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Nonlinear ultrafast acoustics at the nano scale

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ABSTRACT

Pulsed femtosecond lasers can generate acoustic pulses propagating in solids while displaying either diffraction, attenuation, nonlinearity and/or dispersion. When acoustic attenuation and diffraction are negligible, shock waves or solitons can form during propagation. Both wave types are phonon wavepackets with characteristic length scales as short as a few nanometer. Hence, they are well suited for acoustic characterization and manipulation of materials on both ultrafast and ultrashort scales. This work presents an overview of nonlinear ultrasonics since its first experimental demonstration at the beginning of this century to the more recent developments. We start by reviewing the main properties of nonlinear ultrafast acoustic propagation based on the underlying equations. Then we show various results obtained by different groups around the world with an emphasis on recent work. Current issues and directions of future research are discussed.

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1. Introduction

Whenever light cannot enter matter, sound becomes the first choice for imaging buried structures. Megahertz ultrasound techniques have been extensively studied in the past and gave birth to numerous applications such as sonography and nondestructive testing with a resolution of the order of a few micron.

The development of picosecond and femtosecond pulsed lasers in the mid-80's [1] opened the way to all-optical pump probe techniques to study acoustic waves. The archetypal experimental arrangement Fig. 1(a) is nowadays referred to as *picosecond ultrasonics* [2] or *ultrafast acoustics*. A short optical 'pump' pulse heats a (generally metallic) transducer material. Through the thermoelastic effect (the heated material exerts a sudden stress to its surroundings), a coherent longitudinal strain pulse $\eta(z,t)$ is formed. The acoustic wave reflects at the transducer boundaries, locally modifying the optical properties. Such modifications are measured by a delayed optical 'probe' pulse. For a review of developments in detection techniques, we refer to another contribution in this issue.

This technique allows the generation of acoustic pulses as short as a few picosecond, with a bandwidth typically 100 GHz, at strain amplitudes of the order of 10^{-5}–10^{-4}. The principle was first demonstrated and theoretically analyzed for metal films and semiconductors [3,4,1]. Since the sound velocity in semiconductors and metals is typically several nm/ps, picosecond acoustic pulses extend spatially up to several tens of nanometers.

In standard ultrafast acoustics relying on the thermoelastic effect, the range of accessible phonon frequencies is determined by the properties of the generator material, and is limited by either the optical penetration depth or the electron diffusion length (the typical depth over which heat is deposited) [5,6].

To really extend application of ultrasound to the study and manipulation of nanostructures, acoustic frequencies in the 100 GHz–1 THz range are required; an order of magnitude higher than generated by conventional techniques. However, very few techniques can generate such acoustic frequencies. They are mainly based on transient optical excitation by laser pulses.

In some cases, THz frequencies have been generated by optically exciting vibrational modes of a very thin single layer or carefully designed multilayers [7–12]. These heterostructures allow some control over the properties of the generated coherent wave [13]. In strained quantum well structures where strain is generated through piezoelectric screening, tunable phonon generation with a bandwidth of several hundreds of GHz has been demonstrated [14,15], with absolute strain amplitudes up to 2% [16]. A recent paper has shown that by generation in the semiconductor itself broad, and high-amplitude, tunable strain waves can be generated [17]. More information may be found in other contributions in this special issue.

High acoustic frequencies can also be reached by using the intrinsic acoustic nonlinearity of crystals. This does not require the design of specific transducers and (in sharp contrast with nanostructured transducers) results in a broadband acoustic spectrum. We report in this paper on *nonlinear ultrasonics*, the nonlinear propagation of acoustic pulses in lattices, in the framework of femtosecond laser ultrasonics.

Section 2 is an introduction to the propagation equation in anisotropic solids which includes dispersion, diffraction, viscous damping and nonlinearity. The equation is rather complex and impractical to solve mainly because of its tensorial nature. However, making some simplifying assumptions relevant to most nonlinear ultrasonic experiments, the propagation equation is rewritten into an analytical form which we will label the cylindrical Korteweg–de Vries–Burgers equation (cKdVB).

In the remainder of the paper, we will discuss specific regimes, in which one or more of the terms in the cKdVB equation can be neglected. Section 3 treats the linear regime, where nonlinear effects are negligible, to illustrate the effects of the three linear terms (dispersion, diffraction, damping) in the governing equation.

Nonlinearity modifies the acoustic pulse shape and leads to frequency upconversion. Section 5 is devoted to the case when dispersion and diffraction are negligible, and acoustic shock waves are formed, see Fig. 1(b) [18,19].

The group of H.J. Maris was the first to demonstrate the nonlinear propagation of picosecond ultrasonic pulses [20] in SiO_2, Si, Al_2O_3, and MgO in 2001, leading to acoustic wavelengths shorter than the typical wavelength at generation. When nonlinearity is counterbalanced by acoustic dispersion, acoustic *solitons* can form, see Fig. 1(c). These solitons have intriguing properties. Once formed, they show a remarkable resilience to deformation and attenuation, for example by energy transfer to an electronic two-level system [21] or by anharmonic phonon decay [22], and propagate unperturbed over distances as large as several mm [23]. The group of J.I. Dijkhuis has demonstrated the existence of a 0.87 THz component in the spectrum of the acoustic pulse after nonlinear propagation in ruby by means of two-level phonon spectroscopy [21,24].

Acoustic solitons have also been studied by time-resolved reflectometric [25,26] and interferometric [27,28] methods. Although this paper focuses on bulk propagation, it is good to mention that nonlinear propagation of surface waves has been reported as well by beam deflection techniques [29,30]. We will treat nonlinear dispersive propagation in Section 6.

Section 7 addresses current experimental challenges and sketches future directions of research.

Throughout this paper, we will illustrate phenomena by numerical computations and experimental results. The numerical approach we have developed is presented in Section 2. We will first present the setups with which experimental results have been obtained.

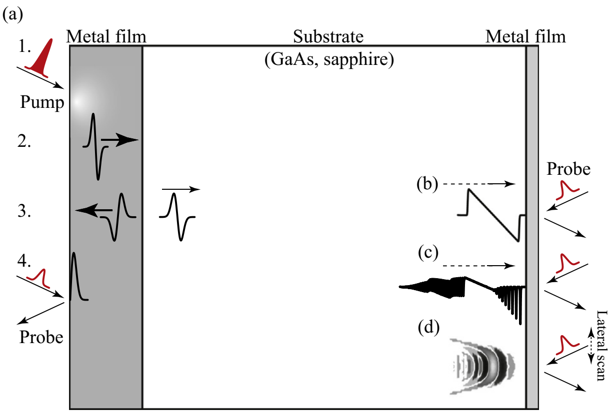

Fig. 1. (a) Picosecond ultrasonics technique. 1. Absorption of energy from pump beam, and heating of the surface region. Coherent strain is generated by thermal expansion of the lattice. 2. The generated strain travels into the film. 3. Part of the wave is reflected at the metal/substrate interface. 4. The part that returns to the interface is detected by a delayed, weak probe pulse. (b) Propagation of the high-amplitude acoustic wave launched into the substrate at room temperature: due to nonlinearity, the wave transforms into a N- or shock wave. The shock wave is detected at the other side by the same technique as in (a). (c) Like (b) but now at low temperatures: the wave develops into a train of solitons, and high-frequency tail. (d) Sketch of diffraction of a soliton train monitored by lateral scanning of the probe beam; brightness indicates strain amplitude.