Surface waves in highly ordered poly-graphite and gold micro-layers studied by picosecond ultrasonic technique

N Chigarev¹, T Dehoux¹, C Rossignol¹, B Audoin¹ and V Levin²
¹Université Bordeaux 1 ; CNRS ; UMR 5469, Laboratoire de Mécanique Physique, F- 33405 Talence, France
²Laboratory of Acoustic Microscopy, Institute of Biochemical Physics, Russian Academy of sciences, Moscow, 119991 Russia

Abstract. Surface waves propagation in micro-layers is very useful for the evaluation of mechanical properties in anisotropic materials. In the present work, they are detected without any transducer by means of beam distortion detection technique in gold and highly ordered poly-graphite (HOPG) micro-layers. The surface wave velocity in HOPG is evaluated as 20 km/s, value close to Rayleigh wave velocity in diamond. The typical frequency (~10 GHz) of surface wave in HOPG is extremely high. Fast diffusion of electrons plays an important role in the process of sound excitation in gold. For this reason, a two-temperature model has been applied to describe the three-dimensional thermoelastic generation of bulk and surface acoustic waves. Good coincidence between experimental and calculated wavefronts is obtained.

1. Introduction
Compared to traditional techniques based on generation and detection of ultrasound by piezo-transducers, picosecond ultrasonic [1] offers many advantages. It is contactless and allows measuring a very broadband acoustic spectrum. For these reasons, the scope of application provided by picosecond ultrasonics broadens increasingly. Since long time, attention has been attracted to the study of the propagation of bulk waves in solid state medium. Meanwhile, a set of interesting applications related to the non-destructive control and testing has been afforded by the use of surface waves. Some of them are related to the focusing of surface phonons [2], which were first imaged at ~1 GHz in several crystals [3]. The use of a sagnac interferometer allows detection of surface displacement due to propagation of the surface wave. In this paper another method to achieve such measurements is proposed. Beam distortion detection (BDD) technique [4] permits simplifying pump-probe experimental set-up. It is based on the detection of a change in the beam wavefront due to photoinduced curvature of the sample surface. This sensitive and stable technique does not require any stabilization system to measure surface displacement. The detection of surface and bulk phonons in gold micrometric layers and highly-ordered polygraphite (HOPG) has been performed using BDD technique. Opaque for visible and infrared light, HOPG is a rather popular material for the preparation of X-Ray monochrometers [5].

2. Experimental set-up
The pump-probe experimental set-up has been designed on the basis of a Ti:Sapphire femtosecond laser (Figure 1). The radiation of this laser is split into two beams. The pump beam passes through an acousto-optical modulator (~300kHz) to provide the reference signal for lock-in amplification. Radiation of probe at 795 nm passes through an optical delay line operating in the range 0-6ns. A x100 microscope objective is used to focalize the beams on the surface of the sample. To reduce the size of
the spot on the surface, the pump radiation is doubled in a BBO crystal. The position of the pump spot on the surface is scanned according to the probe position by means of a system of two lenses [6]. To be sensitive to the displacement of the surface, an iris diaphragm is introduced in the probe beam after its reflection on the surface of the sample (Figure 1). The aperture of the diaphragm is tuneable in the range 0.2-16mm. It is centred according to the maximal transmission of the probe beam. Finally, the probe beam is focused by a lens on a detection photodiode.

3. Experimental results

Micrometric films of HOPG and gold have been studied. The measured wavefronts of surface wave in HOPG are shown in figure 2. Slow temperature decay due to the cooling of the lattice has been removed from the signals for comparison. Typical duration of the surface wave pulse corresponds to a frequency of \( \sim 11 \) GHz. This is a very high frequency, ten times superior to typical values [3]. According to the time of propagation and the scanning distance, the velocity of the surface wave is \( \approx 20 \) km/s. This very high value shows the excellent rigidity and quality of the sample.

Gold films of thicknesses 1.2 and 2.1 \( \mu \)m sputtered on silicon substrates are used. To verify the model of sound generation, an experiment with large laser spots (~10 \( \mu \)m) is first performed. The
measured signal (Figure 3(3)) consists in the first negative electron transition peak and slow thermal decay. In addition, a set of longitudinal echoes arises from the reflections on the boundaries of the film. The shape of the displacement echoes is unipolar, which means there is no influence of diffraction. To study the influence of the laser spot size on the shape of surface and longitudinal acoustic pulses, a 2.1 μm gold film is now used. Using sharp focusing x100 objective, the pump and probe beams are focused on the surface of the film to a spot size ~1 μm. The shape of the displacement wavefronts at epicentre and for a pump-probe distance 2 μm is shown in figure 4 (A) and (B) curve 3, respectively. First and second longitudinal echoes are clearly visible. The shape of the echoes is twopolar, which is a signature of acoustic diffraction. In addition, the displacement caused by the surface wave can be observed in curve 3, figure 4 (B) corresponding to slow vibration with a maximum of the amplitude at ~1.3 ns. Its duration is estimated by τS~1 ns, and its velocity is taken at vR≈1.3 km/s, which is close to the value for shear wave in gold. Then, the length vR×τS~1.3 μm is close to size of beams waists cross-correlation. Therefore, this low frequency oscillation corresponds to the surface wave on the gold film.

4. Theoretical model

As the electronic diffusion is important in gold, the evolution of the electronic temperature Te is distinguished from the one of the lattice Tl. They are described by a two-temperatures model [8]:

\[ C_e \frac{\partial T_e}{\partial t} = \nabla \left( (2)k_e \nabla T_e \right) - g(T_e - T_l) + Q \]
\[ C_l \frac{\partial T_l}{\partial t} = g(T_e - T_l) , \]

where \((2)k_e\), Ce, Cl and g are the tensor of thermal conductivity, the thermal capacities of the electrons and of the lattice, and the electron-phonon coupling constant, respectively. The source term Q describes the absorption of the incident photons by the electrons. To describe the wavefronts, the temperatures Te and Tl are introduced as a source term of the equation of motion:

\[ \rho \frac{\partial^2 \vec{u}}{\partial t^2} = \nabla \left( (4)e \nabla \vec{u} \right) = - (2)\vec{\Lambda} \nabla T_l - (2)\vec{\gamma} C_e \nabla T_e \]

where \(\vec{u}\), \((4)e\), \((2)\vec{\Lambda}\) and \(\rho\) are the acoustic displacement, the stiffness tensor, the rigidity-dilation tensor, and the mass density, respectively. The Grüneisen constant, traditionally defined as a scalar quantity [9], is here introduced as a tensor \((2)\vec{\gamma}\) to represent the acoustic generation by the 3D electronic diffusion. The wavefronts simulated according to this model are in good coincidence with experimental ones (curve 2, figure 3 and figure 4). The description by the model based on a heat diffusion equation is plotted for comparison (curve 1, figure 3 and figure 4). Not surprisingly, the longitudinal echoes are better described by the two-temperature model. An unexpected feature is that the low frequency surface wave is more precisely represented when accounting for the electronic diffusion.

5. Conclusion

Finally, the following points should be underlined:

1) Performed experimental studies in HOPG have shown the existence of surface waves at the impressive frequency of 11 GHz. This could have an application in non-destructive testing of this material. This testing could help to find the cracks and defects of nano- and micrometric
size inside the material. Picosecond ultrasonics allows performing non-destructive control of HOPG without acoustic contact with the sample.

2) Surface and longitudinal wavefronts detected by BDD technique in gold micro-layers are described quite precisely in the frame of the two-temperature model. For the first time this model has been extended to 3D configurations.

![Figure 4](image)

**Figure 4.** Displacement echoes in 2.1 μm gold film at the epicenter (A) and at 2 μm distance from the epicenter (B). 1: parabolic model; 2: two-temperature model; 3: experiment. 2L and 4L: first and second longitudinal echoes, respectively.

**Acknowledgments**

Authors are grateful to T. Pezeril (Université du Maine) for helping in the preparation of micrometric gold layers.

**References**

[1] Thomsen C, Grahn H T, Maris H J and Tauc J 1986 *Phys. Rev. B* **34** 4129
[2] Every A G 1986 *Phys. Rev. B* **33** 2719
[3] Sugawara Y, Wright O B, Matsuda O, Takigahira M, Tanaka Y, Tamura S and Gusev V E 2002 *Phys. Rev. Lett.* **88** 185504
[4] Chigarev N, Rossignol C and Audoin B 2006 *Rev. Sci. Instrum.* **77** 114901
[5] Grigorieva I G and Antonov A A 2003 *X-Ray Spectrometry* **32** 64
[6] Tachizaki T, Muroya T, Matsuda O, Sugawara Y, Hurley D H and Wright O B 2006 *Rev. Sci. Inst.* **77** 043713
[7] Every A G and McCurdy A 1992 Numerical data and fundamental relationships in science and technology (New Series, Group III of Landolt-Burnstein) vol. 29A (Berlin: Springer)
[8] Anisimov S, Kapilevitch S B and Perelman T 1975 *Sov. Phys. JETP* **39** 375
[9] Wright O B 1994 *Phys. Rev. B* **49** 9985