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Phonon frequency down-conversion observed in thin metallic films

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Abstract. The frequency of phonons launched into a sapphire substrate from an electrically heated metal film has been investigated using a heat pulse technique. It was found that for an emitted phonon spectrum calculated using the Perrin and Budd model, the predicted dispersive broadening of the phonon pulse in the substrate was much greater than was observed. We have shown that scattering by subsurface damage is unlikely to account for this observation. By correlating the phonon pulse broadening with the energy dependence and the polarisation magnitudes of the heat pulse signals, we have concluded that phonon frequency down-conversion occurs within the heater film.

1. Introduction

We have obtained evidence that in low-temperature heat pulse experiments the process of phonon down-conversion takes place in the thin metal films used as pulse generators. In an earlier paper (Wybourne *et al* 1985) we reported the study of heat pulses emitted by thin metallic heaters excited on a nanosecond timescale. Broadening of the detected pulses was attributed to a layer of damage below the polished surface of the sapphire immediately beneath the heater film. Our conclusion depended on the prediction that the ballistic escape time of the phonons generated in the film was less than the anharmonic decay time of the phonons.

In the present paper, we describe further nanosecond heat pulse experiments on hydrogen-annealed sapphire surfaces which have previously been shown to exhibit greatly reduced phonon scattering (Eddison and Wybourne 1985). The experiments were performed using both NiCr and AuPd heaters which were excited at sufficiently high power levels to generate a substantial population of phonons within the heater having energies close to the zone boundary. For such high-wavevector phonons, the ballistic escape time exceeds the predicted anharmonic decay time. The low level of dispersive broadening observed within the sapphire indicated that the phonon distribution making up the heat pulse was not that emitted by the electrons, rather it was a modified spectrum resulting from frequency down-conversion. This inference was supported by other unique features of the nanosecond heat pulse data.

2. Experimental details

We used nichrome ($\text{Ni}_{50}\text{Cr}_{50}$) and $\text{Au}_{60}\text{Pd}_{40}$ heaters and $\text{Pb}_{80}\text{Bi}_{20}$ superconducting bolometers. The heaters were of thickness 27 and 10 nm respectively and were within 15% of their bulk values of resistivity suggesting that they were polycrystalline. Both the heaters and the bolometers were only $130 \times 130 \mu\text{m}^2$ in size to reduce geometric pulse broadening. They were evaporated on to opposite faces of a single-crystal sample, 5 mm long, of high-quality sapphire from Union Carbide, oriented along the (2243) axis. The surfaces of the sapphire had first been treated to a high-grade optical polish, and then etched at 1200 °C under fast-flowing hydrogen. This process has the effect of removing many outer atomic layers and hence of eliminating the grossest of the damage introduced by mechanical polishing.

The experiments were carried out in vacuum at a temperature within the superconducting transition region of the Pb–Bi, such that the bolometer impedance was 50 Ω ; typically this was around 7 K. The electrical excitation pulses were between 2.0 and 0.8 ns long, supplied by an Avtech pulse generator. Initially of amplitude 40 V, they could be attenuated by means of a Hewlett–Packard stepped attenuator. The 0 dB power density was $2.1 \times 10^3 \text{ W mm}^{-2}$. Details of the precautions taken in working with such fast heat pulses are given elsewhere (Wybourne *et al* 1985). Because the excitation pulses were so short, the excitation power density levels that we used were considerably larger than those commonly employed in heat pulse studies. The heat pulse signals obtained from the bolometer were recorded using an EG and G boxcar integrator with a timing accuracy of 350 ps. Typical responses are shown in figure 1. The bolometer time constant was determined from its absorption response to direct RF electromagnetic radiation and was found to be of the order of 30 ns. The response time therefore was longer than the heat pulses themselves and the bolometer signal did not follow the incident phonon flux directly. Although we could not determine the exact shape of the arriving heat pulses, we could at least infer their length, since this was equal to the rise time of the leading edge of the bolometer signal.

The inferences that we shall draw from our observations are based on three specific features of the observed heat pulses: their broadening relative to the excitation pulse, their dependence on the width of the excitation pulse, and the relative magnitudes of the different polarisations. We shall anticipate later discussions by summarising the results as they relate to these features.

3. Results

3.1. Pulse broadening

In the experiments, it was found that the lengths of the heat pulses arriving at the bolometer—determined from the rise of the bolometer signals—were considerably greater than that of the excitation pulse. The pulse lengths were also found to be dependent on the excitation power. At the highest power levels, the peaks of the signals became rounded, which indicated that the arriving heat pulses were acquiring significant ‘tails’. The variation in rise time with increasing power is illustrated for the transverse modes in AuPd in figure 2. In figure 3 we plot the variation of the rise times, defined as the 10 to 90% values, for all three polarisations in this material. Of the broadening,

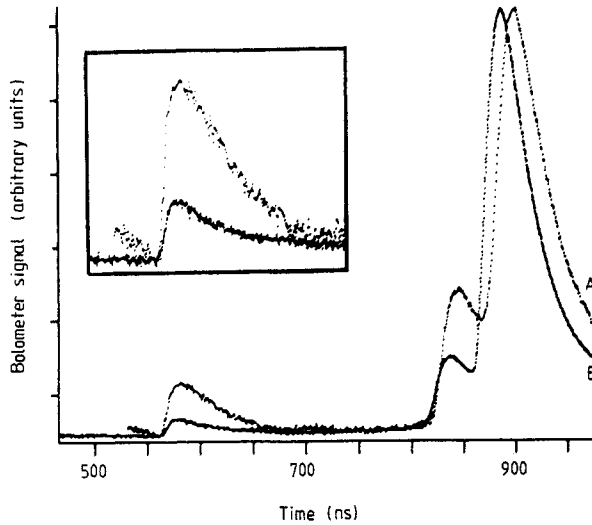


Figure 1. Typical normalised traces for (A) NiCr and (B) AuPd heaters excited at the same power level (950 W mm^{-2}). The differences in the shape of the longitudinal pulses (at 570 ns) and the relative magnitudes of the fast transverse (at 840 ns) and slow transverse (at 890 ns) modes are clearly seen. (Inset: amplification of the longitudinal modes.)

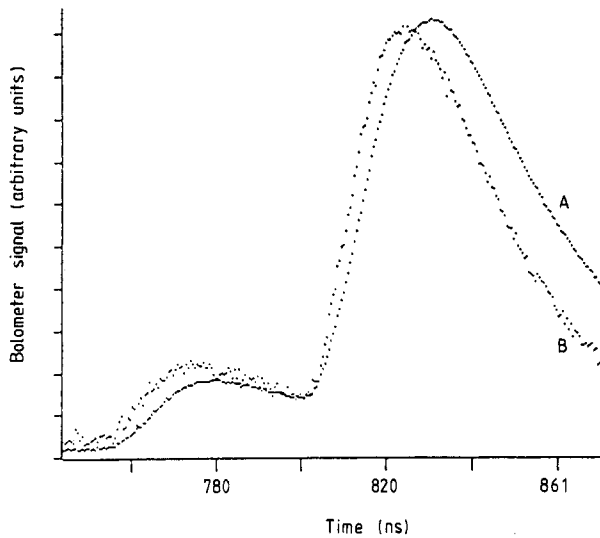


Figure 2. The transverse modes in AuPd measured at different power levels (A) 0 dB ($2.1 \times 10^3 \text{ W mm}^{-2}$) and (B) -13 dB.

approximately 0.5 ns was geometric in origin due to the finite sizes of the heater and the detector, and a further 0.8 ns was due to the band width of the preamplifier.

3.2. Energy dependence

In contrast to the results of previous workers (von Gutfeld and Nethercot 1964) using much longer heat pulses, we observed that for very short pulse lengths the magnitude

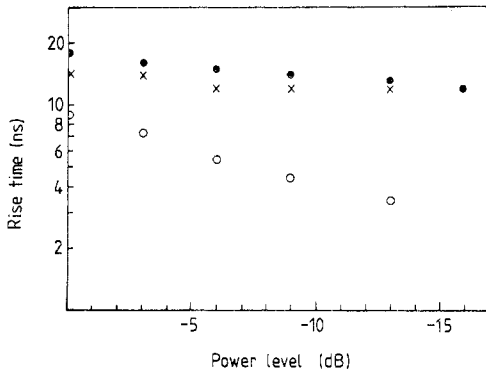


Figure 3. The variation of detected heat pulse rise time as a function of power level (○, longitudinal; ●, slow transverse, ×, fast transverse).

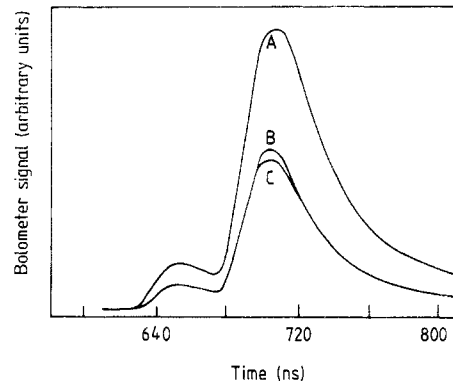


Figure 4. The transverse modes from a AuPd heater excited by electrical pulses of (A) $2.1 \times 10^3 \text{ W mm}^{-2}$ for 2 ns; (B) $2.1 \times 10^3 \text{ W mm}^{-2}$ for 1 ns, (C) $1.05 \times 10^3 \text{ W mm}^{-2}$ for 2 ns.

of the bolometer signal depended on the length as well as the power of the excitation pulse; it varied linearly with both parameters, as shown in figure 4. Halving the excitation pulse length or the pulse magnitude gave bolometer signal rise times that were the same to within the experimental uncertainty, $5 \pm 1 \text{ ns}$ demonstrating that bolometer integration effects could not be responsible for this observation. In experiments using longer heat pulses (20–100 ns) the magnitudes of the different polarisation signals are determined by a power balance condition between the electric power used to excite the heater, and the emitted phonon power. Thus, the magnitudes of the heat pulses are independent of the excitation pulse length. In the present experiments, the observation of a dependence on the pulse length implied that a steady-state situation had not been reached in the duration of the excitation pulse applied to the heater. In the accepted model of heat pulse generation by thin metal films, due to Perrin and Budd (1972), thermal equilibration in films of thickness 10 nm excited at the power levels used in our experiments should take less than 10^{-9} s .

3.3. Magnitudes of polarisations

Again in contrast to observations made using much longer heat pulses in sapphire, it was found that the magnitudes of the different polarisation modes did not depend linearly on the incident power. The results from the 10 nm AuPd film and the 27 nm NiCr film, normalised for bolometer sensitivity, are shown in figure 5 together with data from a more disordered NiCr heater, 58 nm thick, whose resistivity was twice that of the bulk. Furthermore, the ratios of the magnitudes of the fast and slow transverse modes to the longitudinal mode varied as a function of the excitation power level, with relatively more longitudinal phonons being generated at the lowest powers. It can be seen that the data for the different NiCr heaters overlap to a large extent, implying that the mechanism by which the final phonon polarisation was determined did not depend strongly either on heater thickness or on its disorder. Since an increase of the electron coupling to the transverse phonons is expected in more highly disordered films (Keck and Schmid 1976), it follows that the polarisation character of the heat pulse was not being determined

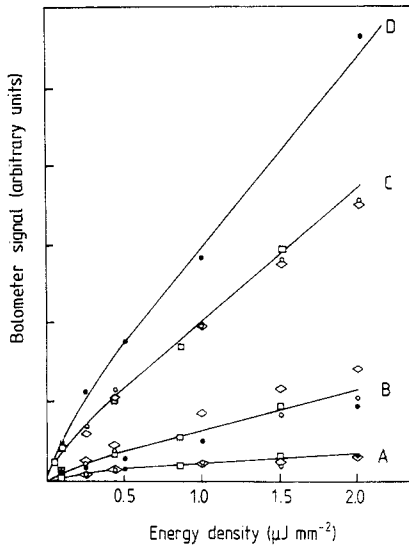


Figure 5. Magnitudes of the different modes as a function of excitation energy density: (A) longitudinal; (B) fast transverse; (C and D) slow transverse. ●, 10 nm AuPd; ◇, 27 nm NiCr; ○, 58 nm NiCr; □, 58 nm NiCr deposited on an unetched surface.

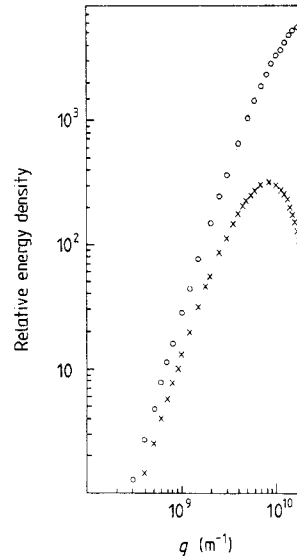


Figure 6. The phonon energy spectra calculated for the two heaters AuPd (○) and NiCr (×) for the same power density ($2.1 \times 10^3 \text{ W mm}^{-2}$). The parameters used in the AuPd calculation are as follows: deformation potential 3.77 eV; mass density $1.64 \times 10^4 \text{ kg m}^{-3}$; electron concentration $6 \times 10^{28} \text{ m}^{-3}$; reflection parameter $\eta = 12.5$.

predominantly by the electron–phonon coupling within the heater. However, the amplitude of the slow transverse mode observed using the AuPd heater was distinctly different to that observed using the NiCr heater (figure 5) showing that the heater material did in some way influence the magnitudes of the polarisations. There was much less difference between the fast-transverse-mode amplitudes for the two materials. As we shall see, these are the most significant observations and provide a key to the interpretation of the experimental data.

4. Discussion

We will first summarise the basic ideas of the heat pulse generation process. Energy incident on the heater film, whether as an optical or as an electrical pulse, appears initially as an increase in the temperature, T_e , of the electron distribution, since the electron–electron scattering rate is much greater than the electron–phonon rate. As T_e rises, the hot electrons begin to emit phonons, rapidly reaching a steady state in which the power incident is equal to the power emitted as phonons. On the assumptions of a spherical Fermi surface, no disorder and an elastically isotropic metal film, only longitudinal phonons would be emitted. For this situation Perrin and Budd (1972) have numerically solved the rate equations describing the approach to a steady state. They assumed that the emitted phonons travel ballistically within the heater film and hence that their mean free path is limited by boundary scattering. We have extended their calculations for the 10 nm thick AuPd film excited at a power density of $2.1 \times 10^3 \text{ W}$

mm^{-2} , and find that a steady-state T_e of 120 K was reached after a time of less than 500 ps. If the phonons detected by the bolometer were those directly emitted by the electrons at this temperature, then a number of experimental features of the data would be different to those we observe.

For an electron distribution of 120 K, the calculated phonon distribution emitted from the film would have a maximum at the Brillouin zone boundary of the AuPd (figure 6). The lattice spacing of FCC $\text{Au}_{60}\text{Pd}_{40}$ is 3.99 \AA , and from this we have estimated that $q_{\text{max}} = 1.72 \times 10^{10} \text{ m}^{-1}$. Taking the dispersion of a particular mode in the AuPd heater film to be described by

$$\omega(q) = v_p(2/\pi) (q_{\text{max}} \sin[(\pi/2) q/q_{\text{max}}])$$

where v_p is the phase velocity at the zone centre, we have calculated the frequency of the longitudinal mode at the zone boundary to be $\sim 6 \text{ THz}$. For this calculation we used $v_p = 3.8 \times 10^3 \text{ m s}^{-1}$ which was estimated by scaling the longitudinal velocity of gold by the weighted average of the Debye temperatures of gold and palladium for the composition of $\text{Au}_{60}\text{Pd}_{40}$.

Using a similar sinusoidal model for the dispersion in the sapphire substrate, the relative heat pulse broadening, Δt , to the time of flight, t , is given by

$$\Delta t/t = 1 - v_g(q, \sigma)/v_p(q, \sigma)$$

where $v_g(q, \sigma)$ and $v_p(q, \sigma)$ are the group and phase velocities respectively for wavevector q and mode σ . On the assumption that the phonon distribution leaves the heater film without modification, the rise times expected would be approximately 30 ns for the longitudinal phonons and 90 ns for the transverse phonons. These values greatly exceed the experimental values of 9 ± 1 and 18 ± 1 ns respectively. Since the observed broadening was much less than the predicted broadening due to the dispersion in the sapphire substrate, the conclusion must be drawn that the emitted phonons are down-converted in frequency, whilst momentum has been converted, before transit through the sapphire. A process by which this down-conversion could take place was proposed by Orbach and Vredevoe (1964). Due to lattice anharmonicity, a phonon of frequency ω splits into two others, each of frequency $\omega/2$. It was initially believed that, because of the non-linearity of the dispersion curve, only the longitudinal mode could down-convert in this way. However, more recent calculations by Tamura (1985) and by Berke *et al* (1988) show that in an anisotropic medium weak down-conversion of transverse modes may also occur. In an isotropic crystal only $L \rightarrow L + T$ and $L \rightarrow T + T$ contribute. The strength of the process can be written as

$$\tau_{\text{sp}}^{-1} = (\hbar/16\pi\rho)\varphi^2 q^5$$

where ρ is the density, φ is an anharmonic factor, and q is the phonon wavevector. Berke *et al* (1988) have determined explicit expressions for φ in terms of second- and third-order elastic constants. In the absence of a detailed knowledge of the elastic constants of the alloys, we used the low-temperature elastic constants of Au and Ni (Landolt-Börnstein 1979) to estimate the anharmonic decay rate, $\tau_{\text{sp}}^{-1}/q^5$, for AuPd and NiCr to be $6 \times 10^{-40} \text{ m}^5 \text{ s}^{-1}$ and $2 \times 10^{-40} \text{ m}^5 \text{ s}^{-1}$ respectively. We took q_{max} for AuPd to be $1.72 \times 10^{10} \text{ m}^{-1}$ and for NiCr to be $0.8 \times 10^{10} \text{ m}^{-1}$ (see figure 6), giving values for τ_{sp} of $1 \times 10^{-12} \text{ s}$, and $1.5 \times 10^{-10} \text{ s}$, respectively.

In order to determine the effect of this process it is necessary to compare its magnitude with that of the ballistic escape time of the phonons from the films. The latter can be written as (Bron and Grill 1977)

$$\tau_{\text{es}} = 4\eta d/v_g(q, \sigma)$$

where d is the film thickness and η is a frequency-independent reflection factor that is equal to the reciprocal of twice the appropriate values of Γ described by Little (1959). For the AuPd films, a lower limit of τ_{es} was obtained by using the zone-centre phonon velocity giving a value of 1.7×10^{-10} s. This is two orders of magnitude greater than the value for τ_{3p} calculated for zone boundary phonons and clearly, therefore, since the escape time increases for higher-wavevector phonons, τ_{3p} is the dominant term for phonons at or close to the zone boundary. For NiCr, however, the effects of improved acoustic matching the higher velocity combine to reduce τ_{es} for this material to 2.8×10^{-11} s. Phonons at the peak of the distribution, therefore, are much more likely to escape from the film before they are down-converted. Since the velocities in sapphire are so much greater than those in either NiCr or AuPd, down-conversion will take place predominantly in the heater films rather than in the substrate.

Support for the above model was obtained by calculating the dominant phonon frequency at which the dispersion in the sapphire would give the observed broadening of the slow transverse mode. At the maximum excitation power density (2.1×10^3 W mm⁻²), the observed rise time of 18 ns leads to a dominant frequency of 2.8 THz. To within the accuracy of this calculation, this value is approximately half the frequency of the longitudinal zone boundary phonons in AuPd estimated above; this is the result expected if anharmonic-down conversion is occurring in the heater film.

Further evidence for this conclusion was found in the observation of a considerably larger ratio of slow transverse to longitudinal modes for the AuPd than the NiCr films, which suggests that greater down-conversion has taken place in the former. The effect can be understood qualitatively in terms of the difference in the phonon distributions between the AuPd and NiCr films. In figure 6 we show the calculated energy density distribution of longitudinal phonons for the two types of heater in the steady state excited with a power density of 2.1×10^3 W mm⁻². For the AuPd the maximum number of phonons occurs at the zone boundary, whereas the NiCr has a maximum number at only half the zone boundary wavevector but at approximately the same frequency. As a result of the higher population of zone boundary phonons produced in the AuPd film, we expect much stronger down-conversion in this material than in the NiCr as indeed was observed (figure 5). The presence of large-wavevector phonons in the AuPd may also be inferred from the data of figure 3. The effect of varying the power was to vary the dominant phonon frequency of the phonon spectrum generated within the heater. Therefore, the more rapid power dependence of the AuPd longitudinal modes indicates greater dispersion and so a high dominant phonon frequency. In contrast the transverse AuPd modes, and all three NiCr modes, consist of dominant phonons with wavevectors no greater than half the zone boundary; hence they exhibit lower dispersion resulting in a lower power dependence.

Finally we discuss the possibility that the pulse broadening might be occurring by diffusive scattering in a damaged layer beneath the heater film. From simple diffusion theory, the broadened pulse width is given by

$$\Delta t = \delta^2 / \frac{1}{3}(v_g(q, \sigma))^2 \tau(\omega)$$

where δ is the thickness of the layer, and $\tau(\omega)$ the scattering lifetime. In our earlier

attempt (Wybourne *et al* 1985) to ascertain the importance of surface defect scattering, we performed an experiment using a NiCr heater on an unetched surface. The rise times of the signals were greater by between 10 and 25% but the mode ratios were the same as those from the etched sample (figure 5). This showed that surface defect scattering was contributing to the heat pulse broadening in the unetched sample. Therefore, residual damage remaining after the etching process used for the present samples could be a source of broadening. The most likely surface defects to scatter the phonons are dislocations introduced by the cutting and mechanical polishing of the sample. For static strain scattering from dislocations, the scattering rate is given by (Nabarro 1951).

$$\tau^{-1} = N_D \gamma^2 b^2 (\omega/2\pi)$$

where N_D is the number of dislocation lines per unit area, γ is the Grüneisen parameter (≈ 1), b is the magnitude of the Burgers vector (3×10^{-10} m) and ω is the phonon frequency. If we suppose that all of the extra broadening observed from the unetched surface was entirely due to dislocations, we estimate that for a depth of damage $\sim 1 \mu\text{m}$ (Hockey 1972), N_D would be $\sim 10^{17} \text{ m}^{-2}$, a value that is between 10 and 12 orders of magnitude greater than the bulk dislocation density. For such carefully prepared surfaces this high value is very unlikely.

5. Conclusions

In conclusion, we believe that our data indicate that the longitudinal phonons emitted by the electrons in a thin heater film are down-converted in frequency and are also mode converted in polarisation before being radiated into the substrate. In general it seems that the mechanism responsible for pulse broadening is a combination of scattering by subsurface damage and dispersive propagation of the phonon flux within the sapphire substrate. In the present work it appears that the dispersive broadening was the dominant effect because of the use of the hydrogen-annealed surfaces.

If these conclusions are generally valid, one consequence is that the standard phonon radiation model (Perrin and Budd 1972), which assumes ballistic flow in the heater film and in the substrate, needs to be modified to take account of frequency down-conversion. This work is currently in progress.

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