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Propagation of high amplitude strain pulses in sapphire

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The propagation of acoustic pulses in sapphire has been investigated using bolometric time-of-flight techniques. Excitation of a thin chromium film by an amplified Ti:sapphire laser pulse resulted in the generation of high-amplitude acoustic pulses. The propagation time of these across the 1-cm-thick sapphire crystal was measured using a superconducting aluminum bolometer. By increasing the excitation power, the growth of an intense fast pulse on the leading edge of the longitudinal heat pulse was observed. This was detected as an additional, separate, signal component, with a propagation time shorter than the heat pulse. This pulse is attributed to the formation and propagation of a high-amplitude coherent strain pulse. The bolometric detection technique allows high resolution images of the acoustic flux over macroscopic propagation distances, and measurements show the strain pulse propagates as a strongly collimated beam. These results demonstrate the applicability of bolometric detection techniques, in addition to Brillouin scattering and pump-probe measurements, to study the propagation of ultrashort strain pulses in crystals.

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Recent progress in ultrafast pulsed laser technology has led to increased interest in the use of picosecond strain pulses to study the acoustic properties of materials. In a typical ultrafast acoustic experiment,[1–3] femtosecond or picosecond laser pulses are absorbed in a metal film deposited on a crystalline substrate. The resulting rapid thermal expansion of the film launches a coherent acoustic strain pulse into the crystal. The spectral content of the strain pulse depends on its duration, which, in turn, is related to the optical penetration depth in the film. Typically, frequency components up to about 100 GHz can be present in the pulse. Normally, the acoustic pulses are detected using a weaker, delayed, optical pulse which probes the change in reflectance or deflection of the sample surface caused by the strain. Excitation with much higher optical power densities gives rise to very intense strain pulses and due to nonlinear effects, the strain pulse develops into a shock front containing phonon components with THz frequencies. This evolves into a stable train of acoustic solitons.[4–6] With lengths of the order of nanometers, such picosecond acoustic pulses could provide an extremely useful tool to study fundamental processes in other systems (e.g., semiconductor nanostructures).

Bolometric detection provides an established technique that can be used to study the propagation of acoustic packets across crystals. The heat pulse technique[7] 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., Ref. 8). Because of the ability of the technique to distinguish between separate phonon modes owing to their different speeds, measurements of this type continue to provide a suitable method to study a range of material properties, as varied as, e.g., hot carrier energy relaxation in semiconductor nanostructures and the role of nonradiative recombinations on the performance of optoelectronic materials. An advantage of being able to use bolometric detection techniques is the ability to study, with high spatial resolution, the angular dependence of acoustic flux propagation (phonon imaging). Heat pulses may also be generated by optical excitation of metal films. However, in this case, the optical pulse used is typically of order nanoseconds in duration and of much lower peak power than used to generate coherent strain pulses in picosecond acoustics experiments.

In this work, granular superconducting bolometers were used to detect the acoustic pulses generated by ultrafast pulsed laser excitation of a metal film on sapphire. The aims were as follows: (1) to demonstrate that superconducting bolometers offer sufficient sensitivity and temporal response to detect the acoustic packet, and (2) to image the acoustic flux following propagation over macroscopic distances. If these could be achieved, then bolometric detection would be shown to be a useful additional technique for the further study of the propagation of short high-amplitude strain pulses in crystals.

The sapphire sample used in the experiments was a highly polished single crystal of dimension $9\times5\times10$ mm^{-3}, with the c axis perpendicular to the 9×5 mm^2 surface. On to this surface, a 100-nm-thick chromium film was deposited by thermal evaporation. On the opposite face of the crystal, superconducting aluminum bolometers were fabricated. The sample was mounted in an optical access helium cryostat and maintained at a temperature of $T_0\simeq2$ K, at the midpoint of the superconducting transition of the bolometers. The chromium film was excited with pulses from a regeneratively amplified Ti:sapphire laser system (Coherent "Libra"). The average power of the output beam is 1 W at 800 nm, and at a repetition rate of 1 kHz, the output beam carries approximately 1 mJ per pulse. Pulses of $\simeq$200 fs (measured using an autocorrelator) were directed onto the sample and focused to a spot size of 200 μm diameter. The optical pump fluence was varied by inserting calibrated neutral density filters into the beam path, and the upper limit was 1.75 mJ cm^{-2}, above which the Cr film was damaged by the beam. When determining the pulse energy, all losses in the optical setup, e.g., reflections from the cryostat windows, and reflection from the Cr film were taken into account. Therefore, the values given are for the energy absorbed in the Cr film and the maximum of this is about 10% of the energy available at the laser output. The generated acoustic packets were detected by measuring the change in resistance of the constant current biased bolometer. This signal was amplified and detected us-

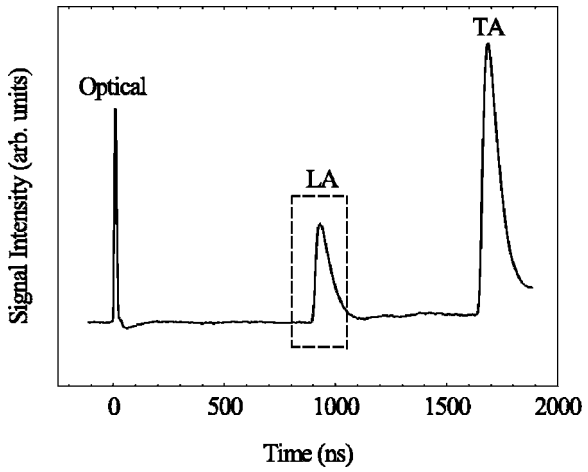

FIG. 1. A typical bolometer trace, obtained for a pulse energy density of about 0.09 mJ cm^{-2}. The boxed area is shown in more detail in Fig. 2.

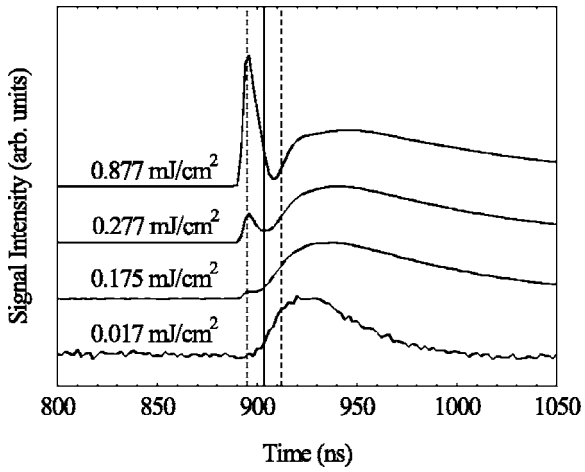

FIG. 2. The LA components of the bolometer signals for a range of excitation energy densities. The signals have been normalized to the LA peak signal intensity and offset for clarity. Also marked on the figure is the range of arrival times which would correspond to published values of both measured and calculated sound velocities for the longitudinal mode in sapphire (the center line is the average of these).

ing a high speed digitizer. To make an image of the spatial dependence of the acoustic strain packet propagation, the laser spot was raster scanned across the Cr film using a pair of galvanometer mirrors under computer control.

A typical bolometer signal obtained at low excitation fluence is shown in Fig. 1. Three distinct peaks are clearly visible, and are marked on the figure. The onset of the arrival of the first peak, at $t \simeq 0$ ns, corresponds to the optical response of the bolometer (a small amount of the incident light falls on the bolometer) and provides the zero time reference point. The second and third peaks correspond to the arrival of the different modes in the acoustic wave packet; the second peak arrives at a time consistent with the arrival of longitudinal acoustic (LA) phonons, and the third peak is due to transverse acoustic (TA) modes, both of which traverse the substrate at their respective speeds of sound. These are attributed to incoherent phonon heat pulses generated by thermal relaxation of the Cr film. Phonon propagation in sapphire has been widely studied experimentally and theoretically, and the obtained LA and TA phonon velocities are in the ranges $c_{LA}=10\,500-11\,230$ m s^{-1} and $c_{TA}=6000-6300$ m s^{-1}, respectively.[9–14]

Figure 2 shows in more detail the change in the characteristics of the LA mode bolometer signals for a range of absorbed fluence. At $\gtrsim 0.1$ mJ cm^{-2} an additional signal component is observed. With increasing fluence, the additional peak increases in intensity, and becomes clearly distinguishable from the slower LA heat pulse component. To properly attribute the peak it is necessary to consider its time of flight across the sapphire substrate. From the start of the rise of the heat pulse signal, the LA phonon velocity is measured as $c_{LA}=11\,150\pm 30$ m s^{-1}. This falls within the range of measured and calculated values of c_{LA} found in the literature and marked on Fig. 2. In all cases, the additional signal we observe appears to arrive several ns earlier than the heat pulse, and its velocity is measured as $11\,240\pm 30$ m s^{-1}, which is about the same as the fastest reported speeds of longitudinal sound in sapphire. No such "fast" pulse is seen on the leading edge of the TA heat pulse at high fluence. At low fluence, $c_{TA}=6138$ m s^{-1}, which is in excellent agreement with published values for the transverse phonon velocity in *c*-axis sapphire. Above 0.1 mJ cm^{-2} a gradual slight decrease in the measured TA velocity is observed. This is possibly due to an increased scattering of high frequency phonons generated at the higher fluences (film temperatures) leading to a small increase in the time of flight.

To discount the possibility that the fast LA pulse is a detector artifact, a number of additional measurements for a range of fluences were carried out. A possible explanation for an apparent increase in the velocity could be that the detector temperature was too low. In this situation, only the peak of the acoustic pulse, which has an amplitude sufficiently large to "push" the bolometer onto its superconducting transition, would be detected. Then, the measured arrival times would suggest a velocity lower than expected. Increasing the fluence would lead to an increase in the intensity of the acoustic pulse and hence a decrease in the onset time of the signal. The result would be an increase in the apparent velocity of *both* the LA and TA components towards the limit of the speed of sound in sapphire. In addition, the variation of the heat pulse signals with fluence would be highly nonlinear at low fluences. No such effects were observed in the measurements. Another potential error in the measurements arises with the possibility of signal saturation at the highest fluences. However, the bolometer signals exhibited clear, well-defined peaks, with none of the flattening that is associated with signal saturation. Finally, the ratio of the heat pulse intensities (LA:TA) remains constant with increasing fluence, which could be expected if the bolometer operating point is optimized.

The fast pulse at high fluences may be attributed to the formation and propagation of a coherent strain pulse. It was already established in pump-probe measurements that the ultrafast excitation of a metal film gives rise to the generation of coherent strain pulses. The spectral content of such pulses is typically $\lesssim 100$ GHz and so they would be expected to

propagate nondispersively over macroscopic distances in high-quality sapphire with little scattering. Furthermore, the wave vectors of phonons in such pulses would be distributed in a very narrow range of angles close to normal to the film. Therefore, the time of flight of the strain pulse from the generation point to the bolometer should correspond closely to the speed of low-frequency sound in sapphire at 2 K. On the other hand, the heat pulse consists largely of high-frequency (THz) thermal phonons emitted with wave vectors in a wide angular range. The combined effects of acoustic anisotropy and phonon scattering will result in most of the energy in the heat pulse reaching the bolometer slightly later than expected on the basis of the speed of low-frequency sound in sapphire.

If the fast pulse is due to the detection of a coherent strain pulse, then it is interesting to consider how the bolometer, which has a response time of order nanoseconds, can so effectively detect an acoustic pulse lasting only a few picoseconds. Probably a very small fraction of the energy of the pulse is thermalized in the bolometer due to phonon scattering and the resulting rise in the bolometer temperature is sufficient to be detected. The temporal response will therefore be determined by the thermal relaxation time of the bolometer and the bandwidth of the detection electronics. The latter is about 0.5 GHz, which could explain the 2 ns rise time of the fast pulse. From the fall time of the fast pulse, the thermal relaxation time of the bolometer can be estimated at about 5 ns. Because this pulse is composed primarily of relatively low frequency phonons ($\lesssim$100 GHz) it is likely that it will not be thermalized very efficiently, especially compared with the heat pulse, which contains much higher frequency phonons. This might explain why there is a fluence threshold above which the fast pulse stands out clearly from the heat pulse. It was also observed that the appearance of the fast LA signal is strongly affected by the width of the excitation laser pulse. By adjusting the compressor in the regenerative amplifier, the pulse width was increased from 200 fs to 4 ps, and it was found that the threshold fluence for the appearance of the fast pulse also increased significantly, but the heat pulse was unchanged. Increasing the laser pulse length will increase the duration of the strain pulse and, hence, reduce the maximum frequency of its spectral content. Therefore, the pulse will be less effectively thermalized in the bolometer and so the threshold for the detection of the fast pulse might be increased, as observed.

An alternative explanation for the fast pulse is that the leading edge of the heat pulse travels at the speed of sound, and the fast pulse is supersonic due to the formation of a train of acoustic solitons at high strain amplitude. In this case, the leading soliton in the pulse travels at a velocity given by

$$u = c_{LA} + \frac{\eta_0(3C_{33} + C_{333})}{6\rho c_{LA}}, \tag{1}$$

where C_{33} and C_{333} are, respectively, the second- and third-order elastic constants, ρ is the density, $c_{LA}=\sqrt{C_{33}/\rho}$ is the longitudinal sound velocity, and η_0 is the peak strain amplitude. For sapphire $\rho=3980$ kg m^{-3}, $C_{33}=4.980\times10^{11}$, and $C_{333}=-3.301\times10^{12}$ kg m^{-1} s^{-2}, and so, for a strain amplitude of about 1%, u is about 0.7% faster than c_{LA}. This could, if the strain was high enough, account for the difference in the arrival times of the fast pulse and the LA heat pulse.

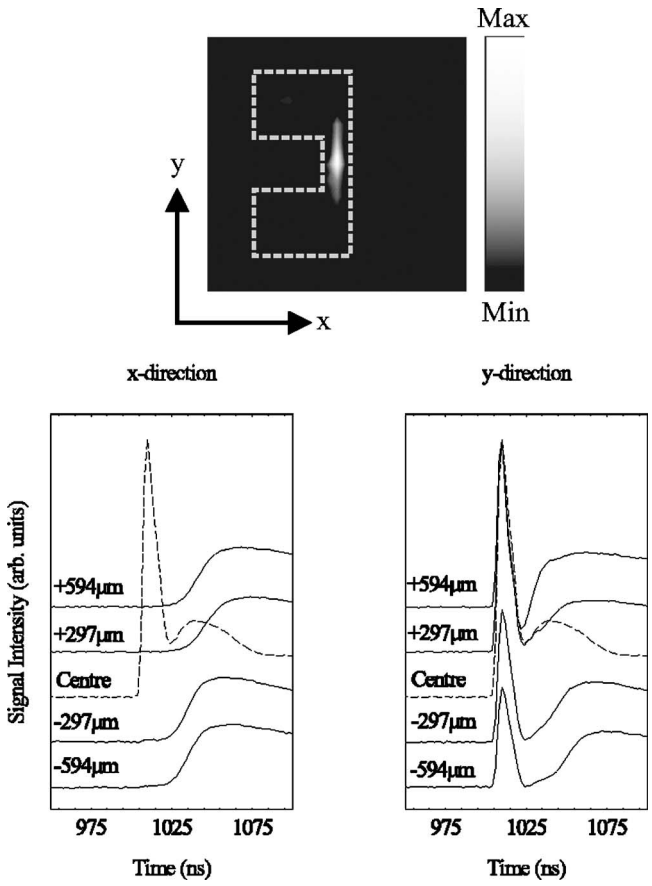

FIG. 3. An image of the supersonic pulse intensity over a 4.75 $\times$4.75 mm^2 area. The outline of the bolometer is indicated by a dashed line. The lower panel shows the bolometer signals obtained for different positions of the laser spot.

The threshold fluence for the appearance of the fast pulse is about 0.1 mJ cm^{-2}; a rough value of the initial strain in the Cr film at this fluence can be estimated using $\eta=\alpha_\ell E/C_V$, where E is the energy absorbed in the film (per unit volume); C_V is the heat capacity per unit volume; and α_ℓ is the coefficient of linear expansion. Assuming that the optical pulse is absorbed within $\approx$10 nm of the surface of the Cr film and that the hot electrons diffuse $\sim$10 nm before they relax,[15] $E=5\times10^7$ J m^{-3}. Using the room temperature values for Cr ($C_V=3.2\times10^6$ J m^{-3} K^{-1} and $\alpha_\ell=5\times10^{-6}$), an initial strain of 10^{-4} is obtained at a fluence of 0.1 mJ cm^{-2}. This is typical of the strain amplitudes used in ultrafast acoustics experiments, but its small magnitude appears to rule out the possibility that the fast pulse is due to the supersonic propagation of strain solitons. However, at the higher fluences used in this experiment, the strain amplitude reaches about 0.15%. This should be sufficient for the formation of solitons, which may assist detection since the higher frequency spectral components present in the solitons would be more readily thermalized and detected by the bolometer. However, the corresponding change in speed would be barely detectable given the experimental limitations.

The direction dependence of propagation of the fast pulse present at fluences $\gtrsim 0.1$ mJ cm^{-2} was investigated using imaging techniques, in which the excitation laser spot is raster scanned across the Cr film. An image of the pulse is shown in Fig. 3; the scan area is 4.76×4.76 mm^2. Only when the excitation point is directly opposite the active area of the bolometer does the detected signal contain the fast LA component. In fact, the image of the fast pulse matches closely the size and shape of the bolometer convolved with the source dimensions. This indicates there is no significant lateral spreading of the fast pulse as it propagates along the 10 mm length of the sample. It effectively propagates as a collimated beam. Phonon focusing effects have been widely studied (e.g., Ref. 8 and 13), and in sapphire, acoustic anisotropy leads to a trigonal focusing pattern for the LA mode that is very different from the observed behavior of the fast pulse. The images could, however, be consistent with the fast pulse being due to the generation and propagation of a coherent strain pulse. Owing to the high degree of spatial coherence across the generator width, which is much greater than the typical acoustic wavelength, there is minimal angular divergence due to diffraction. Furthermore, the fast pulse is clearly not subject to significant divergence due to scattering effects.

In summary, it has been demonstrated that superconducting bolometers are sensitive to ultrashort strain pulses propagating over millimeter distances in sapphire. The strain pulses were generated by the femtosecond laser excitation of a Cr film. At excitation fluences greater than 0.1 mJ cm^{-2}, the strain pulse was clearly resolved ahead of the leading edge of the incoherent LA heat pulse. Imaging measurements indicate that the strain pulse propagation is strongly collimated. Further work is currently underway to observe the effects of scattering and to apply bolometer techniques to study high-amplitude strain pulse generation and propagation in other materials (e.g., Si and GaAs).

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