the Gigahertz range. Delft University of Technology researcher Gerard Verbiest and his team succeeded in reaching vertical nanometer accuracies thanks to the photoacoustic effect. But achieving these accuracies in lateral directions required a second trick: the application of an atomic force microscope.

FRANS ZUURVEEN

In the field of nanotechnology, structures have to be investigated with a resolution of a few nanometers. Existing imaging methods working with either electrons or photons have the disadvantage of not being able to penetrate deep into materials. Moreover, materials like silicon are totally opaque for electrons or photons – i.e. at visible wavelengths; silicon becomes transparent at a wavelength of ~1 µm wavelength, which is however too large for nanometer-scale imaging. And the situation is even worse for metals, which is why ultrasound can help to 'look' deeper into materials with nanometer resolution.

Photoacoustics

Already in 1880, Alexander Graham Bell experimented with sound waves, aiming to reach long-distance signal transmission. He invented the 'photophone', which enabled the transmission of vocal signals by reflecting sunlight from a moving mirror to a selenium-based solar receiver. The essence of his discovery was the awareness that absorbed visible light may cause structural changes inside materials, resulting in the emission of acoustic waves.

Bell did not really understand the origin of these acoustic signals, but today we know that they originate from the local heating of the lattice structure inside materials. This so-called photothermal mechanism causes the generation of acoustic waves in materials, which can be observed by sensitive sound sensors.

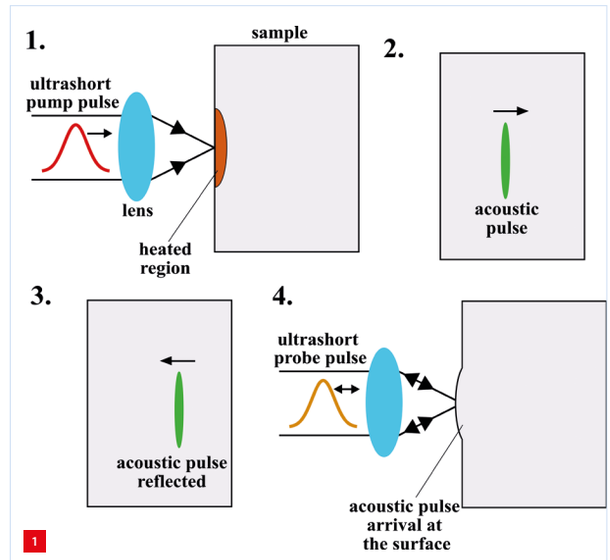
From piezo to light

Already for a few decades, acoustic waves, generated by piezo transducers, have been applied in a highly useful medical aid utilising acoustic 'echoes'. Among other clinical applications they facilitate the widely practiced examination of pregnant women. The frequencies involved here correspond with wavelengths of about 0.1 to 1 mm, depending on the material. For example, in water the wavelength at a medical ultrasound frequency of 10 MHz would be 1,500 ms^{-1}/10 MHz = 150 µm.

Upgrading the ultrasound resolution to the nanometer range requires acoustic signals with extremely short wavelengths, even down to 100 nm, and thus frequencies in the GHz range. In water, see the example above, a wavelength of 150 nm (i.e. a factor of 1,000 lower) would require a frequency of 10 GHz. Piezo transducers are not able to generate such short acoustic waves, so the photoacoustic effect is employed, using femtosecond light pulses. Such pulses are absorbed by the material within 1-2 ps, resulting in an acoustic wave with a duration of 1-2 ps. This research field is therefore called 'picosecond ultrasonics' [1].

Ultrashort optical pulses

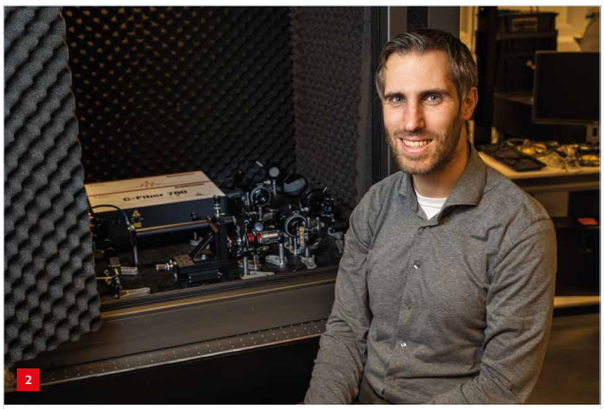
These ultrashort optical pulses are called 'pump pulses'. When these pulses hit the surface of an opaque solid,



The principle of photoacoustics.
(1) A pump pulse of light induces changes in the material structure (heated region).
(2) An acoustic pulse is generated and starts travelling through the material.
(3) The acoustic pulse is reflected from an (internal) interface.
(4) The reflected pulse ('echo') arrives at the surface and is 'detected' by an ultrashort probe pulse; see the text for a more detailed explanation of the detection principle.

AUTHOR'S NOTE

Frans Zuurveen, former editor of Philips Technical Review, is a freelance writer who lives in Vlissingen (NL).
This is his last contribution to Mikroniek; see also page 32.

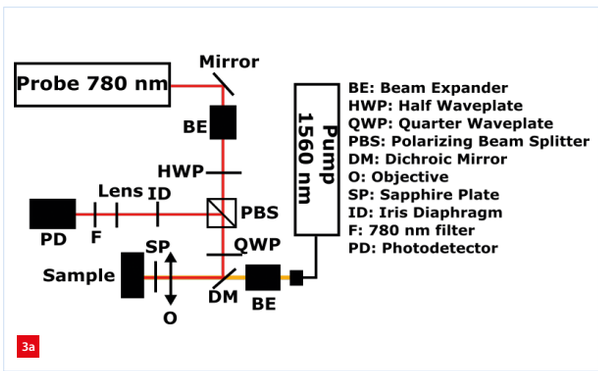


Gerard Verbiest with his photoacoustic set-up. (Photo: TU Delft)

some optical energy is absorbed and converted to heat. Figure 1 illustrates this process, with the final panel showing the reflected acoustic pulse returning (as a kind of 'echo') at the material surface.

Gerard Verbiest (Figure 2) and his team in the Precision and Microsystems Engineering department at Delft University of Technology (TU Delft) participate in this research field and succeeded in realising the emission of these ultrashort acoustic waves by means of the photoacoustic effect.

This phenomenon is highly effective in the investigation of thin films and nanostructures because the acoustic penetration depth is much larger than the optical absorption depth of 10 to 50 nm. The generated acoustic pulse can be modelled as a superposition of longitudinal plane waves of different wavelengths travelling normal to the surface, which is fundamental for the realisation of depth resolutions with nanometer accuracy. When, for example, two interfaces within the material under study each generate a reflected signal ('echo'), the time elapsed between the detection of the two successive echoes can be converted into the distance between these two interfaces by bringing the (material-dependent) velocity of sound into the equation.



Photoacoustic set-up, with labelling of the main components.
(a) Schematic drawing; see the text for explanation.
(b) Practical set-up.

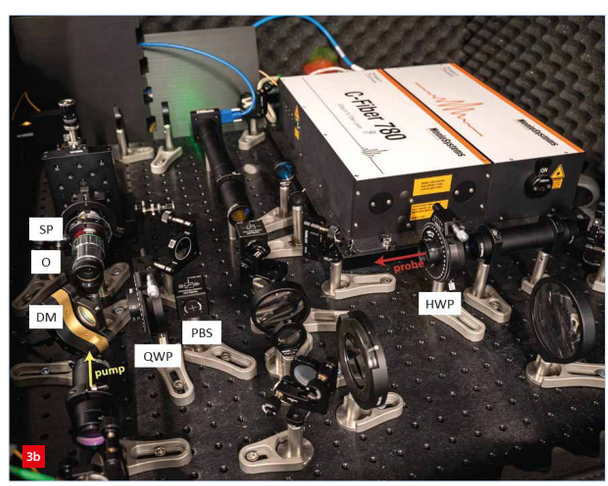
Practical realisation

Among other researchers, the Delft team succeeded in translating the theoretical principles described above into an experimental set-up [2]; see Figure 3. The detection of the acoustic pulses essentially builds on the set-up of a conoscope, an optical instrument designed to study surfaces by exciting interference patterns in (visible) light. In this application, an objective lens focuses light onto a sapphire crystal, which exhibits two different refractive indices (birefringence).

Figure 3 shows the Delft set-up with two femtosecond erbium laser systems: a 1,560-nm laser pump and a frequency-doubled 780-nm probe laser. The drawing shows how the probe beam is expanded to an appropriate size and passed through a half-wave plate HWP to achieve the correct polarisation for the polarising beam splitter PBS. A quarter-wave plate QWP then shifts the polarisation into an elliptical state, which is required to exploit the birefringence.

After passing a dichroic mirror DM, where the pump path joins the probe path, the two beams are focused on the sample through a sapphire plate SP. The pump beam is absorbed by the sample to generate acoustic waves in the sample and a part of the probe pulse, modulated by the acoustic echo signal, is reflected. The conoscopic interferometry takes care of the detection of the reflected probe pulse: the quarter-wave plate QWP restores the linear polarisation, while PBS filtering then leaves one polarisation direction to hit the photodetector.

The silicon-based photodetector has a bandwidth of 250 MHz and is able to detect individual probe pulses. The ultimate aim is to reach vertical nanometer accuracies in opaque materials. To demonstrate the performance, the Verbiest team simultaneously developed another application example; see the text box on the next page.

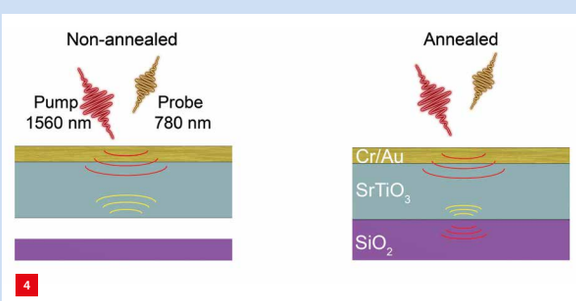


Demonstration

In Delft, photoacoustics have also been applied to improve pressure sensors for MEMS technology (microelectro-mechanical systems) [3]. The challenge was to find ultrathin membranes that show absolutely no gas leakage. 2D sheets of graphene (a regularly arranged configuration of carbon atoms, with one-atom thickness) promised to be ideal for this application, but have not been available in a sufficiently convenient format up until now.

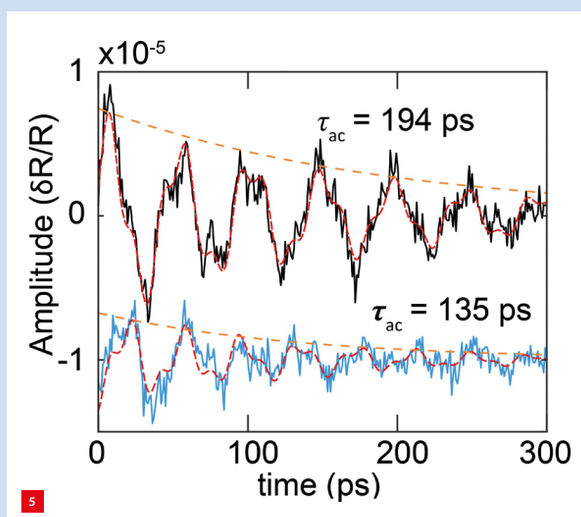
Figure 4 shows how Verbiest tested the adhesion of a new pseudo-2D material: free-standing $SrTiO_3$ (STO, for short). STO is mechanically robust, just like graphene, and can be produced easily in large sheets using epitaxial crystalline growth, which reduces the risk of gas leakage. However, similar to the traditional 2D materials, free-standing STO has a weak (non-bonded) van der Waals interaction with the substrate causing gases to leak along the interface. For microscale pressure-sensing applications using 2D materials, this leakage is a major problem. Better adhesion between the 2D material and the substrate was expected to improve the resistance against leakage.

This schematic figure shows how ultrafast acoustic pulses can be applied to measure the adhesion of STO to a substrate of SiO_2-covered Si. Moreover, the set-up was used to investigate the effect of annealing of the STO. Did it help to improve the resistance to leaking? Yes, because it appeared that annealing causes the forming of extra oxygen combinations between STO and SiO_2 molecules, such that they 'share' oxygen atoms.



Cross-sectional illustration of pump-probe measurements in a non-annealed $SrTiO_3$ (STO) sample on the left and an annealed STO sample on the right. Here, the STO layer measures 82 nm in thickness, while the gold layer on top measures 33 nm.

Figure 5 shows the results of the acoustic measurements: amplitude-versus-time curves for different annealing conditions. Without going into greater details of the meaning of the various curves, the meaning of the general trend highlighted by the dashed lines can be explained as follows. The red dashed curves are fitted to a superposition of damped sine functions and the orange ones depict the exponential decay. This exponential decay is a measure for the energy loss of the acoustic waves at the STO-SiO_2 interface, which strongly depends on the adhesion. From the exponential decay time τ_{AC}, this adhesion can be quantified, with the decrease in Figure 5 showing that the annealing greatly improves the bonding between the STO and SiO_2.

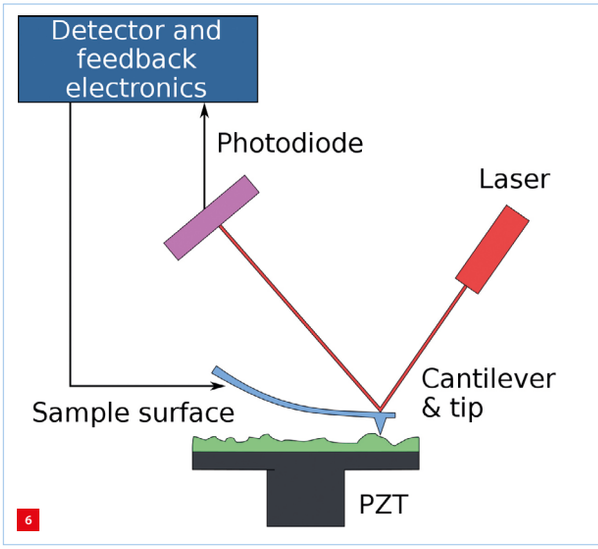


Graphical output of measurements with the set-up of Figure 4, showing the effects of the sound waves reflected from the STO-SiO_2 interface and optically sampled at the top surface of the sample. The black curve is for the non-annealed sample, the blue one for the annealed sample. See the text for further explanation.

Atomic force microscopy

Ultimately, the research resulted in the practical realisation of a vertical (*Z*-direction) nanometer-resolution opto-acoustic material inspection tool. Nevertheless, a solution still had to be found to reach lateral nanometer accuracy. It was not necessary to find a fresh, new approach to this problem because lateral nanometer accuracy can already be achieved with an atomic force microscope (AFM).

The resolution of an AFM as realised in practice, enabled by accurate piezo-electrical drives in the lateral *X*- and *Y*-directions, depends on the dimensions of an extremely sharp cantilever tip. Its radius amounts to only a few nanometers. In the AFM, an optical beam deflection method can be applied to maintain a constant distance between the tip and the surface of a sample, see Figure 5. In this set-up, the tip is mounted at the end of a beam of which the nanometer displacements are detected with



Schematic drawing of a cantilever-based AFM. PZT (lead-zirconate titanate) is a piezoelectric ceramic widely used for nanopositioning (in this case, in the lateral directions). (Source: Wikipedia / OverlordQ).

a four-quadrant photodetector measurement system. This results in a nanometer-resolution height map of the sample surface.

Integration of an AFM into the picosecond ultrasonics set-up of Figure 3 means that the sample is replaced by an AFM cantilever system, as shown in Figure 6; see also [4]. The backside of the cantilever tip is then simultaneously illuminated by a laser for the conventional AFM optical beam deflection measurement and the pump and probe pulses for photoacoustic measurement. Thus, the integration enables a two-stage probing process: an AFM tip probes the 'real' sample, in either contact or oscillating non-contact mode, while the picosecond ultrasonics set-up probes the acoustic reflections coming from the tip and the sample. Further details of this combination of interferometry and optical beam deflection cannot be provided here.

To conclude

Looking into non-transparent materials appeared to be an absolute impossibility in the realm of nanotechnology. But Gerard Verbiest and his Delft team are now set to make it reality by building on the 'old' expertise of photoacoustics as explored by the famous 19th-century inventor Bell. By combining the high vertical resolution enabled by photoacoustics with the high lateral AFM resolution, they are developing a new nanometer-resolution inspection tool for which novel applications can be explored.

REFERENCES

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