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Ultrafast laser pulse chirp effects on laser-generated nanoacoustic strains in Silicon

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ABSTRACT

Nanoacoustic strains are generated in Silicon by chirped femtosecond laser pulses using thin Titanium films as transducers. We investigate the effect that the generating laser pulse chirp has on the amplitude of the induced strains, manifested as Brillouin oscillations observed in degenerate femtosecond pump-probe transient reflectivity measurements. The strain amplitude is larger when negatively chirped pulses are used, which is attributed to the more efficient conversion of laser pulse light into acoustic strain in the Titanium transducer. Our present studies clearly show that the dependence of the Brillouin amplitude and the lattice strain is a non-monotonous function of the laser chirp parameter. An optimum negative laser pulse chirp is found for which the strain amplitude is maximized. A detailed thermomechanical model satisfactorily supports the experimental findings. In such a way, it is possible to suppress or enhance the induced nanoacoustic strain amplitude, thus all-optically controlling it by at least a factor of two.

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1. Introduction

The study of very high frequency acoustic waves at a microscopic and nanoscopic level is a particularly active field of research. While MHz ultrasonic wave techniques have been extensively applied to many imaging and non-destructive testing applications, the development of ultrafast lasers and related all-optical pump-probe techniques opened the way for the generation and study of acoustic waves as short as a few picoseconds and of a bandwidth typically 100 GHz, thus opening the way for new applications. These include the non-destructive measurement of thermal and elastic properties of composite materials [1–3], the high-resolution microscopic characterization of interfaces [4,5] the study of vibrational modes of nanoparticles and nanostructured materials [6–8] the measurement of the ultrasonic attenuation in glasses [9,10] and the study of biological materials [11,12].

In what is usually referred to as *picosecond ultrasonics* [13,14] or *ultrafast acoustics*, an ultrashort "pump" laser pulse interacts with a transducer material (usually metallic) and a longitudinal strain pulse is formed. The transducer's lattice dynamics are governed by micro- and nano-acoustic strain localization that is generated either thermally or non-thermally [15–17]. The spatial extend of the localization depends on the material's response to the ultrafast laser excitation and the structure of the solid target and can be of the order of few nanometers [18] up to few tens of nanometers. The generated strains are monitored with a time-delayed "probe" laser pulse usually measuring changes in reflectivity. Applying these techniques in the nanoscale, for example in layered [19,20] and structured [21,22] materials, acoustic (phonon) frequencies up to the THz range have been demonstrated, while the generation of giant acoustic strain pulses in layered systems has been also reported [18].

To really extend the application potential of very high frequency acoustic waves their all-optical control on ultrafast time scales is highly desirable. This has been investigated both theoretically [23] and experimentally by using shaped laser pulses, for example, double-pulse excitation [24] and four-pulse excitation schemes [25]. An exciting possibility is the all-optical control using ultrashort laser pulses whose spectral content is rearranged in time, i.e. chirped pulses. For example, it has been demonstrated that the introduction of chirp affects the direct coherent phonon

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generation in semiconductors [26]. This field is relatively unexplored experimentally, while there are no works that deal with the effect of chirped laser pulses on thermomechanical nanostrains induced in semiconductors through metal thin film transducer coating.

In a previous work we have studied the laser-generated acoustic strains in Ti:Si and Ag:Si layered systems using a degenerate transient reflectivity technique [27]. By probing the Brillouin scattering of laser photons from the generated strain waves within Si, we have investigated the role of the transducer film characteristics (electron-phonon coupling factor, acoustical impedance, thickness). We have demonstrated that localized, giant acoustic strains are produced when a 25 nm Ti film is used as a transducer. More recently, we have reported in a Letter clear indication that negatively chirped femtosecond (fs) laser pulses are capable of inducing larger strain amplitudes than unchirped and positively chirped pulses [28]. Here, we extend our studies in order to provide with more data and increase the resolution in the connection between the lattice strain and the laser chirp values aiming to both determine optimum values for strain amplitude maximization and to explore the strain dynamic range that can be accessed by this method. These new data reveal a remarkable effect, i.e. a non-monotonous behavior. This is supported by additional theoretical calculations, using a modified thermomechanical model based on the combination of a revised two-temperature model (TTM) and elasticity theory, which verify the experimental findings and predict the behavior for larger absolute values of the chirp parameter where experimental data are still not available.

2. The experimental method

The samples used in this study were layered structures (see Fig. 1a) consisting of a 25 nm thick Ti film deposited on 0.5 mm thick Si (1 0 0) monocrystal substrates by unbalanced dc magnetron sputtering. The Ti film served the role of a transducer for the conversion of laser pulse energy into acoustic strain. Ti was the material of choice since it has high electron-phonon coupling strength, high compressive yield strength, and its acoustical impedance is very well matched to that of Si [27]. Therefore, it converts very efficiently the laser pulse energy into acoustic strain, and it transfers very efficiently the acoustic strain into the Si substrate. Furthermore, it allows for a part of the fs probe laser pulses to reach the Si crystal, thus allowing for probing the acoustic strain that propagates inside it.

For the generation and detection of acoustic strains a degenerate pump-probe transient reflectivity technique was developed, described in detail in Refs. [27,28]. Both pump (generation) and probe (detection) laser pulses were derived from a Ti:Sapphire-based amplifier laser system, capable of producing pulses with energy up to 2 mJ and time duration 35 fs (full-width at half-maximum – FWHM). The repetition rate was 1 kHz while the center wavelength of the laser pulses was ~795 nm. The pump pulses were focused normally to the sample surface using a spherical mirror to achieve a fluence of 13 ± 1 mJ/cm^2, which was below the ablation threshold of Ti and into the thermoelastic regime for Ti. The probe pulses were focused at an angle of ~40° relative to the pump pulses using a parabolic mirror. The probe focusing area was centered with respect to the pump focusing area and its size was ~4 times smaller, yielding a fluence ~20 times smaller than the pump fluence, thus avoiding any contribution of the probe pulses to the excitation dynamics.

The probe pulses were variably delayed with respect to the pump pulses with a minimum temporal delay step of 0.8 fs. A lock-in detection technique, incorporating a balanced photodiode, was used for the detection of reflectivity changes after excitation (i.e. differences in reflectivity in the presence and the absence of the pump pulses). The detection scheme was sensitive enough for reflectivity changes of the order of ~10^{-5} to be resolved. Specially developed software was used to simultaneously control the temporal delay and detection instruments, and for data recording.

A linear chirp was introduced to the laser pulses, i.e. the instantaneous frequency of the laser pulses varied linearly with time:

$$\omega_{inst}(t) \equiv \omega - \frac{d\varphi(t)}{dt} \quad (1)$$

where ω and $\varphi(t)$ are the laser pulse carrier angular frequency and temporal phase, respectively. A pulse whose instantaneous frequency increases (decreases) linearly with time is positively (negatively) chirped. It has to be mentioned that the introduction

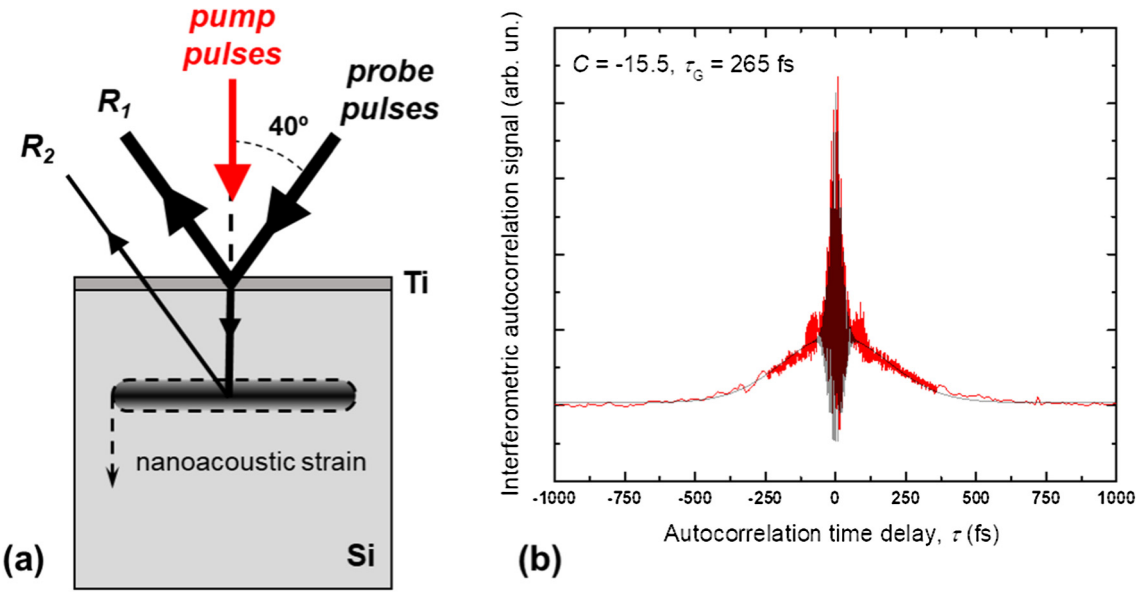

Fig. 1. (a) Schematic of the structure of the used samples and the principle of the detection scheme, (b) Typical experimental second-order interferometric autocorrelation signal (red line) and fit (grey line) using Eq. (3) (extracted parameters C and τ_G are also shown).

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