Picosecond ultrasonics on a single micron carbon fiber

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Abstract. A pump-probe picosecond technique is used to reveal the opto-acoustic response of micrometric carbon fibers. Two carbon fibers having the hexagonal symmetry are experimentally studied. A dedicated experimental setup allows to generate acoustic waves that propagate in a cross section of the fiber. The elastic properties in the transverse direction of a single carbon fiber are thus evaluated. Complex optical refractive index is also measured. The results provide interesting perspectives for the non-destructive evaluation of elastic and optical properties at a micron scale.

1. Introduction
Carbon reinforced composites are widely used materials in various fields such as transport, civil engineering, sports, etc. Carbon fibers are of particular interest owing to their outstanding strength to weight ratio. It is mainly composed of carbon atoms forming microscopic crystals that are more or less aligned with the fiber axis, giving the fiber a hexagonal symmetry.[1] Such carbon fibers have diameters of only a few microns. They are assembled in millimetric yarns before weaving the carbon plies that are finally embedded in a matrix. Usually, manufacturers measure the mechanical properties of those millimetric sized yarns by a traction test. At the same time, the knowledge of these properties at the scale of a single fiber is of strong interest for a better understanding of the micromechanical behaviour of composite materials. A recent work reports experimental results of a micro-traction test on a single micron carbon fiber assessing the elasticity in the direction of its axis.[1] The elasticity in a normal direction to the fiber axis has not yet been investigated essentially due to the experimental difficulties. We propose the first application of the picosecond ultrasonics technique [2] to generate and detect high frequency acoustic waves in such a single micron carbon fiber. Owing to its non contact generation and detection process, this optical technique is particularly well suited for the non-destructive evaluation of cylindrical shaped targets. A description of the experimental setup used in this study is first given; then its application to the non-destructive evaluation of the elastic and optical properties of carbon fibers is detailed.

2. Experimental setup and samples
The pump-probe picosecond ultrasonics technique has been successfully applied since the end of the 1980s to analyze plate structures with sub-micrometric thicknesses.[2] The absorption of a subpicosecond laser pulse (pump) suddenly heats the surface material that releases the incoming
energy through a picosecond acoustic pulse propagating back and forth in the sample. A second light pulse (probe) is used to detect the change of the optical reflectivity of the sample caused by the propagating transient strain pulses. This optical technique allows non-contact acoustic generation and detection with very high resolution in time and space, making it particularly well suited for the non-destructive evaluation of cylindrical targets with micron diameters.

Two samples made of carbon fibers are used in this study. A high elastic modulus UMS40 carbon fiber (from Tenax Co.) made from a polyacrylonitrile precursor (ex-PAN) and a low elastic modulus XN05 carbon fiber (from Nippon Graphite Fiber Co.) produced from an isotropic pitch precursor also called an ex-brai precursor. These fibers have respectively a mean diameter of $d = 5 \, \mu m$ and $d = 10 \, \mu m$ and show a very regular cylindrical section and a small surface roughness. A small piece of a given fiber of one millimeter length is stretched at its two ends in a dedicated experimental setup to avoid free vibrations.

Experiments are performed with a conventional picosecond ultrasonics setup.[2, 3] Pump and probe beams are collinearly focused on the sample surface by the same objective with a 50X magnification and a high numerical aperture NA = 0.8. The widths of the pump and probe beams on the surface of the fibers are estimated to be 1.5 $\mu m$ and 3 $\mu m$, respectively.

3. Experimental results and characterization of the carbon fibers

3.1. Elastic and acoustic properties

Transient reflectivity changes measured on the UMS40 and the XN05 carbon fibers are shown in Fig. 1 (left) and Fig. 1 (right) respectively. The absorption of light creates a sudden temperature rise followed by a low thermal decay. Acoustic signal is then superimposed at corresponding times for which the reflected acoustic wavefronts reach the probed surface. The thermal background is removed to emphasize the acoustic signature of the signal as shown in Fig. 1 (right) for the XN05 fiber, where two acoustic echoes are detected corresponding to successive reflections of the longitudinal acoustic wave at the free surface of the sample. These echoes are shown with more details in the insets in Fig. 1 (right). Their dispersive feature appears clearly, with the low frequency components, about 15 GHz, of the transient echoes arriving earlier to the probed surface than the high frequency components. This negative dispersion cannot be due

![Figure 1](image.png)

**Figure 1.** Reflectivity change for the UMS40 carbon fiber of 5 $\mu m$ mean diameter (left). A sudden change in the reflectivity of the sample is followed by a slow thermal decay. The thermal background is subtracted to reveal the acoustic content measured for the XN05 fiber (right). The two upper insets focus on each acoustic echo.
to the structural relaxation (e.g. caused by visco-elasticity), since then the dispersion would be opposite, with high frequencies propagating faster than low frequencies.[4] This phenomenon results actually from the intrinsic geometric scattering of the acoustic waves in the cylindrical cavity.[3]

The acoustic displacements generated by the absorption of a laser pulse along a radial direction in a cylinder have been predicted theoretically [5] and have permitted to successfully describe experimental results observed at the surface of a glass cylinder with a diameter of 5 mm.[6] The wavelength of the bulk acoustic waves approximately equals the optical penetration depth. In the case of carbon fibers, acoustic diffraction is weak since the acoustic wavelength ($\sim 0.2 \mu m$) is small with respect to the source width, and hence only bulk waves with a longitudinal polarization are detected. Their successive arrivals are observed at the times $\tau_{2L} = 4.65 ns$ and $\tau_{4L} = 9.3 ns$ for the XN05 fiber whereas only one echo has been observed for the UMS40 at 3.4 ns. From the round trip of the acoustic pulses we find a sound velocity $c_L$ in the cross section equal to $4.3 \pm 0.4 \text{ km s}^{-1}$ and $2.9 \pm 0.3 \text{ km s}^{-1}$ for the XN05 and UMS40 fibers, respectively. The mean diameters $d$ of 10 $\mu m$ are provided by the manufacturer with standard deviation of 1 $\mu m$.[7] The elastic coefficients $c_{11}$ can be evaluated as follows: $c_{11} = \rho c_L^2 = 30 \pm 6 \text{ GPa}$ (where the density is $\rho = 1.65 \text{ g cm}^{-3}$) for the XN05 fiber and $c_{11} = 15 \pm 3 \text{ GPa}$ (where $\rho = 1.79 \text{ g cm}^{-3}$) for the UMS40 fiber. These elastic properties are in the same range of order whereas their elastic moduli in the axial direction are drastically different: $E = 53 \text{ GPa}$ and $E = 380 \text{ GPa}$, respectively.[1] This can be understood from the microstructure of such carbon fibers. The XN05 fiber is made from an ex-braided isotropic pitch that leads to a low-modulus fiber in the axial direction with a weak anisotropy between axial and transverse directions. This is essentially due to axial traction forces during the graphitization process. The UMS40 fiber, made from a PAN pitch precursor, is a highly anisotropic carbon fiber where microcrystals, made of graphene sheets, are preferentially aligned in the axial direction leading to a high-modulus fiber.[8] Acoustic attenuation can also be measured for the XN05 fiber from the amplitudes of the two successive echoes.[3]

3.2. Optical properties

Interest is now focused on the Brillouin oscillations detected in the first picoseconds, Fig. 2. The transient reflectivity changes are sensitive to the modulation of the optical properties created by the in-depth temperature rise and strain. Part of the probe light penetrates into the material and is scattered by acoustic phonons provided that their wave number $k = 2q \cos \theta$, where $q$ stands for the probe light wave number and $\theta$ is the angle between the two wave vectors. The

Figure 2. Brillouin oscillations observed in the first picoseconds (solid lines), with frequencies of 16.7 GHz and 20 GHz for UMS40 (left) and XN05 (right) fibers, respectively. The thermal background has been subtracted. The corresponding curve fitting (dashed lines) is also reported.
scattered light interferes with the part of the beam reflected at the sample surface giving rise to the so-called Brillouin oscillations. Their frequency is

\[ f_B(\theta) = 2 n c_L \cos(\theta)/\lambda, \]

where \( \lambda \) is the wavelength of the probe light and \( c_L \) is the sound velocity. Using Eq. (1), the real part of the optical refractive index \( n \) at a wavelength of 796 nm can now be deduced from \( f_B(0) \) at normal incidence of the probe beam: \( n=1.9 \pm 0.2 \) and \( n=2.3 \pm 0.2 \) for the XN05 and the UMS40 fibers respectively. In addition, the lifetime of the Brillouin oscillations can be the result of either acoustic or optical absorption. Since the acoustic echoes are detected after propagation along a long distance of 40 and 10 \( \mu m \) for the XN05 and UMS40 fibers respectively, the decrease of the first Brillouin oscillations comes mainly from the optical absorption. The experimental signal can be fitted by a damped oscillation function. The value of the optical penetration depth at 796 nm, defined as the inverse of the absorption coefficient (\( \alpha^{-1} \)), can then be calculated using \( \xi = C\tau \), giving \( \xi = 91 \pm 10 \) nm and \( \xi = 94 \pm 10 \) nm for the XN05 and UMS40 fibers, respectively. It yields the values for the imaginary part of the complex refractive index of the fiber as \( k = 0.7 \) and \( k = 0.6 \) for the XN05 and UMS40 carbon fibers, respectively. The measured refractive index for the carbon fibers compares well with the complex refractive index \( \tilde{n} \) given in [9] for graphite at 800 nm, \( \tilde{n} = 2.6 + 1.5i \). The measured physical parameters are reported in Tab.1.

| fiber   | \( d \ (\mu m) \) | \( \rho \ (g \ cm^{-3}) \) | \( c_L \ (\text{cm} \ s^{-1}) \) | \( c_{11} \ (\text{GPa}) \) | \( \alpha^{-1} \ (\text{nm}) \) | \( n \) | \( k \) |
|---------|-------------------|-----------------------------|-------------------------------|--------------------------------|-------------------------------|-------|-------|
| XN05    | 10                | 1.65                        | 4.3 \( \pm \) 0.4             | 30 \( \pm \) 6                | 91 \( \pm \) 10              | 1.9 \( \pm \) 0.2 | 0.7 \( \pm \) 0.07 |
| UMS40   | 5                 | 1.79                        | 2.9 \( \pm \) 0.3             | 15 \( \pm \) 3                | 94 \( \pm \) 10              | 2.3 \( \pm \) 0.2 | 0.6 \( \pm \) 0.06 |

Table 1. Physical parameters for the XN05 and UMS40 carbon fibers: the mean diameter \( d \), the longitudinal sound velocity \( c_L \), the elastic constant \( c_{11} \), the optical penetration length \( \alpha^{-1} \) at 796 nm, and the real and imaginary parts \( n \) and \( k \) of the refractive index.

4. Conclusion

The picosecond ultrasonics technique has been applied to the non-destructive evaluation of two carbon fibers of 5 and 10 \( \mu m \) diameter. Detection of the acoustic echoes propagating back and forth through the fiber cross section has led to measurement of the fiber mechanical properties. In consequence, theses data complements tensile tests that provide the elasticity only in the axial direction of the fiber. In addition, the complex optical refractive index has been measured locally. Such measurements may find interesting applications for the non-destructive evaluation of elastic anisotropy at a micron scale.

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