Picosecond ultrasonic investigations of phonons in 2D nano-scaled lattices

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Abstract. We time-resolved the acoustical response of lattices of aluminum nano-dots with a step of a few hundreds nanometers using tunable femtosecond laser pulses in a pump and probe scheme. We detected two kinds of modes, the first being the individual modes of the dots. The other modes are shown to be both dependent on the dot size and on the lattice and are thus interpreted as collective modes. Using several step sizes we show that we can plot the phonon dispersion relation. A simple analytical model very well reproduces the data from which we can describe completely the dependence of the lattice modes on the sample parameters.

1. Introduction
Acoustic phonons in periodic materials behave like photons in a photonic crystal. Changes due to zone folding occur and affect elastic wave propagation such as band gaps or dispersion [1]. In order to obtain a Hypersonic Phononic material (i.e. beyond 1 GHz) one has to periodize a sample with a few hundreds nanometers scale. This way the sample is simultaneously photonic for visible light and phononic for hypersounds. Many reports on acoustics in nanostructures concern individual vibrations of unordered or isolated particles resolved in the time domain [2, 3, 4, 5] or by light scattering [6]. Other studies adress artificial objects such as metal stripes and report proper modes detection [7, 8, 9]. Finally recent works concern investigations of Phononic Crystals in the frequency domain by Brilouin Light Scattering [10].

Here we report on a time-resolved study of acoustic modes in 2D lattices of nanometric cubes. We demonstrate the coexistence of two kinds of vibrations. The first ones are identified to be those of the individual cube. Other modes present a dependence on the step size and cube width which let us assign them to collective acoustic modes. We study the dispersion and explain it thanks to an analytical model. Finally we discuss their acoustic nature.

2. Samples and experimental details
The 2D lattices of aluminum (Al) cubes are fabricated using electronic lithography. This technique enables designing patterns of various shape and width with a precision down to a few nanometers. This way we obtain defect free samples with well defined dot width(d) and lattice constant (a). The e-beam prints directly in a positive photoresist and the dots are formed by lift-off of an evaporated Al layer. All these lattices lay on an 100 nm thick Al film deposited on a pyrex substrate.
Samples are listed in Table 1. Some samples are made of 50, 100 and 200 nm cubes (samples A, B and C) the cube width and lattice step are proportional. Other samples are whole sets of 200 nm sized cubes with increasing step sizes from 400 to 1400 nm (series D and E). A SEM image of an hexagonal lattice is shown on Figure 1(b).

Table 1. Sample list

| Single square lattices | Cube width (nm) | Lattice step (nm) | Series | Lattice Type | Cube width (nm) | Lattice step (nm) |
|------------------------|----------------|-------------------|--------|--------------|----------------|-------------------|
| A                      | 50             | 100               | D      | Square       | 200            | 400 - 1200        |
| B                      | 100            | 200               | E      | Hexagonal    | 200            | 400 - 1400        |
| C                      | 200            | 400               |        |              |                |                   |

The time-resolved experiments were conducted using a pump and probe setup associated with a tunable Ti:sapphire oscillator. The laser produces 120 fs optical pulses at a repetition rate of 76 MHz. The wavelength is adjustable between 700 nm and 990 nm. The probe can be frequency doubled to produce a blue pulse. All the experiments are performed at room temperature. The maximum fluence of the pump beam is 1 J/m² and the ratio between pump and probe intensities is about 1000:1. A sketch of the experimental setup is shown on Figure 1. The focus sizes of both pump and probe beams are large (50 µm) compared to the lattice constant of the sample so that a large assembly of dots is excited coherently.

3. Individual modes of the dots

We first focus on samples A, B and C using both infrared pump and probe (905 nm). The transient reflectivity, shown on Figure 2(a) exhibit several components. First, at the zero delay a sudden change in the reflectivity occurs. Then the initial heat decays exponentially on a hundred ps time scale which can be seen on Figure 2(a). The interesting contribution here is the oscillating part resulting from the modulation of reflectivity by the vibration of the dots. This oscillation last on a hundred picosecond time scale. On Figure 2(b) we notice that the period of this signal is proportional to the cube width which is consistent with an acoustic origin. Performing the same experiment on the whole D set of samples gave the same frequency. Which establishes that the signal measured here originates from the dot but is not dependent on the lattice constant. These first acoustic contributions are associated to the individual vibrations of the cubes.
The detection of these modes uses the photo-elastic constants of aluminum. Indeed we performed the same experiment at a wavelength of 800 nm and observed a sign change in the contribution of the individual modes. This fact was previously reported as a cancellation of the photo-elastic couplings around the interband transition of Al at 850 nm [11].

4. Collective modes of the lattice

If we now test the sample C with a blue probe (400 nm) the transient reflectivity is much different (Figure 3(a)). The oscillating part of the signal is quite larger and less damped, the time scale is of a few nanoseconds. Furthermore as shown on Figure 3(b) the frequency content is much lower. Two sharp peaks dominate the spectrum which correspond to the long lived oscillations. A smaller pattern at 20 GHz is similar to the signal obtained with an infrared probe, the individual mode. We studied the whole sets of samples E with a blue probe. For each step size we retrieved this vibration at 20 GHz corresponding to the individual mode. New lower frequencies are observed whenever the lattice step is shorter than the probe wavelength. Contrary to the individual modes they strongly vary with the lattice constant.

On Figure 4(b) we plot these lower frequencies as a function of the inverse of the lattice parameter for the D (square lattice) series. First one notices that the experimental points order very well as branches which reach zero for infinite lattice constant. Thus this contribution does not exist for a free surface without dots. In the extent \( d/a \ll 1 \) the curves are linear and the relation between the frequency and the step size reduces to \( f = c/a \). Where \( c \) is a sound velocity (3.07 nm/ps) very close to the Rayleigh wave velocity in Al\((c_R = 3.01 \text{ nm/ps} \ [12])\). Which proves the acoustical nature of these modes and suggests a similarity with surface waves.

For experimental values of \( d/a \) the branches exhibit a large curvature which indicates that the increasing dots density affects the lower frequencies modes. We assign these signal which depend
on both the dots width and the lattice constant to collective vibrations resulting from the dots. This fact is well supported by a model plotted as continuous lines on Figure 4(b). In this model we consider the base cell of the lattice as an harmonic oscillator. The frequency of the oscillator depends on the lattice step and reproduces the branches curvature [13].

The ratio between the two branches for the square lattices (D series) is equal to $\sqrt{2}$. This noticeable factor manifests that the different branches result from the excitation of the same collective mode along the surface in different directions [(1,0) and (1,1) in this case]. To confirm this point we made the same measurements on hexagonal lattices (E series) shown on Figure 4(b). In that case the ratio between branches equals $\sqrt{3}$ as expected in the hexagonal geometry. These results show that the collective modes of the dots propagate at the surface of the sample in the crystal directions.

5. Conclusion
We show in the time domain the simultaneous existence of collective modes and individual vibrations of metallic dots. First the individual modes of the cubes are detected via a photoelastic mechanism. Second we explored the dispersion of collective vibrations. This revealed their propagation at the surface. The optical detection of these last is original due to the role played by the probe wavelength. It could be linked to the peculiar optical properties of nano-periodized metallic surfaces.

References
[1] Brillouin L 1953 Wave Propagation in Periodic Structures (Dover publications)
[2] Nisoli M, De Silvestri S, Cavalleri A, Malvezzi A M, Stella A, Lanzani G, Cheyssac P and Kofman R 1997 Phys. Rev. B 55 R13424–R13427
[3] Voisin C, Christofilos D, Fatti N D and Vallée F 2002 Physica B 316-317 89
[4] VanDijk M A, Lippitz M and Orrit M 2005 Phys. Rev. Lett. 95 267406
[5] Petrova H, Lin C H, de Lieger S, Hu M, McLellan J M, Siekkinen A R, Wiley B J, Marquez M, Xia Y, Sader J E and Hartland G V 2007 The Journal of Chemical Physics 126 094709
[6] Lim H S, Kuok M H, Ng S C and Wang Z K 2004 Appl. Phys. Lett. 84 4182–4184
[7] Lin H N, Maris H J, Freund L B, Lee K Y, Luhn H and Kern D P 1993 Journal of Applied Physics 73 37–45
[8] Antonelli G A, Maris H J, Malhotra S G and Harper J M E 2002 Journal of Applied Physics 91 3261–3267
[9] Bienvill T, Robillard J, Belliard L, Roch-Jeune I, Devos A and Perrin B 2006 Ultrasonics 44 1289–1294
[10] Gorishnyy T, Ullal C K, Maldovan M, Fytas G and Thomas E L 2005 Phys. Rev. Lett. 94 115501
[11] Devos A and Louarn A L 2003 Phys. Rev. B 68 045405
[12] Anderson O L 1965 Physical Acoustics vol 3B ed Mason W P (Academic, New York) pp 43–95
[13] Robillard J F, Devos A and Roch-Jeune I 2007 Physical Review B 76 092301