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[INVITED] Laser generation and detection of ultrafast shear acoustic waves in solids and liquids

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ABSTRACT

The aim of this article is to provide an overview of the up-to-date findings related to ultrafast shear acoustic waves. Recent progress obtained for the laser generation and detection of picosecond shear acoustic waves in solids and liquids is reviewed. Examples in which the transverse isotropic symmetry of the sample structure is broken in order to permit shear acoustic wave generation through sudden laser heating are described in detail. Alternative photo-induced mechanisms for ultrafast shear acoustic generation in metals, semiconductors, insulators, magnetostrictive, piezoelectric and electrostrictive materials are reviewed as well. With reference to key experiments, an all-optical technique employed to probe longitudinal and shear structural dynamics in the GHz frequency range in ultra-thin liquid films is described. This technique, based on specific ultrafast shear acoustic transducers, has opened new perspectives that will be discussed for ultrafast shear acoustic probing of viscoelastic liquids at the nanometer scale.

Contents

1. Introduction

The development of picosecond ultrasonics [1], in which single-cycle acoustic wavepackets are generated through sudden laser heating of a thin film and are detected optically after propagation through one or more film and/or substrate layers, has been exploited for thin film diagnostics and for measurements of acoustic properties in the ~GHz-THz frequency and ~4–100 nm wavelength ranges [2,3]. The method has been applied to a wide range of materials, but in general it has been restricted to generation and characterization of longitudinal acoustic waves only [4]. Contrary to longitudinal acoustic waves, the excitation of ultrafast shear acoustic waves requires more sophisticated experimental configurations that are not yet efficient in the THz frequency range. For the moment, efficient and simple ultrafast shear transducers are lacking and novel ultrafast shear transducers

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would be of great interest for the GHz–THz viscoelastic investigation of many material families, especially disordered and partially ordered systems such as supercooled liquids and glasses, mixed ferroelectrics and multiferroics, and giant magnetoresistance or other correlated electron systems.

This article concatenate the up-to-date findings and references related to the laser excitation and detection of ultrafast shear acoustic waves in solids and liquids above GHz frequencies. In Section 2, examples in which the axial symmetry of the sample structure is broken in order to permit shear wave generation in solids through sudden laser heating are described. Other types of photoacoustic mechanisms and alternative pathways for the ultrafast shear acoustic excitation at GHz-THz frequencies are discussed. Section 3 summarizes the special requirements for the optical detection of ultrafast shear acoustic waves. In Section 4, a promising technique dedicated to measurements of shear relaxation dynamics in liquids, that yielded results of shear viscoelastic properties in glycerol and water at GHz frequencies is described.

2. Generation of ultrafast shear acoustic waves

2.1. Thermoelasticity in off-axis crystals

In picosecond ultrasonics, plane longitudinal acoustic waves at GHz-THz frequencies can be generated by laser light absorption and subsequent local transient thermal expansion [1–3]. From this thermoelastic process and in case of isotropic materials or materials with out-of-plane axial symmetry, shear acoustic waves are not excited in the material volume at all. From the physical point of view, this is the consequence of the in-plane transversal isotropy of thermal expansion which proceeds equivalently along all possible directions from the heated point. As a result, the particle displacement preserves spherical symmetry, and the shear deformation is not generated because transverse displacement orthogonal to this spherically symmetric excitation is forbidden by virtue of symmetry. Thus in many situations with axial symmetry along the direction of propagation, the thermoelastic generation mechanism is coupled to the excitation of plane shear acoustic waves only.

In some circumstances, shear acoustic waves can be excited through mode conversion of longitudinal waves upon oblique reflection at an interface. This situation can occur at the edges of the laser excited area where longitudinal waves are obliquely incident to the interface. The emitted shear waves in this situation are not planar and the predominant emission direction is inclined relative to the interface normal which reduces the detected frequency bandwidth of the shear waves [5–7]. As a result oblique longitudinal mode conversion is not adapted to the excitation of shear frequencies above a few GHz.

To overcome this shortcoming, it is required to break the axial sample symmetry. Experimental results of this sort obtained in such a geometry with an off-axis Zn crystal are shown in Fig. 1. The underlying mechanism of plane shear acoustic waves excitation with broken axial symmetry, described in [4], is efficient even in the common situation of an isotropic thermal expansion tensor β_{kl}, a high symmetry cubic elastic tensor C_{ijkl}, and a diagonal thermoelastic stress tensor $\sigma_{ij} = C_{ijkl}\beta_{kl}T$ which couples directly to the laser excitation of out-of-plane stresses σ_{33} and strains η_{33} upon a transient temperature rise T. In such a geometry, the laser excitation of the out-of-plane stress σ_{33} excites the two quasi acoustic modes, quasi-shear (QS) and quasi-longitudinal (QL), as depicted in Fig. 2. The off-axis orientation of the symmetry axis of the crystal implies quasi acoustic eigenmodes and forbids pure longitudinal and shear modes along the x_3 direction of propagation. The fact that the off-diagonal components of the thermoelastic stress tensor σ_{ij} are zero means that, at the initial stage of laser excitation, there is no direct excitation of the shear stress σ_{23}. The in-plane components of the QL and QS modes cancel each other, which leads to an initially virtual source of shear acoustic waves. However, since the two QL and QS modes propagate with different acoustic velocities v_{ql} and v_{qt}, later on, the acoustic wave field will be spatially and temporally decomposed into two individual acoustic eigenmodes, each of them carrying in-plane shear components, see Fig. 2.

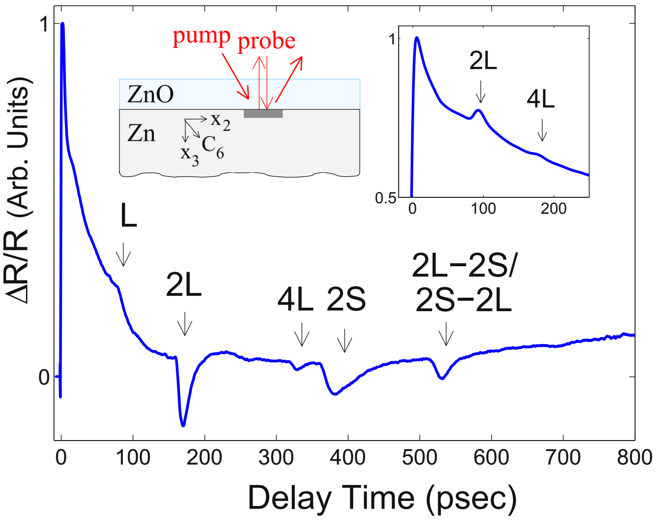

Fig. 1. Change in transient reflectivity for a Zn single-crystal substrate with tilted C_6 axis relative to the interface normal and on which a transparent ZnO film has been deposited. The break of shear symmetry ensures the direct thermoelastic generation of plane shear waves that are partially transmitted into the ZnO film. After a round trip inside the ZnO film, the shear acoustic wavepacket is detected (see the 2S echo in the transient reflectivity signal). The excitation of longitudinal waves does not require such a canted symmetry. Thus, in the case of a Zn single crystal whose normal surface coincides with the C_6 axis, only longitudinal waves are excited (as revealed by the detection of the 2L and 4L longitudinal echoes in the transient reflectivity signal only). Reprinted from [4]. Copyright (2007) by The American Physical Society.

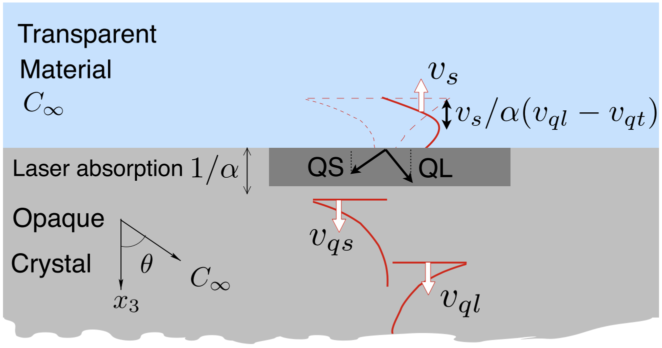

Fig. 2. Due to the exact compensation of the individual in-plane shear strain contributions of the QS and QL quasi modes, excited in the off-axis crystal, the total shear strain is canceled at the first stage of laser excitation. However, thanks to the mismatch propagation of the QS and QL modes at different acoustic speeds v_{qs} and v_{ql} across the laser excited area of spatial extension $1/\alpha$, the separation of the shear strain contributions arises.

In case of an off-axis crystal coated with an isotropic transparent thin film, each of the individual components of the two quasi-acoustic modes QS and QL will be transmitted into purely longitudinal or shear modes across the interface. The in-plane cancellation of the two quasi-acoustic modes QS and QL at the initial stage of laser excitation will partially vanish due to the mismatch propagation of the modes QL and QT in the direction