Investigation of strain soliton formation and propagation

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Introduction

The availability of high power, ultrafast lasers has resulted in the application of new techniques to study non-linearity and dispersion in solids. One such effect is the generation and propagation of solitons - acoustic strain packets with a group velocity faster than the speed of sound. Previously, soliton formation in crystals has been studied using picosecond (ps) excitation of metal transducers[1-3]. Ultrafast (ps) laser excitation of the transducers deposited on the surface of a variety of crystals (eg sapphire, MgO, Ge, Si, GaAs) resulted in the formation of an acoustic strain packet; at low excitation powers, the packet propagated through the crystal at the speed of sound. When the initial excitation pulse amplitude was large enough (governed by the metal film and the crystal studied), the strain packet evolved into a soliton. Theory shows this soliton traverses the crystal at a speed slightly greater than the speed of sound, and that the velocity increases as the excitation power is increased[3]. The Korteweg-de-Vries (KdV) equation can be used to describe the acoustic wavepacket behaviour, and to predict the effect of strain amplitude on the number of solitons which evolve, and the soliton velocity and shape. One disadvantage of conventional pico-ultrasonics is that because the fraction of the available energy from the laser pulse that is converted into a strain packet is so low, the strain amplitude is not always sufficient to observe non-linear effects, unless the propagation distance is long.

It is possible to generate solitons over shorter distances if the strain amplitude is increased. This can be achieved using femtosecond Ti:sapphire amplifiers. Recent work using systems of this type have demonstrated that femtosecond laser excitation of thin metal films deposited on the surface of crystals (sapphire, ruby[4,5]) also results in the formation of solitons. The higher amplitude excitation has been shown to generate solitons over smaller distances than observed with Ti:sapphire oscillators.

Bolometric detection provides an established technique which can be used to study the propagation of acoustic packets across crystals. The heat pulse technique, first demonstrated by von Gutfeld and Nethercot[6], has been used in recent years to study a variety of systems, including semiconductor crystal structures (for an introduction to the techniques, see e.g.[7]). Because of the ability of the technique to distinguish between separate phonon modes, measurements of this type continue to provide a suitable method to study a range of material properties, as varied as e.g. coherent phonon generation in semiconductor structures, hot carrier energy relaxation and the role of non--radiative recombinations on the performance of optoelectronic devices. The bolometric detection technique allows for satisfactory temporal resolution of the acoustic wavepacket, and so is an obvious choice to aid in the further study of non--linear effects following ultrafast optical excitation of crystals.

The basic idea of our experiment is to generate a short, high amplitude acoustic pulse and to launch it across the crystal. The pulse is then detected by the bolometer on the opposite face of the crystal at some short time later. If the excitation power is relatively low, the shock wave generated when the laser pulse is thermalised in the metal film traverses the crystal at the speed of sound. However, if the excitation pulse amplitude is high enough, the strain pulse amplitude is large enough evolve into a soliton train (the number of solitons can be determined using the KdV equation); the propagation time across the crystal reduces since the soliton velocity exceeds the sound velocity. In recent studies, the soliton has been detected either by Brillouin scattering techniques[4,5], or by detecting changes in the pulse shape following reflection back across the sample[1,3]. The time-of-flight technique allows us to more easily achieve sufficient temporal resolution over long propagation distances than is possible using the methods reported by other groups, and is well suited to this application. (Although pump-probe measurements offer good temporal resolution, the technique cannot be easily applied to study long propagation distances because of the long delays necessary). By using bolometric detection, we can obtain an accurate value for the acoustic packet velocity as a function of excitation power, and so can observe the onset of soliton generation. In this project, the first loan of the Coherent Libra fs amplifier system (UFL2), we have studied soliton generation in sapphire, silicon and GaAs; however, in this report, we outline only our results obtained from sapphire.

Experimental Details

The sapphire sample used in this experiment was a highly polished single crystal of dimension 9x5x10 mm^{-3}, with the c-axis perpendicular to the 9x5 mm^{-2} surface. Onto this surface, a 100nm thick chromium film was deposited by thermal evaporation. On the opposite face of the crystal, superconducting aluminium bolometers were fabricated. The sample was mounted in an optical access, continuous flow, helium cryostat and maintained at a temperature of $T_o \approx 2K$, at the mid-point of the superconducting transition of the bolometers. The chromium film was excited with pulses from the regeneratively amplified Ti:sapphire laser system (Coherent "Libra", UFL2). The average power of the output beam was 1W at 800nm, and at a repetition rate of 1kHz, the output beam carried approximately 1mJ per pulse. Pulses of ≈ 120 fs were directed onto the sample without focussing. The spot size on the sample was varied using aperture plates, and was approximately 2mm diameter. The optical pump fluence was varied by inserting calibrated neutral density filters into the beam path, and the upper limit in our measurements was estimated to be 0.6 mJ/cm^{-2}. The generated acoustic packets were detected by measuring the change in resistance of the constant current biassed bolometer. This signal was amplified and detected using a high speed digitiser. To make an image of the spatial dependence of soliton propagation, the laser spot was raster scanned across the metal film using a pair of galvanometer controlled mirrors.

Results and Discussion

A typical bolometer signal from the sapphire is shown in Fig 1. Three distinct peaks are clearly visible, and are marked on the figure. The onset of arrival of the first peak, at $t \approx 0$ns, corresponds to the optical response of the bolometer (a small amount of the incident light is transmitted through the film) and provides the zero time reference point. The second and third peaks correspond to the arrival of the different modes in the acoustic wavepacket; the second peak arrives at a time consistent with the arrival of longitudinal phonons, and the third peak to transverse acoustic modes, both of which traverse the substrate at their respective speeds of sound. Phonon propagation in sapphire is well studied, and the longitudinal (LA) and transverse (TA) phonon velocities are $c_{LA} \approx 11\ 100 ms^{-1}$ and $c_{TA} \approx 6000 ms^{-1}$ respectively[8,9]. Due to the large crystal dimension, these signals are well separated in time. In Fig 2, we plot the results of measurements of the acoustic velocities as a function of pulse energy for both components of

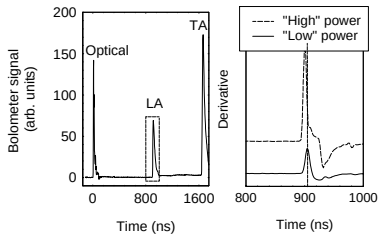

Figure 1. Left: A typical bolometer trace, obtained for a pulse energy of ≈0.009 μJ. Right: The derivative of the bolometer signals for "high" and "low" excitation powers, where the change in arrival time of the signal is visible. The straight dashed line is a guide to the eye.

the wavepacket. First, we consider the third peak to arrive, due to transverse acoustic phonons. We measure the phonon velocity as c_{TA} = 6100 ∓ 50 ms^{-1}. This value is in excellent agreement with published values for the transverse phonon velocity in c-axis sapphire. Across the range of excitation powers used, we see no significant change in the velocity Δc_{TA} is negligible within the experimental error). Now we consider the second signal peak in Fig 1; at low excitation power, we measure a velocity of 11 100 ∓ 50 ms^{-1}, which is also what is expected for LA phonons. As we increase the excitation power, we see that the arrival time of this signal decreases, and continues to decrease across the range of powers used. The measured change in the time-of-flight corresponds to an overall change in velocity of ≈ 1%. This indicates that, as the excitation power is increased, there is an additional fast component arriving at the bolometer, and the contribution from this component increases with increasing power.

Using the value of the velocity shift we observe at the highest excitation powers, we calculate that a negative (compressional) strain of ~1% is required. This value seems high considering the material parameters of chromium and sapphire, and the pump fluence that might be achieved using a Ti:sapphire amplifier. However, the pump fluence used in our experiments is comparable to other reported works. The high strain amplitude required to produce the velocity enhancement we measure indicates that the simple thermal expansion approach to strain determination, (which has been previously applied to estimate strain in picoultrasonic measurements) is not entirely appropriate for such high power ultrafast laser pulses.

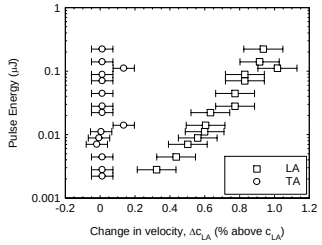

Figure 2. The change in velocity, Δc_{LA}, as a function of excitation pulse energy for both acoustic modes. The increase in the LA component is clearly visible, while the TA mode remains constant within experimental error.

To further confirm detection of the soliton pulse, we have performed imaging measurements, where the excitation spot is raster scanned across the metal film. Measurements of group velocity as a function of angle have been used previously to discuss phonon focussing effects in a variety of crystals (e.g. [7,9]). The soliton is expected to propagate as collimated wavefront[4], whereas the heat pulse component is subject to focussing effects. This means that (at high laser powers) for all positions where some part of the laser spot is directly opposite the detector, the bolometer signal will include a component due to the soliton. At the point where the laser and detector no longer overlapping, the soliton contribution to the bolometer signal falls to zero and the propagation distance across the crystal now becomes a significant factor in determining the measured velocity of the wavepacket. The low power excitation case is always subject to focussing effects and changes in propagation distance. Geometrical and acoustic anisotropy considerations are rather detailed, and a comparison of measured velocity for high and low power excitations for different angles is difficult. Instead, we consider the arrival time as a function of angle, and this is plotted in Fig 3. We find that for small deviations in angle from the centre point, the arrival time remains constant, and is shorter than would be expected from consideration of c_{LA}. For angles greater than ≈ 5°, the arrival time increases and continues to increase with propagation distance. This can be explained as follows; taking into account the diameter of the aperture plate and the beam divergence, we estimate the spot size at the sample is ≈ 2mm. If the soliton is not subject to significant lateral spread (divergence) as it propagates, then due to the size of the spot, we would expect to see a constant (and short) arrival time for all angles which where the laser spot and detector overlap. This is what we observe and provides additional strong evidence that we are indeed detecting the arrival of the soliton pulse.

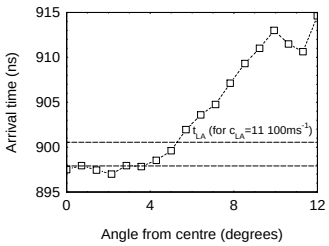

Figure 3. The arrival time of the fastest signal component as a function of angle of excitation spot from the normal.

Conclusion

To summarise, we have shown that the bolometric detection technique is sensitive to strain solitons, and the temporal resolution is sufficient to observe the small change in velocity of ultrashort THz pulses in sapphire. Imaging measurements suggest soliton propagation is strongly collimated. Future work is planned to measure the positional dependence in more detail, to observe the effects of soliton decay and to extend the preliminary measurements of soliton generation and propagation in silicon and gallium arsenide. With a better understanding of the fundamental behaviour of solitons, we can then use these ultrashort, high amplitude strain packets to probe and modify strain in other systems.

This work is has been submitted to Physical Review B as a Rapid Communication.

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