Ultrasonics xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Ultrasonics

journal homepage: www.elsevier.com/locate/ultras

Fundamentals of picosecond laser ultrasonics

Osamu Matsuda [a,*], Maria Cristina Larciprete [b], Roberto Li Voti [b], Oliver B. Wright [a]

[a] Division of Applied Physics, Faculty of Engineering, Hokkaido University, Sapporo 060-8628, Japan
[b] Dipartimento di Scienze di Base ed Applicate per l'Ingegneria, Sapienza Università di Roma, Via A. Scarpa 14, 00161 Roma, Italy

ARTICLE INFO

Article history:
Received 16 December 2013
Received in revised form 30 May 2014
Accepted 3 June 2014
Available online xxxx

Keywords:
Optical pump-probe technique
Gigahertz and terahertz acoustic waves
Longitudinal and shear acoustic waves
Interferometer
Green's function

ABSTRACT

The aim of this article is to provide an introduction to picosecond laser ultrasonics, a means by which gigahertz–terahertz ultrasonic waves can be generated and detected by ultrashort light pulses. This method can be used to characterize materials with nanometer spatial resolution. With reference to key experiments, we first review the theoretical background for normal-incidence optical detection of longitudinal acoustic waves in opaque single-layer isotropic thin films. The theory is extended to handle isotropic multilayer samples, and is again compared to experiment. We then review applications to anisotropic samples, including oblique-incidence optical probing, and treat the generation and detection of shear waves. Solids including metals and semiconductors are mainly discussed, although liquids are briefly mentioned.

1. Introduction

Picosecond laser ultrasonics, or picosecond ultrasonics, is the study of materials using high frequency acoustic pulses generated and detected by ultrashort optical pulses typically <1 ps in duration [1–5]. When such an optical pulse, known as a pump pulse, is incident on the surface of an opaque solid, some optical energy is absorbed and converted to heat. In the simplest models one assumes that the process of conversion to heat is instantaneous and occurs within a depth approximately equal to the optical absorption depth. In reality nonequilibrium electron heating and relaxation processes will delay the heat generation, and diffusion processes will spatially blur the excitation region. The production of heat in a solid results in a lattice temperature rise, and this leads to a thermal stress that launches a strain pulse propagating in three dimensions. We shall concentrate on the acoustic propagation normal to the surface, because it is in this direction in an isotropic solid that the frequency of the generated acoustic pulse is highest and the wavelength smallest. Moreover, if the optical spot size (typically a few microns) is much larger than the optical absorption depth (~10–50 nm for example), the generated acoustic pulse can be accurately modelled as a superposition of longitudinal plane waves travelling normal to the surface. Acoustic frequencies are usually in the 10–1000 GHz range. Because of the correspondingly small acoustic wavelength, down to the nanometer range, picosecond laser ultrasonics is ideal for investigating thin films and nanostructures.

Sub-surface structures or inhomogeneous regions under the surface, typically at nm to μm depths, can reflect the acoustic pulse back to the surface. This is shown schematically in Fig. 1 for the example of an opaque thin film on a substrate. We can detect the return of the acoustic pulse to the surface by illuminating the sample with a second ultrashort light pulse, known as a probe pulse, focused to the same point on the sample. A set of measurements of the probe beam reflectivity or phase change is obtained by changing the time delay between the pump and probe pulses in an optical delay line, thus avoiding the need for an ultrafast photodetector. This is known as the optical pump and probe technique. Noise reduction is achieved by chopping the pump beam and using lock-in detection. We shall deal separately with the acoustic generation and detection processes without going into any details of the experimental apparatus.

The purpose of this article is to provide a summary of the fundamentals of generation and detection of longitudinal and shear acoustic pulses in picosecond laser ultrasonics. It should serve as an introduction to the subject for those not familiar with this field. After considering the theory of longitudinal acoustic wave generation through the thermoelastic effect in an opaque isotropic solid in the absence of diffusion processes, we briefly review the effect of electron and thermal diffusion processes and other generation mechanisms on the generated acoustic pulses. We then present the theory of optical detection using a normally-incident probe beam, including the detection of both optical reflectivity and phase

* Corresponding author. Tel.: +81 11 706 7190.
E-mail address: omatsuda@eng.hokudai.ac.jp (O. Matsuda).

http://dx.doi.org/10.1016/j.ultras.2014.06.005
0041-624X/

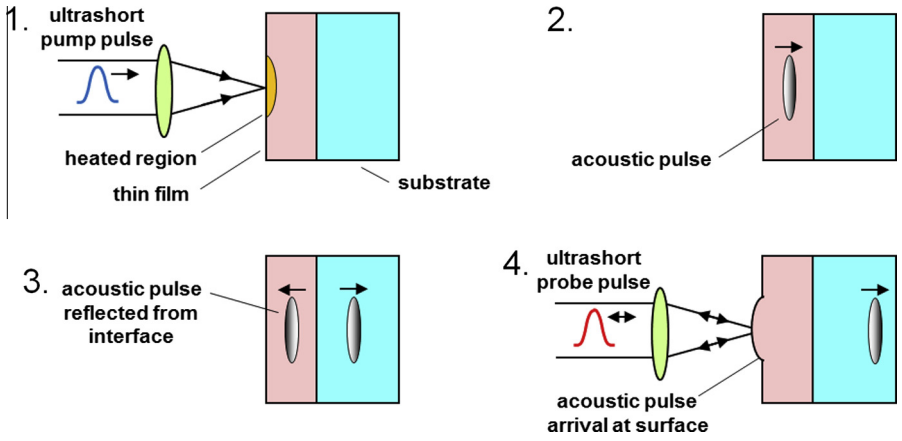

Fig. 1. Sequence of events in picosecond laser ultrasonics.

changes. Different contributions to the acoustic echoes are explained and elucidated using the example of thermoelastically generated acoustic pulses in the absence of diffusion processes. We then describe the theory of optical detection of longitudinal acoustic waves in multilayers, including photonic crystals. Finally, advances in oblique-incidence optical detection techniques and the generation and detection of picosecond shear waves are briefly reviewed.

2. Basic theory of laser picosecond ultrasonics

2.1. Theory of picosecond strain generation and propagation

2.1.1. Thermal stress

When an ultrashort optical pulse is absorbed at a free surface it produces a thermal stress. To calculate the strain pulse shape we first need to know the effect of thermal stress on the stress–strain relation [6]. We shall assume an infinitely wide illumination of a solid by an infinitely short optical pulse. This is a one-dimensional problem in which acoustic diffraction effects do not occur. We shall also at first neglect the effect of any electron or thermal diffusion processes. The thermal expansion tensor α_{ij} of a solid (expressed as a 3×3 matrix) is defined by $\eta_{ij} = \alpha_{ij}\Delta T$, where η_{ij} is the strain tensor, here describing a mechanical equilibrium situation, and ΔT is the position-dependent change in temperature compared to a reference temperature before the optical pulse arrival. For small displacements, the strain tensor is given by $\eta_{ij} = (\partial u_i/\partial x_j + \partial u_j/\partial x_i)/2$, where u_i is the displacement vector and x_i is the position vector. If a material undergoes an instantaneous change of temperature at time $t = 0$, it will expand at a rate governed by the speed of sound. The relation between the stress tensor σ_{ij} and strain η_{ij} in the presence of a thermal stress σ'_{ij} is given by [7]

$$\sigma_{ij} = c_{ijkl}\eta_{kl} + \sigma'_{ij}, \tag{1}$$

where c_{ijkl} is the elastic constant tensor, and the summation is understood to be over repeated subscripts. When the solid has expanded (at time $t = \infty$ in the case of no thermal diffusion), $\sigma_{ij} = 0$ and $\eta_{ij} = \alpha_{ij}\Delta T$. So, from Eq. (1), $\sigma'_{ij} = -c_{ijkl}\alpha_{kl}\Delta T$, where ΔT is a function of x, y, and z in general. For isotropic solids, we define

$$c_{ijkl}\eta_{kl} = 2\mu\eta_{ij} + \lambda\eta_{kk}\delta_{ij}, \tag{2}$$

where λ and μ are elastic constants known as the Lamé constants, and $\delta_{ij} = 1$ when $i = j$ but $\delta_{ij} = 0$ otherwise. So

$$\sigma'_{ij} = -(2\mu\alpha_{ij} + \lambda\alpha_{kk}\delta_{ij})\Delta T.$$

This can be seen by comparison with Eq. (2). Alternatively, Eq. (2) implies that $c_{ijkl} = 2\mu\delta_{ik}\delta_{jl} + \lambda\delta_{ij}\delta_{kl}$. However, for isotropic solids, $\alpha_{ij} = \alpha\delta_{ij}$, where α is the coefficient of linear thermal expansion. So

$$\sigma'_{ij} = -(2\mu + 3\lambda)\alpha\delta_{ij}\Delta T.$$

Alternatively, in terms of the bulk modulus B, Poisson's ratio ν, and Young's modulus Y, where the definitions

$$B = \frac{Y}{3(1-2\nu)}, \quad \nu = \frac{\lambda}{2(\lambda+\mu)}, \quad Y = \frac{\mu(2\mu+3\lambda)}{(\lambda+\mu)}$$

apply, we may write $(2\mu + 3\lambda) = 3B$. Therefore, for isotropic solids,

$$\sigma'_{ij} = -3B\alpha\delta_{ij}\Delta T.$$

The generated thermal stress is hydrostatic and compressive for positive α and ΔT.

2.1.2. Generation of longitudinal waves with picosecond optical pulses in isotropic materials

We assume that $\Delta T = \Delta T(z,t)$ is a function of depth z and time t only, where $z = 0$ corresponds to the flat sample surface and $+z$ is directed into the material. $\Delta T(z,t)$ is assumed to be zero for $t < 0$, and $\Delta T = \Delta T(z)$ to be independent of time for $t > 0$ (in the absence of thermal diffusion). Also, because of this one-dimensional model, the only non-zero tensor component of the strain is η_{zz}. For an isotropic solid,

$$\sigma_{zz} = (2\mu + \lambda)\eta_{zz} - 3B\alpha\Delta T(z). \tag{3}$$

Also, $\sigma_{xx} = \sigma_{yy} = \lambda\eta_{zz} - 3B\alpha\Delta T(z)$. The components σ_{xx} and σ_{yy} are present to prevent lateral contraction (i.e., to make $\eta_{xx} = \eta_{yy} = 0$). Eq. (3) can be rewritten as

$$\sigma_{zz} = 3\frac{1-\nu}{1+\nu}B\eta_{zz} - 3B\alpha\Delta T(z) = \rho_0 v_l^2\eta_{zz} - 3B\alpha\Delta T(z), \tag{4}$$

where v_l is the longitudinal sound velocity and ρ_0 is the density. The elastic wave equation for zero body forces, expressed in terms of the z-directed displacement u_z, where $\eta_{zz} = \partial u_z/\partial z$, is given by

$$\frac{\partial\sigma_{zz}}{\partial z} = \rho_0\frac{\partial^2 u_z}{\partial t^2}, \tag{5}$$

$$u_z(z,t) = \int_{+\infty}^{z}\eta_{zz}(z',t)dz'. \tag{6}$$

To solve the elastic wave equation we need to know the form of $\Delta T(z)$. For ultrashort pulse optical absorption at the surface of a homogeneous isotropic solid,

$$\Delta T(z,t) = \frac{(1-R)Q}{AC\zeta_0}e^{-\frac{z}{\zeta_0}} \text{ for } t > 0, \quad \Delta T(z,t) = 0 \text{ for } t < 0,$$

where R is the optical intensity reflection coefficient, Q is the incident optical pulse energy, A is the area over which the energy Q is distributed (assumed to be a uniform distribution), C is the heat