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1990 J. Phys.: Condens. Matter 2 1475

(http://iopscience.iop.org/0953-8984/2/6/007)

J. Phys.: Condens. Matter 2 (1990) 1475–1483. Printed in the UK

Phonon dispersion sheets and group velocities in GaAs

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Received 12 May 1989, in final form 25 August 1989

Abstract. Phonon frequencies on two dispersion sheets for TA modes have been determined by inelastic neutron scattering in the (001) and the (01$\bar{1}$) planes at 12 K. Phonon group velocities for these phonons have been derived. The results are used to calculate heat transport properties. Comparison with results from phonon-focusing experiments shows good agreement and yields an estimate of the lifetime of ballistic phonons.

1. Introduction

The semiconductor GaAs is attracting much interest for technical applications since crystals of very high quality have become available with almost perfect stoichiometry and crystal structure. The lattice dynamics of this material was investigated by Dolling and Waugh (1965), who have measured phonon dispersion curves for the three principal symmetry directions and compared them with model calculations. Since then several other lattice-dynamical models have been developed. But it was difficult to decide which of the models gives the best representation of the features of GaAs. In this context we found it necessary to produce new reliable experimental data. Therefore, we have determined phonon dispersion curves at about 12 K with increased precision and in more symmetry directions, and we have compared these with the available models (Strauch and Dorner 1990). To allow a test of the different models beyond the comparison with measured phonon frequencies, we have determined phonon eigenvectors at the X- and L-points (Strauch and Dorner 1986).

The propagation of 'non-equilibrium' phonons is investigated in heat-transport experiments at low temperatures (Eisenmenger *et al* 1984, Bron 1985, Eisenmenger and Kaplyanskii 1986, Anderson and Wolfe 1986). The experimental work in GaAs is reviewed by Ulbrich (1985), (see also Wolfe 1989), theoretical work is reviewed by Tamura (1986). Until recently these experiments could be well explained by the propagation of long-wavelength phonons, and, e.g., the phonon-focusing patterns could be reproduced with the mere knowledge of the elastic constants. Calculations by Northrop *et al* (1985) using various lattice-dynamical models resulted in different phonon-focusing patterns, indicating the different degree to which the elastic constants are reproduced by the different models. At the present time these experiments are being carried into the frequency and wavevector regime where dispersive effects are starting to be observed (see Hebboul and Wolfe 1989 and references therein). The different lattice-dynamical

0953-8984/90/061475 + 09 $03.50

properties predicted by the different models will have even more pronounced effects for larger wavevectors, as can be anticipated from two rather different dispersion sheets obtained from two different models by Hebboul and Wolfe (1989) or from the difference in the dispersion curves (Strauch and Dorner 1990). An experimental determination of a larger set of low-frequency, large-wavevector phonons seems to be desirable, and we have thus determined two phonon dispersion surfaces for (dominantly transverse) acoustic phonons, one for so-called slow transverse phonons in the (001) plane and one for fast transverse phonons in the $(01\bar{1})$ plane.

Phonon frequencies and eigenvectors as harmonic properties are those data which can be obtained by inelastic neutron scattering (Dorner 1982). But phonon lifetimes due to anharmonicity at low temperature are too long to be observed within the energy resolution possible today. A particular development of the neutron spin-echo technique (Zeyen 1988) may change this situation in the future. However, there is a way to obtain information on lifetime and mean free path of phonons by determining phonon group velocities by inelastic neutron scattering and comparing them with results from the heat transport experiments.

The experiment is described in § 2; the results for the dispersion sheets, group velocities and phonon lifetime will be described in § 3 and the paper is summarised in § 4. A short account of this work has been published in Dorner and Strauch (1989).

2. Experiment

A single crystal (5 cm diameter, 4 cm height) of high quality (resistance $\rho \simeq 10^7\ \Omega$ cm), kindly supplied by Dr Stath of Siemens AG, was mounted in a Displex cryostat cooled to about 12 K. We used the lattice constant $a = 5.646$ Å. The experiments were performed on the three-axis spectrometer IN3 of the ILL with a Cu(111) monochromator and the pyrolytic graphite (002) analyser fixed to $k_F = 2.662$ Å^{-1}. A graphite filter in front of the analyser served to reduce second-order contamination.

To determine the dispersion sheet of the lowest (slow TA) phonon frequencies in the (001) plane we performed constant-$\boldsymbol{Q}$ scans within the section of the Brillouin zone between the [100] and the [110] direction (see figure 1). The distance between neighbouring wavevectors was 0.02 RLU (reciprocal lattice units) in the frequency range $0.5 < \nu < 1$ THz, 0.014 RLU for $1 < \nu < 2$ THz and 0.05 RLU for higher frequencies up to the zone boundary.

In the $(01\bar{1})$ plane we examined the second-lowest (fast TA) dispersion surface in the irreducible quarter of the plane spanned by the [100] and [011] directions (and including the [111] direction), see figure 2 below. The distance between neighbouring $\boldsymbol{Q}$ vectors was 0.033 RLU in the [100] direction and 0.025 RLU in the [011] direction. In each plane we determined about 600 phonon frequencies.

3. Results

3.1. Dispersion sheets

The frequency value for each wavevector has been determined by a least-squares fitting routine. The statistical error of these frequencies is in the order of 0.002 THz, which is comparable to the experimental reproducibility for the frequencies of neighbouring

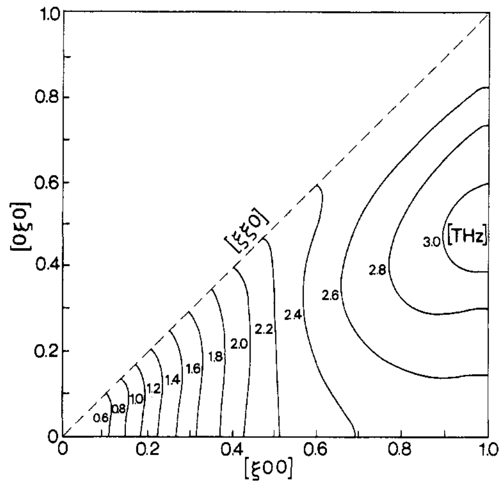

Figure 1. Experimentally determined dispersion sheet of the lowest-frequency (slow transverse) phonons with wavevectors in the (001) plane in GaAs, represented by equal-frequency contour lines.

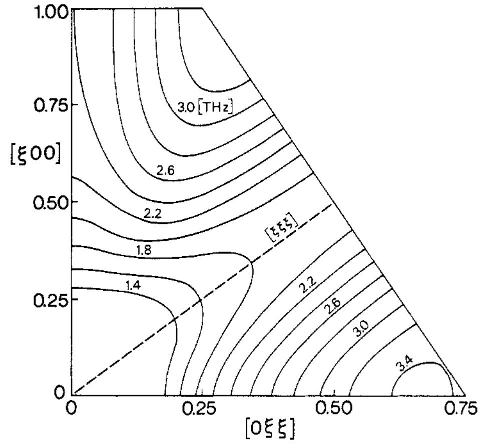

Figure 2. Experimentally determined dispersion sheet of the second-lowest (fast transverse) phonon frequencies for GaAs in the $(01\bar{1})$ plane, represented by equal-frequency contour lines. In [ξ00] and [ξξξ] directions the frequencies are degenerate with those from the dispersion sheet of lowest frequencies.

wavevectors and which represents a relative error between neighbouring values. The absolute uncertainty of the frequencies is in the order of 0.02 THz due to possible systematic mis-settings of the three-axis spectrometer.

The frequencies lie on two dispersion sheets, and the corresponding wavevectors point to the corners of small squares in reciprocal space. For each square the fitted frequencies have been smoothed by averaging over the four adjacent values, thus creating a frequency value situated in the centre of the square. At boundaries of the area of measured frequencies, such as symmetry directions and Brillouin-zone boundaries, we have mirrored the experimental data into the neighbouring areas to avoid boundary problems in the averaging procedure. The resulting dispersion surfaces derived in this way are represented by constant-frequency contours as shown in figures 1 and 2.

3.2. Group velocities and heat transport

In the present study, the phonon group velocity (the gradient of the frequency) lies always in the plane of measurement for symmetry reasons, and its direction is perpendicular to the constant-freqeuncy contours. One can deduce from figures 1 and 2 that there are preferred directions of group velocities, such as [100] in figure 1. As a consequence of this there appears a preferred direction of energy transport by non-equilibrium phonons (Ulbrich 1985, Stock *et al* 1985, Ramsbey *et al* 1988, Tamura and Harada 1985, and others). This phenomenon is known as focusing (Taylor *et al* 1969).

The spectral phonon intensity impinging on the detector has been analysed by Tamura (1983). In the experiments to be discussed below the more natural entity is time.

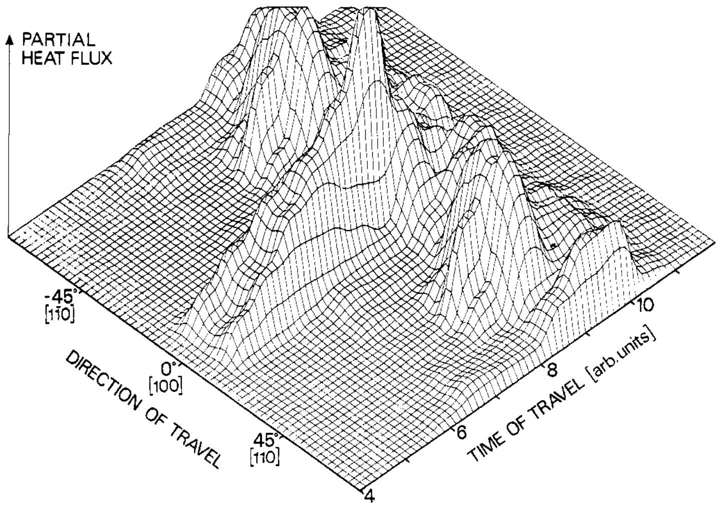

Figure 3. Partial heat flux $d^2E(t)/dtd\varphi$ through a GaAs crystal as derived from the experimental determination of phonon group velocities in the (001) plane, see figure 1.

We obtain for the phonon intensity passing the detector surface element (subtended by the solid angle $d\Omega$) per unit time

$$d^2E/dtd\Omega = (v^2/r)(d^2E(\boldsymbol{v})/dv\, d\Omega) = (v^2/r)\varepsilon(\boldsymbol{v}) \quad (1)$$

with the energy density (per velocity volume element d^3v)

$$\varepsilon(\boldsymbol{v}) = v^2 \sum_{qj}{}' \hbar\omega(\boldsymbol{q}j)n(\boldsymbol{q}j)\delta[\boldsymbol{v}(\boldsymbol{q}j) - \boldsymbol{v}]. \quad (2)$$

The prime on the summation sign in equation (2) indicates that the sum is restricted to phonons with group velocities

$$\boldsymbol{v}(\boldsymbol{q}j) = \partial\omega(\boldsymbol{q}j)/\partial\boldsymbol{q}. \quad (3)$$

from within d^3v.

In the calculations we have used differences rather than differentials in equation (3). In practice, we have derived orthogonal components v_1 and v_2 at the position of an experimentally observed frequency as the average of v_1 and v_2 from the surrounding four frequency values created by the smoothing procedure described above. In this way we have obtained a value for the group velocity corresponding to each experimentally investigated position (q_1, q_2) in the Brillouin zone. We have taken care to always normalize to constant mode density in reciprocal space.

From these results and with the help of equation (8) we have calculated the 'partial' (two-dimensional) heat flux shown in figures 3 and 4. (The determination of a sufficient number of phonon frequencies in three dimensions is beyond the time available on spectrometers for inelastic neutron scattering.)

In this analysis the occupation factor $n(\boldsymbol{q}j)$ has been taken as a constant for all modes. For a sample at low temperature, for instance at 2 K, the (equilibrium) Bose factor is practically zero for all frequencies above 0.05 THz. But the artificial excitation of non-equilibrium phonons by a laser pulse does not necessarily generate a Bose occupation but populates phonons preferably in the lowest sheet of dispersion, because these have

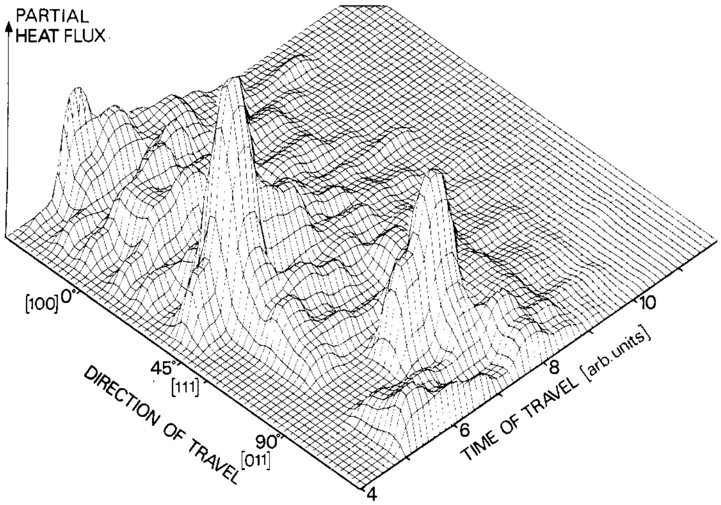

Figure 4. Partial heat flux $d^2E(t)/dt d\varphi$ through a GaAs crystal as derived from the experimental determination of phonon group velocities in the $(01\bar{1})$ plane, see figure 2.

a long lifetime. A constant occupation factor is then the simplest assumption to start with.

For the results of figures 3 and 4, the arriving heat has been sampled in angular intervals of $\Delta\varphi = 2.5°$. The zero of time of travel is given by the creation of phonons on one side of the crystal. In other words, the time axis gives the arrival time of phonons at the other end of the crystal. Note that the distance from the source point to the opposite face of the crystal has been assumed to be constant. This means that figures 3 and 4 correspond to a crystal of hemispherical shape with the source point in the centre of the sphere.

Each of the results in figures 3 and 4 is derived from one phonon dispersion sheet in two dimensions of reciprocal space. On the other hand, transport experiments with non-equilibrium phonons include all occupied phonons. Therefore, a comparison of these latter results with ours is limited. Nevertheless, as we will show, some conclusions can be drawn.

Heat transport by phonons depends upon the direction as well as on time of travel. For example, the TA phonons in the lowest dispersion sheet produce a strong focusing in and around the [100] direction (see figure 3). In the early signal a narrow maximum of heat transport even appears about 10° away from the [100] direction towards [110]. This effect can be easily understood by inspecting figure 1. At lower energies the contour lines are closer together due to the higher group velocity. In this region many constant-frequency contours have a large section with an inclination of about 10° to the [100] direction. Therefore, many modes with group velocities 10° away from the [100] direction contribute to the early signal.

From figure 3 we see that the partial heat transport is confined to ±10° away from the [100] direction for a long time. Only as late as 1.7 times the travel time of the fastest signal could one expect a signal in the [110] direction (at 45°).

Figure 4 shows that the second-lowest TA dispersion sheet in the $(01\bar{1})$ plane produces much less focusing. Around [100] again a maximum appears about 10° away but now in

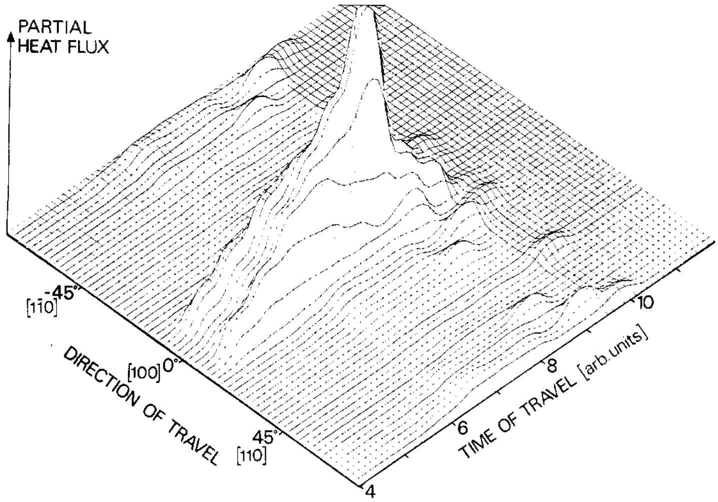

Figure 5. Partial heat flux $d^2E(t)/dt d\varphi$ as in figure 3, but 'filtered' for frequencies $0.7\text{ THz} < \nu < 2.2\text{ THz}$ corresponding to the Pb tunnelling diode (Ulbrich 1985).

the [111] direction. The signal along [011] is not as late as that in figure 3, but is late nevertheless. A particular phenomenon is the focusing at about 40° which is distinctly not the [111] direction. From figure 2 we see that the frequencies for $\boldsymbol{q} \parallel [111]$ are very low, and so are the group velocities. The focusing at about 40° can be explained by the many contour lines which are almost parallel to the [111] direction, and correspondingly many modes have group velocities perpendicular to [111]. This is an extreme situation, where the energy transport is almost perpendicular to the wavevectors of the contributing phonon modes.

For a real experiment of heat transport by non-equilibrium phonons it is not directly evident to what extent the second-lowest TA modes, figures 2 and 4, are activated (occupied). However, the lowest-frequency modes, figures 1 and 3, are expected to have a high occupation of non-equilibrium phonons. Therefore, we will concentrate on these modes in the following comparison of our results with data from heat transport experiments.

The superconducting Pb detector generally used is sensitive only for frequencies above 0.7 THz. Also, during the generation of the heat pulse phonons of higher energy will have a decreasing or even vanishing population. Arbitrarily we assume a cut-off frequency of 2.2 THz. Therefore, we have performed a new analysis of our data and derived the partial heat flux by phonons within a window of $0.7\text{ THz} < \nu < 2.2\text{ THz}$ with the results shown in figure 5. The main difference between figures 3 and 5 is the disappearance of slow-velocity heat transport in the 45° direction.

3.3. Phonon lifetime

In the preceding section the spreading of the heat pulse was idealised by the assumption of negligible anharmonic decay and defect scattering, i.e., infinite lifetime and thus infinite mean free path, in which case the occupation numbers $n(\boldsymbol{q}j)$ and the group velocities $\boldsymbol{v}(\boldsymbol{q}j)$ in equation (2) do not change in time. We will now consider the effect of finite lifetime.

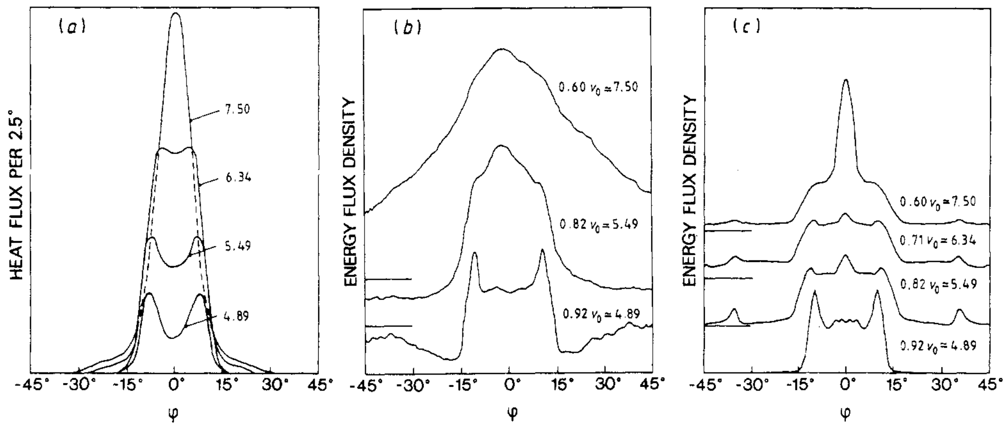

Figure 6. Heat flux in the (001) plane of GaAs for different times of travel (*a*) as derived from experimentally determined group velocities (this paper), (*b*) as determined from heat flux measurement at about 2 K by Stock *et al* (1985), and (*c*) as calculated with a bond-charge model by Schreiber *et al* (1987). The numbers on the curves give the velocities relative to the maximum group velocity v_0. The correspondence of the velocities given here to the times given in figures 3–5 is $t = 4.5v_0/v$.

For the early signals by high-velocity phonons, the experimental focusing patterns exhibit a sharp crown-like structure (Stock *et al* 1985, Ramsbey *et al* 1988), very similar to our results for times up to 5 (figure 5). The later signals are much more blurred and broadened than expected from our results. This is detailed in figure 6, where we compare our results with the experimental heat-transport data and bond-charge model calculations (Schreiber *et al* 1987). The angle φ in figure 6 has the same meaning as that in figure 5. The signal in figure 6(*a*) increases with arrival time due to the assumption of constant $n(qj)$ and increasing phonon density for decreasing group velocities, i.e., increasing frequency.

Although the experimental heat flux is carried by three-dimensional phonons with limited lifetime, analysis of the angular distribution of the heat transport for different arrival times (different group velocities) should allow a distinction between ballistic phonons with a mean free path longer than the length of travel in the crystal and those phonons with shorter lifetimes. Ballistic phonons should have an angular distribution of the heat flux similar to the one derived from the inelastic neutron scattering experiment. Phonons with a shorter lifetime and mean free path, on the other hand, decay on their way into other phonons and/or are scattered by defects (Tamura 1986). Even though in the decay processes (total) momentum (phonon wavevector) and energy (phonon frequency) are conserved, the group velocities are not conserved and thus the heat flux is not. In a scattering process not even the momentum is conserved. Thus in both cases the angular distribution of the heat transport is changed from the one assuming infinitely long mean free path. Therefore we can conclude that the onset of the deviation of the real heat flux from the predicted one indicates the onset of non-ballistic transport.

From the two-dimensional dispersion and assuming infinite lifetime one would expect a crown-like heat distribution for arrival times up to $t \sim 6$ $(v/v_0 \sim 0.75)$ with decreasing width, see figure 6(*a*). The experimental crown-like structure becomes smoother for *shorter* times, $t \sim 5$ (corresponding to $v/v_0 \sim 0.9$), and this we take as the onset of non-ballistic transport in which the mean free path is equal to the crystal thickness. With

$v_0 = 3.34 \times 10^5$ cm s^{-1} (for long-wavelength TA phonons) and a path length $l = 2$ mm we find a lifetime of

$$\tau = l/v \sim 0.67 \times 10^{-6} \text{ s}$$

of the dominating phonons. This is in good agreement with results from other studies: Fieseler *et al* (1988) find $\tau = 0.48 \times 10^{-6}$ s from a comparison of their Monte Carlo simulations of the heat transport including isotope scattering (where the scattered phonons propagate mostly along or slightly off the [100] directions) with the experimental data. A slightly larger number is obtained by Ramsbey *et al* (1988) using the same method. Tamura (1984) in his calculation of the lifetime due to isotope scattering in GaAs obtains

$$\tau = 0.136 \times 10^{-6} \, \nu^{-4} \text{ s THz}^4$$

giving $\tau = 0.56 \times 10^{-6}$ s for $\nu = 0.7$ THz.

4. Summary

We have determined two phonon dispersion sheets by inelastic neutron scattering, and from these we have derived group velocities. Our data provide additional information for the design of lattice-dynamical models (and determination of their parameters) which may then be used for various predictions or interpretations of theoretical results. Comparison of our dispersion sheet of figure 1 with the theoretical one of Hebboul and Wolfe (1989)—their figure 3(*c*)—shows clearly the superiority of their bond-charge model over their shell model, at least in the frequency range of interest to them, and thus the failure of the latter in their calculations is to be expected.

Finally, we have obtained a hypothetical heat flux—from the two-dimensional set of group velocities—predicted for phonons with infinite lifetime. From the change of the angular distribution of the experimental heat transport in comparison with theoretical results (assuming harmonic phonons with infinite lifetime) we have estimated the lifetime of the long-wavelength phonons travelling in and near the [100] direction. It should be noted that our conclusion about the lifetime is based on the analysis of only one dispersion sheet in only two dimensions of reciprocal space and that the number is but a rough estimate, even though in surprising coincidence with other results.

Acknowledgments

We thank Professor R G Ulbrich and his collaborators and Professor F P Wolfe for stimulating discussions. Dr Stath (Siemens) kindly donated the crystal. Mr A Oppenländer helped with the computer graphics.

References

Anderson A C and Wolfe J P 1986 (ed.) *Phonon Scattering in Condensed Matter V* (Berlin: Springer)
Bron W E (ed.) 1985 *Nonequilibrium Phonon Dynamics* (NATO ASI series B, vol 124) (New York: Plenum)
Dolling G and Waugh J L T 1965 *Lattice Dynamics* ed. R F Wallis (Oxford: Pergamon) p 19
Dorner B 1982 *Coherent Inelastic Scattering (Springer Tracts in Modern Physics* vol 93*)* ed. G. Höhler and E A Niekisch (Berlin: Springer)

Dorner B and Strauch D 1989 *Phonons '89* ed. S Hunklinger, W Ludwig and G Weiss (Singapore: World Scientific) at press
Eisenmenger W and Kaplyanskii A A (ed.) 1986 *Nonequilibrium Phonons in Nonmetallic Crystals* (Amsterdam: North-Holland)
Eisenmenger W, Lassmann K and Döttinger S (ed.) 1984 *Phonon Scattering in Condensed Matter* (Berlin: Springer)
Fieseler M, Wenderoth M and Ulbrich R G 1988 *Physics of Semiconductors* ed. W Zawadzki (Warsaw: Institute of Physics—Polish Academy of Sciences) p 1477
Hebboul S E and Wolfe J P 1989 *Z. Phys.* B **74** 35
Northrop G A, Hebboul S E and Wolfe J P 1985 *Phys. Rev. Lett.* **55** 95
Ramsbey M T, Wolfe J P and Tamura S 1988 *Z. Phys.* B **73** 167
Schreiber M, Fieseler M, Masur A, Pollmann J, Stock B and Ulbrich R G 1987 *Physics of Semiconductors* ed. O Engström (Singapore: World Scientific) p 1373
Stock B, Fieseler M and Ulbrich R G 1985 *Physics of Semiconductors* ed. J D Chadi and W A Harrison (Berlin: Springer) p 1177
Strauch D and Dorner B 1986 *J. Phys. C: Solid State Phys.* **19** 2853
—— 1990 *J. Phys.: Condens. Matter* **2** 1457–74
Tamura S 1983 *Phys. Rev.* B **28** 897
—— 1984 *Phys. Rev.* B **30** 849
—— 1986 *Phonon Scattering in Condensed Matter V* ed. A C Anderson and J P Wolfe (Berlin: Springer) p 288
Tamura S and Harada T 1985 *Phys. Rev.* B **32** 5245
Taylor B, Maris H J and Elbaum C 1969 *Phys. Rev. Lett.* **23** 416
Ulbrich R G 1985 *Nonequilibrium Phonon Dynamics* (NATO ASI series B vol 124) ed. W E Bron (New York: Plenum) p 101
Wolfe J P 1989 *Festkörperprobleme (Advances in Solid State Physics)* vol 29 ed. U Rössler (Braunschweig: Vieweg) p 75
Zeyen C 1988 *The Time Domain in Surface and Structural Dynamics* ed. G J Long and F Grandjean (Dordrecht: Kluwer) p 213