Interferometric detection of acoustic shock waves

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Abstract. We excite high-amplitude longitudinal coherent acoustic strain waves in a chromium film deposited on a thin sapphire slab, by 100-fs pulses from a 1-kHz Ti:sapphire regenerative amplifier. The bipolar strain pulse that is launched into the crystal, gradually transforms into a shock wave when attenuation by thermal phonons is weaker than the nonlinear action. We detect such shock waves at the opposite side of the slab by probing the reflection of a second chromium film in a standard reflection geometry, with a common-path interferometer to resolve both the induced amplitude and phase changes. We first discuss the interferometer and its standard performance. We then present the measurements on propagating shock waves, and analyze the results on a qualitative level.

1. Introduction
In recent years, the subject of nonlinear phonon propagation has obtained increasing interest. The detection of acoustic shock waves [1] and solitons [2, 3] has possibly opened up a new field of research, where coherent phonons of terahertz frequencies can be used to acoustically investigate and manipulate nanometer-sized materials [4]. In this developing field, interferometric detection is an appropriate technique to use [5]. Its main benefit is the ability to detect the phase retardation between a signal and reference pulse due to the (instantaneous) displacement of the surface when strain reaches the surface of the detection layer.

In this paper, we study high-amplitude acoustic strains by means of a Sagnac-type interferometer [6]. After introducing the technique and typical performance of the setup, we present results obtained on nonlinear strain propagation through a 126 \( \mu \text{m} \) slab of sapphire. We compare these results to previous results obtained in a pure reflection experiment [1].

2. Experimental setup
Fig. 1 (a) shows a sketch of the experimental setup. The design is similar to the one presented in Ref. [6]. However, the addition of focusing lenses in both sidearms and the use of a third photodetector for reference measurements before entering the interferometer decreased the noise level with an order of magnitude. In our specific experimental setup, which operates on a 1 kHz amplified Ti:sapphire laser system at a wavelength of 800 nm, we make use of three fast digital multimeters (Agilent 3458A) to correct noise in the probe signal on a pulse-to-pulse basis [7]. Additionally, since we can measure three channels separately, we can infer the absolute amplitude signal \( \rho \propto D_1 + D_2 \), and phase signal \( \delta \phi \propto D_1 - D_2 \) at the same time.

The pump light is doubled to 400 nm before sending it to the sample, and focused to a waist \( w_{\text{pump}} \) of 165 \( \mu \text{m} \), to obtain pump fluences as high as 30 mJ/cm\(^2\). The probe pulses were
focused down to $\sim 10 \mu m$. In this report, we discuss two different pump-probe geometries, both shown in Fig. 1 (a): pump-probe on the same metal film, and pumping on the other side of the substrate, so that we can monitor acoustic wave propagation through the substrate.

In Fig. 1 (b), a typical measurement result for $\rho$ and $\delta \varphi$ is shown. Here, pumping and probing occurs on a Cr layer of $\sim 200 \text{ nm}$ thickness deposited on a Si substrate. Comparing with previous measurements on the same system [8], we derive an interferometric contrast around 85 percent. This value is significantly below maximum, and we attribute it to the poor amplifier beam quality; it is somewhat elliptic, and the beam profile shows large deviations from the (ideal) Gaussian profile. The typical noise level is $\sim 10^{-5}$ for both amplitude and phase signal (corresponding to surface displacements of the order of 1 pm) at an integration time of 1 second. We note that for high signals the noise is larger, due to slow variations in pump intensity and/or position, of the order of 1% of the total signal amplitude. The inset in Fig. 1 (b) shows the fast response at $t = 0$, due to fast heating of the electron gas. The response time of the phase signal indicates that the temporal resolution in this geometry is better than 300 fs.

3. Results and discussion
Before presenting the results on strain propagation through sapphire, we start by estimating the effects of diffraction, attenuation, and nonlinearity, for a typical strain found in this experiment [1]. It can be seen in Table 1 that indeed the nonlinear action is dominant at the highest powers, and this sample thickness.

The strain is generated in a 200 nm thick Cr layer, and detected on a 30 nm Cr layer at the other side of the 126 $\mu m$ sapphire slab. In Fig. 2 (a), we show the detected phase signal after propagation for a range of fluences, up to 26 mJ/cm$^2$, roughly the damage threshold of
Typical lengths over which wave deformation effects become apparent. These estimates were made for an input strain wave shape of amplitude $s_0 = 10^{-3}$ and central frequency $f = 40$ GHz. For sapphire: sound velocity $v_s = 11.23 \times 10^3$ m/s, density $\rho = 3.97 \times 10^3$ kg/m$^3$, elastic constant $c_{33} = 4.97 \times 10^{11}$ N/m$^2$, nonlinear coefficient $\alpha = -18.5 \times 10^{11}$ N/m$^2$, and viscosity $\eta_{33} = 5 \times 10^{-4}$ Ns/m$^2$.

| Sample thickness $d_{sample}$ | Shock formation length $\lambda_{nml}$ | Attenuation length $\lambda_{att}$ | Diffraction length $\lambda_{diff}$ |
|-------------------------------|--------------------------------------|----------------------------------|----------------------------------|
| Expression                    | $\frac{v_s^3 \rho}{3\pi |\alpha| s_0 f}$ | $\frac{c_{33}^{3/2}}{2\pi^2 \rho^{1/2} \eta_{33} f^2}$ | $\frac{w_{pump}^2}{v_s}$ |
| Typical value                 | 126 $\mu$m                           | 8.1 $\mu$m                       | 352 $\mu$m                       | 97 mm |

Chromium. The minus sign with respect to the measurements in Fig. 1 (b) is due to the reflection at the Cr/Si interface in the latter case. For the highest fluence, the 1% phase signal change corresponds to a surface displacement as high as 0.6 nm. Fig. 2 (b) shows the typical pulse width after propagation through the sapphire. At low powers, the temporal width has broadened slightly ($\pm$ 2 ps) with respect to the results shown in Fig. 1 (b), due to attenuation in the sapphire substrate (affecting mainly the high frequencies in the wave packet), and the limited parallelity of the substrate front and rear planes; these were determined to have an angle of 0.076 $^\circ$, corresponding to a broadening of 1.2 ps over the probe spot.

At the highest fluences, the increase is roughly a factor of six with respect to the incoming pulse. The nonlinear dependence on pump fluence is due to a nonlinear dependence of the input strain amplitude on fluence. This is likely due to the temperature rise of the order of 1000 K for the highest powers, and the corresponding increase of the thermal expansion coefficient. In the inset in Fig. 2 (b), we show a typical N-wave strain formed in the sapphire substrate for 24 mJ/cm$^2$. Since the sample traversal time $t_{travel}$ is 11.2 ns and the decrease in arrival time $\delta t$ is around 30 ps for this fluence, we can ascribe an average Mach number of $(1 + \delta t/t_{travel}) = 1.0027$ to the propagation. The inset of Fig. 2 (b) shows that this corresponds to the average strain amplitude during traversal of the sample.

Knowing that the phase signal is dominated by the surface displacement, which effectively means an integration of the strain that has passed the surface, we would expect the measured signal to develop into a parabolic shape. We have plotted such a shape for the highest power in Fig. 2 (a), showing that this approximation is indeed correct. With respect to the measurements performed on acoustic shock waves before [1], under similar circumstances (sapphire substrate thickness, absorbed energy in Cr), we can say that the broadening of the acoustic wave is of the same order.

A nice illustration of nonlinear action can finally be seen in the inset in Fig. 2 (a), where we show the reflection from the 200 nm Cr generation layer/sapphire interface, arriving 76 ps after the main pulse. Since the sign of this wave is opposite to the original one, here nonlinearity does not broaden but narrow the original pulse, giving rise to a sharply peaked signal.

Besides the previously mentioned loss in temporal resolution due to the parallelity of the sample planes, there is an additional loss of high-frequency phonon information. This is most probably not due to diffraction (see Table 1), or the variation of pump intensity over the probe spot (giving rise to arrival time differences of only 300 fs). Our assumption is that there is a large amount of high-frequency scattering at the Cr/sapphire interface. The quality of the Cr film is poor, and the typical Cr grain size matches the $\sim 300$ GHz cutoff frequency. With our large probe spot, one averages over a number of scatterers (Cr grains with different sizes and axis orientations).
Figure 2. (a) Phase signal obtained by probing at the other side of the sapphire crystal, as a function of pump fluence. Inset shows the $\sim 10\%$ amplitude reflection from the 200 nm Cr (generation layer)-sapphire interface. (b) Typical width as determined from the traces shown in Fig. 2 (a). Inset shows the initial wave shape (blue line), and the wave shape after propagation through 126 µm of sapphire (red line), for a pump fluence of 24 mJ/cm$^2$, obtained from a simulation.

4. Conclusions
We have presented a common-path interferometer setup, with an interferometric contrast of $\sim 85\%$, capable of detecting amplitude and phase modulations of the probe light of the order of $10^{-5}$, with a temporal resolution better than 300 fs. We have succeeded in detecting the formation of acoustic shock waves in sapphire, giving surface displacements of 0.6 nm and a wave stretching to 75 ps after propagation over only 126 µm.

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