

## Towards ultrafast pump-probe spectroscopy on trains of strain solitons

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We describe a low-repetition-rate ultrafast pump-probe setup, based on the picosecond ultrasonics technique, to be used to detect trains of strain solitons. The acoustic pulses are generated and probed with a kHz-amplified Ti:sapphire laser. By correcting on a pulse-to-pulse base, the sensitivity of the setup reaches  $2 \times 10^{-5} \text{ Hz}^{1/2}$ . We demonstrate the effectiveness of our setup by a series of pump-probe measurements on a thin chromium transducer at room temperature, and show that the injected strain in sapphire can exceed  $2 \times 10^{-3}$ . A first measurement is presented on the arrival of a high-amplitude acoustic pulse that has travelled through a sapphire plate at room temperature, and qualitatively explained. This paves the way to detect high-amplitude individual solitons in the train at low temperatures with this pump-probe setup.

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**1 Introduction** Picosecond ultrasonics [1] has recently found new applications in fundamental solid state research. Acoustic strain pulses, that are generated in thin metallic transducer layers by subpicosecond pulses, are studied after propagation through crystals typically over several hundreds of micrometers. At high enough strain amplitude and at room temperature, nonlinearity and thermal damping will result in a reshaping of the injected bipolar strain pulse into a damped N-wave [2]. At low temperatures thermal damping is absent, and the presence of increasingly high frequencies will lead to a significant role for lattice dispersion. The compressional part of the waveform will eventually develop into a train of strain solitons [3].

Hao and Maris [4], and more recently, Singhsomroje and Maris [5], were able to observe one or more of these solitons in several materials by pumping with the focused beam of an 80-MHz modelocked Ti:sapphire laser and using picosecond ultrasonics as detection method. Muskens and Dijkhuis obtained much higher strain amplitudes over much larger areas by pumping with an amplified 1-kHz Ti:sapphire laser system, and observed trains of up to ten individual solitons, as appeared from a detailed analysis of Brillouin scattering data [3, 6].

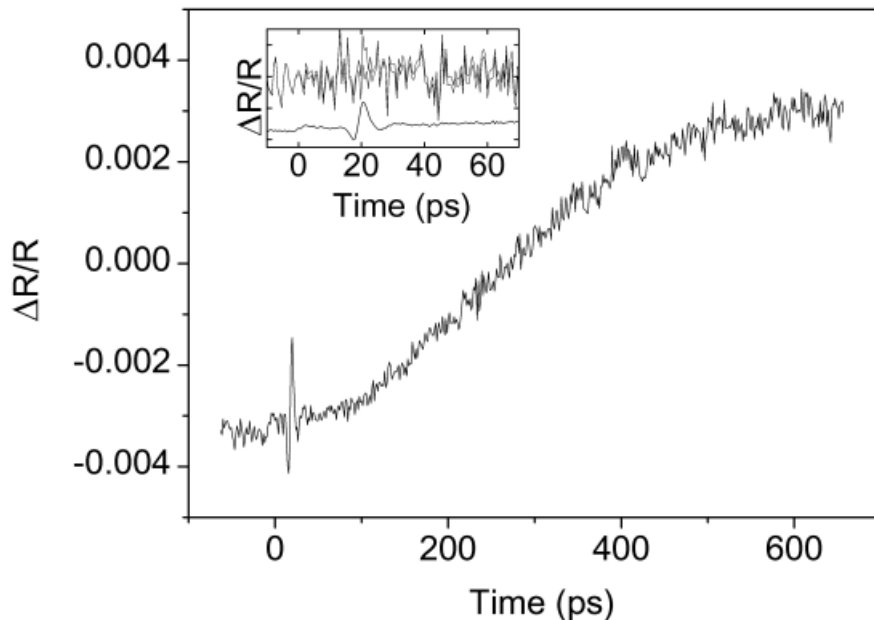
In this paper, we present the first results obtained with a pump-probe reflectometry setup employing a kHz-amplified laser system. We performed measurements at room temperature on several metallic layers to show directly that the induced strain exceeds 0.2%, confirming the results of earlier Brillouin experiments [6]. The main purpose of this setup, however, is to perform sensitive measurements on propagation of ultrashort strain pulses over large distances at liquid helium temperatures. We present the first measurements on sapphire at room temperature showing the formation of a shock wave, indicating that measurements on soliton trains are feasible with our kHz setup.

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**2 Samples and setup** The experiments are performed on three samples. Two 115- $\mu\text{m}$  thin, c-cut sapphire crystals and a lead molybdate crystal are used. A thin metallic layer is evaporated on respectively one or two of the adjacent sides of the sapphire crystals: either a 100-nm Cr/5-nm Al layer at one side (sample A), or with a 100-nm Cr layer at one side and 200-nm Al layer at the other (sample B). To enhance the acoustic reflection at the Cr-crystal interface we use a 100-nm Cr layer deposited on a  $10 \times 4 \times 5 \text{ mm}^3$  lead molybdate crystal (sample C).

Our experiments are performed with a standard pump-probe setup [1]. We determine the differential reflection from the signal with and without pump. The relatively long time between pump pulses in our kHz system allows us to correct for low-frequency noise in the probe signal (mainly intensity fluctuations, specified to 1%) per shot by fast electronics. For this purpose, a reference signal is split off from the probe, and both signals are processed by two high-precision (Agilent 3458A) multimeters. The division of the two signals reduces the noise, resulting in a typical uncertainty of  $2 \times 10^{-5} \text{ Hz}^{1/2}$ . Figure 1 shows a typical trace of our setup, taken on sample A.



**Fig. 1** Long-time differential reflectivity measurement on a 100-nm Cr/5-nm Al layer (sample A, crystal-side pumped and surface-side probed). Coherent acoustic strain pulse is clearly visible at  $\sim 20$  ps, followed by an incoherent heat-pulse. Inset shows typical probe and reference signals (upper traces) and corrected signal (lower trace).

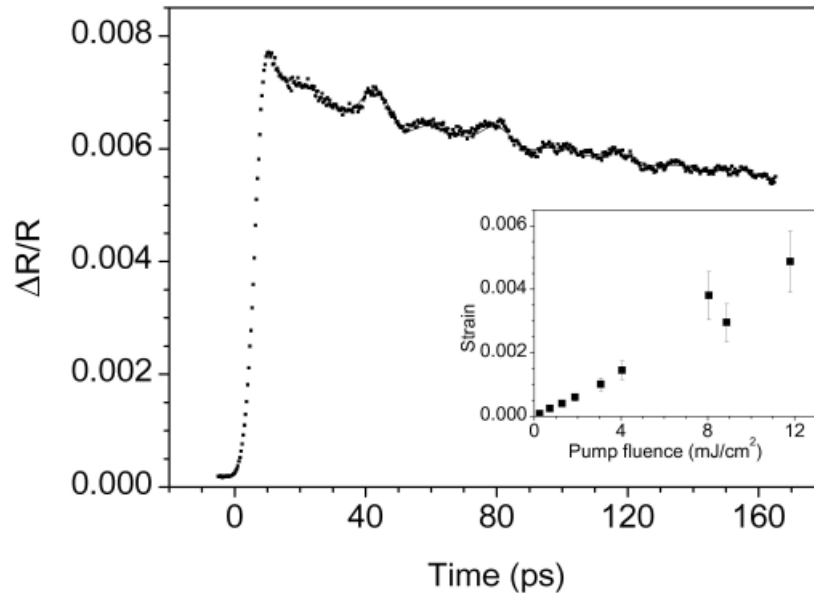
### 3 Results and discussion

**3.1 Cr transducer** We have performed a series of pump-probe measurements on the Cr thin film transducer on lead molybdate (sample C), as a function of pump fluence. In Fig. 2(a), we show a typical trace, together with the best fit, explained below. One can see the characteristic acoustic reflections returning from the metal-crystal interface, superimposed on a slow decaying background due to diffusion loss of heat deposited in the film by the pump pulse.

By applying standard theory [1] and taking into account only the pure acoustic reflections, we can extract from the trace the typical amplitude and temporal width of the induced strain and the reflection coefficient at the Cr-lead molybdate interface. Here, we approximate the initial strain pulse by the derivative of a Gaussian [6].

Figure 2(b) shows the strain as a function of pump fluence. As expected, we find a linear dependence. Clearly, the strain in the film exceeds  $4 \times 10^{-3}$ , at intensities still significantly below the damage threshold of  $15 \text{ mJ/cm}^2$  [3]. When we compare these results with those obtained by Muskens for the strain

injected into a sapphire crystal [6], taking due account of differences in sound speed of film and crystal and of reflection at the interface, we find that the results are in reasonable agreement.



**Fig. 2** Typical trace (squares) of acoustic echoes for a Cr film on lead molybdate, obtained at a pump fluence of 8.9  $\text{mJ}/\text{cm}^2$ , together with a best fit (full line). Inset shows strain versus pump fluence.

The temporal width of the strain pulse is estimated at  $9.8 \pm 0.5$  ps. The reflection coefficient for the Cr-lead molybdate interface exceeds the ideal acoustic mismatch [7] strongly. Strikingly, the reflection coefficient at the interface increases with increasing pump intensity (not shown). We attribute these effects to imperfect mechanical attachment of the film to the crystal. In the case of the Cr layer on sapphire (sample B), assuming identical values for the input strain pulse, however, we find only a minor deviation of the reflection coefficient from the acoustic mismatch value of 0.11, indicating an excellent mechanical attachment.

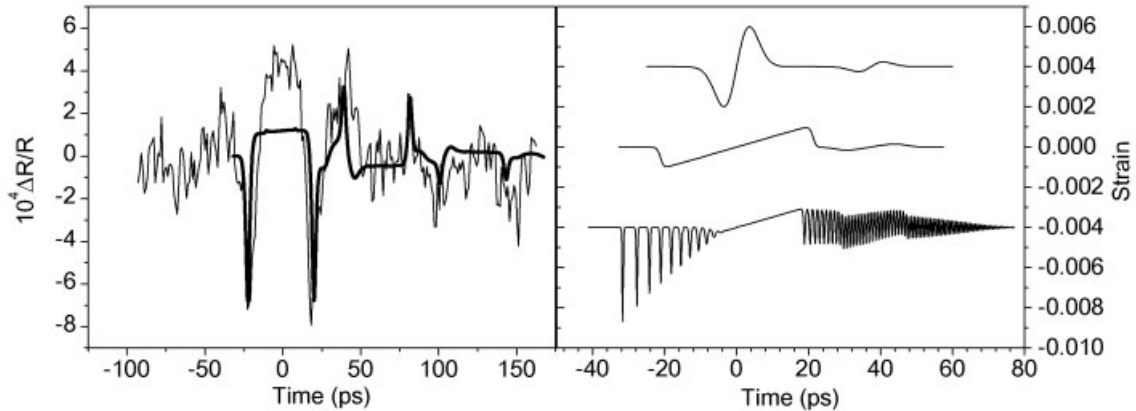
**3.2 Acoustic pulse in sapphire** To make the step towards pump-probe measurements at different sides of the crystal slab, we took sample B, pumped the Cr layer (500- $\mu\text{m}$ -diameter spot) and probed the Al layer. Thanks to the large speed of sound in sapphire of 11.23 km/s (c-axis) and the thin crystal, the traversal time amounts to about 10 ns, a delay that we can handle with our 50-cm long, 8-pass optical delay line.

Figure 3(a) shows our first result on the differential reflectivity for this configuration. One can see two sharp peaks, indicating the two edges of the N-wave developed from the bipolar pulse by nonlinearity. Also visible are the first and second order reflections of the N-wave on the Al-sapphire interface.

In a first attempt to quantitatively explain this result, we take the parameters obtained in Section 3.1 as input for a simulation based on the Korteweg-de Vries-Burgers equation (KdVB) [2], and describe the propagation of strain pulses in the presence of nonlinearity, damping, and moderate dispersion. At 11  $\text{mJ}/\text{cm}^2$  pump fluence, we estimate the strain in the film at  $4.5 \times 10^{-3}$ . Setting the reflection at the Cr-sapphire boundary to 12%, and taking into account the difference in sound velocities between the two materials, the injected strain amplitude in sapphire is  $2.0 \times 10^{-3}$ . Again, we approximate the initial strain waveform as a Gaussian derivative.

Figure 3(b) shows the strain pulse as it enters the sapphire (upper trace), and the shape after traversing 115  $\mu\text{m}$  (center trace). Obviously, the bipolar input pulse has changed into a N-wave with smooth edges,

damped to about 50% of the initial maximum amplitude. In Fig. 3(a) (bold line) we show the effect such a strain pulse has on the reflectivity of the Al-film. When the width of the initial pulse is set to 5 ps, and the reflection coefficient at the Al-sapphire interface is taken to be 0.4, best qualitative agreement is reached. Deviations may well be caused by input pulses with a different shape from the one assumed in calculations. Further experiments are clearly needed at this point.



**Fig. 3** (a) Measured differential reflection signal at  $11 \text{ mJ/cm}^2$  pump fluence, and fit obtained by solving the KdVB-equation (bold line). (b) Upper trace: input strain pulse at given pump fluence. Center trace: result for given initial pulse after travelling through  $115 \mu\text{m}$  of sapphire at room temperature. Lower trace: as for center, but now at liquid helium temperatures. In both figures, zero time indicates  $10.25 \pm 0.09 \text{ ns}$  after firing the pump pulse.

When lowering the temperature, the most important effect on strain propagation is the decrease of thermal damping. In Fig. 3(b) (lower trace), we have plotted the result of simulations for viscosity 0 (being  $4.54 \times 10^{-4} \text{ Ns/m}^2$  for sapphire at room temperature [7]). It is apparent that this initial strain amplitude will lead to the development of several strain solitons within this distance at low enough temperatures. We hope to detect these trains in the near future.

**4 Conclusions** We have tested a high-power pump-probe setup operating at 1-kHz repetition rate by a series of picosecond ultrasonics measurements on a chromium thin film transducer. Ti:sapphire-regenerative-amplified-laser-system-induced strains may exceed  $5 \times 10^{-3}$  over a focus of  $\sim 500 \mu\text{m}$ .

In addition, we managed to detect the arrival of a damped N-wave developed from a high-amplitude strain wave travelling through a  $115\text{-}\mu\text{m}$  thick sapphire crystal at room temperature. The result found qualitatively agrees with theoretical predictions. Under the same circumstances and at liquid helium temperatures, simulations show that the same input strain will produce a not yet fully developed train containing up to 11 solitons.

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