Spatiotemporal mapping of surface acoustic waves in isotropic and anisotropic materials

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Abstract

We demonstrate a new method for a real-time imaging of surface acoustic waves at frequencies up to 1 GHz with picosecond temporal and micron spatial resolutions using an ultrafast optical pump and probe technique combined with a common path interferometer. Using samples with isotropic or anisotropic substrates coated with metallic thin films, we observe the propagation of Rayleigh-like modes and surface-skimming bulk modes as well as resolving surface phonon focusing effects. In addition we image surface acoustic wave propagation in a laterally inhomogeneous sample. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Laser ultrasonics is an attractive method for the measurement of the elastic properties of thin films and substrates because of the non-contact nature of the measurement. By optically generating and detecting surface acoustic waves (SAW) with wavelengths of a few microns and frequencies in the 100 MHz to 1 GHz range, the thickness and elastic properties of thin films of micron or sub-micron thickness can be probed non-destructively. Recently SAW has been studied extensively because SAW filters are widely used in communication systems and cellular phones. One of the main interests in SAW physics is the surface phonon focusing effect in anisotropic materials, arising because the SAW phase and group velocities depend on the propagation direction. Surface phonon focusing has been studied theoretically for about twenty years [1–3]. In SAW experiments using laser ultrasonics techniques, two kinds of SAW excitation have predominantly been used: one involves focusing the laser radiation to a point or line on the sample surface [4–6]; the other involves transient gratings produced by crossing two laser pulses at the sample surface [7–9]. The intrinsic SAW propagation properties of anisotropic materials, such as the cuspidal structure of the wavefronts, can be effectively demonstrated by focusing the laser radiation to a point source.

Particularly important in the study of anisotropic materials are SAW imaging methods, because the SAW wavefronts can then be directly viewed. Imaging methods for SAW were developed over the same period as those for bulk phonon focusing [10]. Various methods for imaging SAW have been proposed, such as dust removal [11], point-focus transducers in water [12], stroboscopic X-ray topography [13], voltage-contrast scanning electron microscopy [14], scanning acoustic force microscopy [15], stroboscopic phase-shift interferometry [16], and laser knife-edge detection [17]. The SAW propagation properties in anisotropic materials have been most intensively investigated with point-focus transducers in water and by the dust removal method. However, the ideal technique for such investigations would be a totally non-contact optical method for imaging surface phonon focusing phenomena in real time. Conventional optical detection methods are promising [4–6,17], but the signal-to-noise ratio at present seems insufficient for real-time imaging of surface phonon focusing patterns in anisotropic materials where in some propagation directions the signals can become very small.

In this paper, we demonstrate a new non-contact method for imaging SAW in real time using the optical
pump and probe technique combined with an ultrafast Sagnac interferometer [18]. With this method SAW at frequencies up to 1 GHz generated from a point source in isotropic and anisotropic materials can be imaged, allowing ‘movies’ to be made with picosecond temporal and micron spatial resolutions.

2. Experimental set-up

Experiments are carried out with metallic thin films on transparent isotropic and anisotropic substrates. The SAW are excited by visible pump pulses of duration \( \sim 1 \) ps, repetition rate 80 MHz (one pulse every 12.5 ns), wavelength 415 nm, and incident fluence \( \sim 1 \) mJ cm\(^{-2}\), derived from a mode-locked Ti:sapphire laser. The pump light is focused at normal incidence through a \( \times 50 \) or \( \times 100 \) long-working-distance microscope objective lens from the substrate side of the sample to a spot of diameter typically 1.3–2.8 \( \mu \)m (full width at half maximum), that one can control by changing the incident pump beam diameter. Increasing the spot size results in longer wavelength SAW generation. For the present samples, the generation is governed by the thermoelastic effect. For the case of a thin film on a substrate the SAW phase velocity will vary significantly with acoustic wavelength when the film dimensions are of the same order as the film thickness [19]. For this reason the optimum choice of pump spot size, when it is required to probe the film thickness through SAW measurements, will depend on the sample in question. The SAW detection is done interferometrically by two probe pulses of wavelength 830 nm, temporally separated by 510 ps from one another, focused at normal incidence through another objective lens onto the front surface of the film. We use a highly stable common-path interferometer [18], slightly modified from the form proposed by Hurley et al. (Fig. 1). A tilted polarizing beam splitter is used instead of a polarizer, and this makes it possible to obtain the optical phase change signal directly. By scanning the relative positions of the pump and probe spots, the interferometer allows SAW wavefronts to be observed with \( \sim \)pm vertical and \( \sim \)\( \mu \)m lateral spatial resolution. Scanning of the relative positions of the two beams is performed by moving the pump or probe objective lens with an \( x-y \) scanning stage. By changing the optical path of the pump beam with a 0–4 m optical delay line, we can obtain a scanning time range of 0–12.5 ns and picosecond temporal resolution (determined by the optical pulse duration). Spatial scans at fixed times or temporal scans at fixed positions are possible.

3. Results and discussion

Fig. 2(A) shows the phase-change image for a 200 \( \times \) 200 \( \mu \)m region of a polycrystalline gold film of thickness 70 nm on a Crown-glass substrate of thickness 1 mm at a fixed delay time \( \sim \)6 ns after the pump pulse arrival. A horizontal section through the centre of the pattern is shown in Fig. 2(B). Circular SAW wavefronts can be clearly seen emanating from the centre of the pattern, indicating elastic isotropy. Consecutive wavepackets are
temporally separated by 12.5 ns, the period of the laser pulses.

Fig. 3 shows the measured optical phase change plotted in the time domain at a point corresponding to the centre of the pattern in Fig. 2(A). This figure is important for understanding the interferometric detection technique. In the interferometer, two probe pulses are incident at the same point of the sample surface with a time difference equal to 510 ps in our case. At delay time -500 ps on the graph the first probe pulse arrives at the sample surface at the same time as the pump pulse; the second probe pulse arrives at a time corresponding to 0 ps. Sharp peaks appear at these positions owing to hot electron excitation and relaxation. After these peaks the effect of longitudinal acoustic pulses bouncing backwards and forwards inside the film can be seen [18,20,21]. The vertical axis in Fig. 3 therefore represents the phase difference (that is, probe 2 phase minus probe 1 phase) between the two optical pulses reflected from the film surface at different moments in time. For the case of gold the optical phase is expected to be largely governed by the out-of-plane acoustic displacement [18], and so we are effectively measuring the difference in surface displacements for a temporal separation of 510 ps. Provided that the frequency of the SAW is less than \( \frac{1}{2C_2} \) GHz, one is effectively imaging the out-of-plane surface particle velocity. The acoustic generation mechanisms leading to the generation of both longitudinal acoustic waves and surface waves (as well as other polarizations) is complex, and should be treated with a three-dimensional theory of the thermoelastic generation. This problem is not considered in this paper. One advantage of being able to detect longitudinal acoustic waves and SAW with the same apparatus is that the film thickness can be easily calibrated from the longitudinal echo interval, provided that the longitudinal sound velocity of the film is known. (The film 70 nm thickness of sample of Fig. 3 was determined by this method).

In Fig. 2(B) the time-domain signals in the spatial regions (i), (ii) and (iv) represent the same mode excited at 12.5 ns intervals. This corresponds to the first-order Rayleigh-like mode [19], which is the lowest-order mode. The polarization lies in the sagittal plane. The phase velocity of this mode depends on the wavenumber. We therefore see a wavepacket of longer duration in spatial region (i) than that in spatial regions (ii) and (iv). The wavepacket in spatial region (iii) corresponds to a different mode because the velocity is greater (as discussed below). In order to analyse this faster mode, measurements in the time domain at the points (a) and (b) of Fig. 2(A) were carried out. The results are shown in Fig. 4(A) and (B), respectively. Also shown in Fig. 4(C) and (D) are the numerical Fourier transforms of single periods of Fig. 4(A) and (B), respectively. In Fig. 4(C) a high frequency peak (of central frequency \( \sim 750 \) MHz) and a low frequency peak (of central frequency \( \sim 400 \) MHz) can be observed. (Because of the periodicity in the time domain, the Fourier transforms consist of a series of delta functions spaced by 80 MHz.) This implies a difference of \( \sim 2 \) in the velocities of the two modes because the spatial extent of the acoustic pulses (and corresponding excited wavelength spectrum) for these two modes is similar (as expected because this is controlled by the pump spot diameter). We associate the faster mode with a surface-skimming bulk longitudinal wave. In the spectrum of Fig. 4(D), corresponding to the measurement at a point b situated 65 \( \mu \)m from the pump spot position, this mode is not visible, presumably because of the initial lower amplitude of this mode.

![Fig. 3. The optical phase change plotted in the time domain at a point corresponding to the centre of the pattern in Fig. 2(A).](image)

![Fig. 4. (A) and (B): Optical phase change plotted in the time domain at points corresponding to points (a) and (b) in Fig. 2(A), respectively. (C) and (D): Numerical Fourier transforms of single periods in (A) and (B) of this figure, respectively. Broken lines are just guides to the eye.](image)
In order to confirm the above we have also taken three frames of a SAW movie in Fig. 5(A), (B) and (C) at 2.13 ns intervals, for a 90 × 90 μm area of the same sample. The measurement time for one image is 8 min. The arrows in the three figures show the position of the wavefront of the surface-skimming bulk mode. The estimated group velocity (~5300 m/s) is, as expected, close to the longitudinal sound velocity for the substrate (~5650 m/s) calculated from the known values of the Young’s modulus $E = 71.5$ GPa, Poisson’s ratio $\nu = 0.219$ and density $\rho = 2.55$ g/cm$^3$ of crown glass.

Fig. 6(A) shows a SAW image of a 150 × 250 μm region of a polycrystalline gold film of thickness 400 nm on the (1 0 0) surface of a single-crystal TeO$_2$ substrate of thickness 1 mm at a fixed delay time. The vertical and horizontal directions correspond to [0 0 1] and [0 1 0] respectively. The SAW wavefronts are approximately elliptical in shape, as expected from the tetragonal symmetry of the substrate. Strong focusing is observed at ~81° from [0 1 0] to [0 0 1]. This can be seen more clearly in Fig. 6(B), where the intensity has been plotted (proportional to the square of the amplitude in Fig. 6(A)). Theoretical considerations ignoring piezoelectricity and the finite film thickness show an expected focusing.
direction at \( \sim 84^\circ \), in reasonable agreement with experiment [22]. This shows that surface phonon focusing can be imaged effectively with our method.

In Fig. 7(A) an image of SAW propagating in a laterally inhomogeneous structure is shown for a 200 \( \times \) 200 \( \mu \text{m} \) region of a Au/Cr/glass structure. Fig. 7(B) corresponds to an image of the optical reflectivity at the probe wavelength of the same region, obtained simultaneously with the SAW image. In Fig. 7(C) the two images are superimposed for comparison. The sample was made as follows. A chromium film of thickness 400 nm was deposited by electron beam deposition on a crown-glass substrate of thickness 1 mm. Then a second thermal evaporation of gold of thickness 470 nm was made through a mask consisting of a grid of square holes of separation 60 \( \mu \text{m} \) and hole-size 25 \( \times \) 25 \( \mu \text{m} \). This mask was some distance from the substrate, leading to a smearing of the gold pattern in the horizontal direction (since the gold evaporation boat extended in this horizontal direction). The white regions in Fig. 7(B) therefore correspond to gold, whereas the darker regions correspond to chromium.

As seen by examination of Fig. 7(C), the presence of the gold structure on the chromium tends to decrease the group velocity, leading to the distorted wavefronts shown. In addition, the edges of the gold structures scatter the SAW into arc-shaped wavefronts at various points on the surface. This example shows the potential of the technique for imaging SAW in complex geometries.

4. Conclusions

In conclusion we have demonstrated a new method for real-time imaging of surface acoustic waves at frequencies in the 100 MHz to 1 GHz range with picosecond temporal and micron spatial resolutions using an ultrafast optical pump and probe technique. Isotropic samples show circular wavefronts, and we have succeeded in resolving both first-order Rayleigh-like modes and surface-skimming bulk waves. Using anisotropic substrates we have demonstrated the imaging of surface phonon focusing effects. We have also shown how wavefronts in complex nanostructures can be mapped. In future we expect to see a wide variety of uses of this technique in both fundamental physics and in the evaluation of SAW devices.

References