Femtosecond pump-probe spectroscopy has been carried out on suspended gold nanostructures with a rectangular cross section lithographed on a silicon substrate. With a thickness fixed to 110 nm and a width ranging from 200 nm to 800 nm, size dependent measurements are used to distinguish which confined acoustic modes are detected. Furthermore, in order to avoid any ambiguity due to the measurement uncertainties on both the frequency and size, pump and probe beams are also spatially shifted to detect guided acoustic phonons. This leads us to the observation of backward propagating acoustic phonons in the gigahertz range (∼3 GHz) in such nanostructures. While backward wave propagation in elastic waveguides has been predicted and already observed at the macroscale, very few studies have been done at the nanoscale. Here, we show that these backward waves can be used as the unique signature of the width dilatational acoustic mode.

Backward Propagating Acoustic Waves in Single Gold Nanobeams

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We use time-resolved pump-probe spectroscopy with spatially shifted pump and probe beams to excite and detect the backward wave motion. As a first application, unambiguous mode identification using backward wave propagation is investigated.

In order to study guided modes in one dimensional elastic waveguides, gold nanostructures with a rectangular cross section are conceived (Fig.1). Their thickness, $h$, is fixed to $h = 110\, \text{nm}$ and their width, $W$, ranges from 200 to 800 nm. As shown in Fig.1(a), the gold stripes are connected to large mesas at both ends. To reduce the influence of the silicon substrate on the elastic confinement, the gold nanostructures are suspended several micrometers above the substrate as illustrated in Fig.1(b). This geometry results in what we call “gold nanobeams”. All boundaries are thus mechanically free. As their length is greater than 15 μm, they are considered as infinitely long and the fixed boundary conditions at the extremities can be neglected. The samples are fabricated by electronic lithography coupled with anisotropic wet etching on silicon substrates. To prepare these structures, single face polished Si(001) substrates are used. First, an electronic resist (PMMA-950K-A6) is spin coated on the Si wafer surface. The exposition is made at 20kV using a Zeiss Supra 40 scanning electron microscope equipped with a Raith Elphy Quantum module. After being developed, rinsed with water and blown dry, the sample is coated with 10 nm of chromium to improve the adhesion, followed by a 110 nm thick gold layer. Then, a lift-off process in acetone with careful ultrasonic agitation is performed to delimit the future gold nanobeams. Finally, the sample is dipped in a hot 40% KOH solution.
to partially etch the Si substrate and free the desired gold nanobeams. This quick anisotropic Si etching process leads to the formation of suspended gold nanobeams between two big gold pads. The key point of this process is that the main axis of the beams has been rotated 45° with respect to the [110] direction of Si substrates to allow this underetching.

Our experimental setup working in reflection geometry was described in detail elsewhere. Ultrafast pump-probe spectroscopy experiments are performed using a mode-locked Ti:sapphire (Mai-Tai Spectra) laser source operating at 800 nm with a pulse duration below 100 fs at the laser output and a repetition rate of 78.8 MHz. Synchronous detection on the sample reflectivity is performed by modulating the pump beam at 1 MHz. Both pump and probe beams are focused by means of a microscope objective with a NA = 0.9 and are normally incident on the sample. The laser spots can be focused around 1 µm diameter at 1/π². A telescope is fixed on a XY piezoelectric stage such that the probe beam is laterally positioned with respect to the fixed pump beam. A two-color experiment is performed by doubling the pump frequency (λ = 400 nm) with a nonlinear crystal (BBO) to avoid scattered light coming from the pump beam. A dichroic filter located in front of the diode system suppresses the light of the pump beam, its power is reduced around 500 µW and the power of the probe beam does not exceed 15 µW.

Such experimental conditions place us in the thermoelastic regime and the acoustic signal and optical reflectivity remain stable during all the average processing. The reflectivity from the sample is measured by an avalanche photodiode and analyzed with a lock-in amplifier. A maximum pump-probe time delay equal to 12 ns is achieved using a mobile reflector system mounted on a translation stage.

First, let us consider the case where the pump and probe beams are spatially superimposed. Fig. 2 shows the time domain reflectivity change measurements ∆R/R obtained on a 400 nm ± 20 nm wide (determined by SEM measurement) and 110 nm thick gold nanobeam in a usual reflectometry set-up. After a sharp and sudden rise, a slowly decreasing non-oscillatory background in ∆R/R is observed. This is the signature of the rapid heating of the electron gas induced by the pump pulse absorption, followed by the slow cooling down process. This thermal stress launches the acoustic vibrations of the nanobeam.[27]. The time-resolved signature also shows a superposition of high (inset Fig.2(a)) and low frequency oscillations. By performing a numerical fast Fourier transform (Fig.2(b)) or by fitting the time oscillation with a damped sinus function, one can extract a 2.8 GHz ± 0.5 GHz and a 16 GHz ± 0.5 GHz acoustic signatures. In the following we consider that the Young modulus, the Poisson’s ratio and the density of gold are respectively 79 GPa, 0.44 and 19 300 kg/m³[27]. The longitudinal $v_L$ and transverse $v_T$ sound velocities are then 3.6 µm ns⁻¹ and 1.2 µm ns⁻¹ respectively. Thus, the higher 16 GHz frequency appears to be the thickness vibration of the beam owing to the fact that $v_L/(2h) = 16.5$ GHz. The displacement field of this $d_2$ eigenmode is plotted in Fig. 3(b). At this point, we verify...
that this thickness resonance signature is also observed when the pump-probe experiment is undertaken on the gold mesa using an interferometric scheme in this case as

FIG. 3. (a) Acoustic modes dispersion relations in a 370 nm wide and 110 nm thick gold nanobeam (with a 10 nm thick adhesion layer of chromium). $f_1$, $s_1$ and $d_1$ refer respectively to flexural, shear and dilatational modes. The inset shows the parabolic fit of three specific modes at low wave number used to evaluate the corresponding propagating wave packet. The parabolic fit is respectively $f_1(k) = -3.4 \times 10^{-2} k^2 - 9.2 \times 10^{-5} k + 3.2$ GHz, $f_2(k) = 5.0 \times 10^{-2} k^2 + 5.0 \times 10^{-4} k + 3.6$ GHz and $f_3(k) = 5.7 \times 10^{-3} k^2 + 2.8 \times 10^{-4} k + 3.4$ GHz for the modes $d_1$, $s_2$ and $f_2$ with the wave number $k$ in $\mu$m$^{-1}$. (b) Cross-sectional representation of the displacement field of three flexural ($f_1$,$f_2$ and $f_3$), two dilatational ($d_1$ and $d_2$) and three shear ($s_1$,$s_2$ and $s_3$) acoustic modes. The black arrows show the displacement direction.

FIG. 4. Using a finite elements method, the acoustic mode frequency at wave number $k=0$ is plotted versus the nanobeam width. Flexural, shear and dilatational modes are respectively the orange dotted lines, the green dashed lines and the blue solid lines. Experimental measurements are the full black circle with bars. The vertical black dashed line at 370 nm is the specific width where dispersion relations have been calculated (Fig.3) and propagation has been observed (Fig.4).

photoelastic signals are negligible in gold layers. Consequently, the reflectometry measurement undertaken on the gold nanobeam is probably partly an interferometric like detection with the light reflected by the silicon substrate underneath acting as a reference mirror.

The case of the lower frequency $\sim 3$ GHz is dealt with by performing an eigenmode analysis by finite elements method. The dispersion relation of a 370 nm wide nanobeam is plotted in Fig.3. Several acoustic modes with non-zero cut-off frequencies are revealed. Three distinct families emerge: the flexural modes $f_1$, $f_2$ and $f_3$, the dilatational modes $d_1$ and $d_2$ and the shear modes $s_1$, $s_2$ and $s_3$ (see Fig.4(b)). The frequency evolution of $s_1$, $s_2$, $f_1$, $f_2$, $f_3$ and $d_1$ with the nanobeam width is plotted in Fig.4. The measured frequencies on different gold nanobeams are also reported. Owing to the measurement uncertainties, the experimental frequencies ranging from 1.7 GHz to 5.5 GHz may be consistent with the frequencies of the modes $f_2$, $s_2$ and $d_1$ which exhibit close cut-off frequencies. Consequently, additional evidence on the detected mode is needed. Given that the excitation process imposes the relative amplitudes of the excited modes, one can use the initial displacement field projection on the orthogonal basis formed by the nano-object eigenmodes to identify which mode will be efficiently excited. Here we propose to discriminate unambiguously between the different acoustic modes by probing their propaga-
The vertical scale corresponds to the pump-probe spatial separation. The exponential thermal background has been removed. The offset corresponds to the spatial pump-probe shift and the reflectivity modulation is normalised by the maximum. The envelope of the theoretical signal 

\[ \Delta r(t) \propto \text{Re} \left( \frac{\exp(-i\gamma t)}{\sqrt{\sigma^2 + 32i\alpha t}} \exp \left( -\frac{8(z_0 - \beta t)^2}{\sigma^2 + 32i\alpha t} \right) \right) \]  

where \( z_0 \) is the spatial pump-probe separation and \( \sigma \) is the root of the sum of squares of the pump and probe beam diameters at 1/e². A polynomial fit at small wavenumber for \( d_1, f_2 \) and \( s_2 \) according to the parabolic fit of the dispersion relation (Fig.5(a)), (e), (f), (g), (h) are respectively the same signals as in (a), (b), (c), (d) zoomed in a shorter time window. Only half of the eight curves are plotted for readability. The offset is also suppressed in order to observe the propagation direction of the phase of the wave. The black arrow indicates this propagation direction.

In conclusion, two dilatational acoustic modes are investigated on e-beam lithographed gold nanobeams by pump-probe time-resolved spectroscopy leading to the observation of backward wave propagation in the gigahertz range. As a first application, we show unambiguous acoustic mode discrimination using this unique property in the dispersion relations. Furthermore, as already observed in 2D Lamb waveguides, one can imagine to tune the width and thickness of our nanobeam, thus opening the possibility to control the guided wave and its dispersion.
the way to negative refraction physics in acoustics at the nanoscale.

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