

## Max-Kade Foundation.

\*Work performed under the auspices of the U. S. Atomic Energy Commission.

†On leave from University of Bonn, Bonn, Germany.

‡In partial fulfillment of Ph. D.

<sup>1a</sup>See, for example, E. O. Kane, Phys. Rev. 146, 558 (1966).

<sup>1b</sup>D. J. Stukel, T. C. Collins, and R. N. Euwema, in *Electronic Density of States, Proceedings of the Third International Materials Research Symposium, Gaithersburg, Maryland, 1969*, edited by L. H. Bennett, National Bureau of Standards Special Publication No. 323 (U. S. GPO, Washington, D. C., 1971).

<sup>1c</sup>M. L. Cohen and J. D. Joannopoulos, private communication.

<sup>2a</sup>See, for example, F. Herman, R. L. Kortum, C. D. Kuglin, and J. L. Shay, in *Proceedings of the International Conference on II-VI Semiconducting Compounds, Brown University, 1967* (Benjamin, New York, 1967).

<sup>2b</sup>M. L. Cohen and J. D. Joannopoulos, private communication.

<sup>2c</sup>M. Cardona and F. H. Pollak, Phys. Rev. 142, 530 (1966).

<sup>3a</sup>See, for example, T. M. Donovan and W. E. Spicer, Phys. Rev. Lett. 21, 1572 (1968).

<sup>3b</sup>D. T. Pierce and W. E. Spicer, Phys. Rev. B 5, 3017 (1972); T. E. Fischer and N. Erbudak, Phys. Rev. Lett. 27, 1220 (1971).

<sup>3c</sup>D. T. Pierce and W. E. Spicer, Phys. Rev. Lett. 27, 1217 (1971).

<sup>4</sup>M. F. Thorpe and D. Weaire, Phys. Rev. Lett. 27, 1581 (1971).

<sup>5</sup>J. D. Joannopoulos and M. L. Cohen, to be published.

<sup>6</sup>K. Siegbahn, C. Nordling, G. Johansson, J. Hedman, P. F. Hedén, K. Hamrin, U. Gelius, T. Bergmark, L. O. Werme, R. Manne, and Y. Baer, *ESCA Applied to Free Molecules* (North-Holland, Amsterdam, 1969).

<sup>7</sup>D. W. Langer, Z. Naturforsch. 24a, 1555 (1969).

<sup>8</sup>G. Wiech and E. Zöpf, in Proceedings of the International Conference on Band-Structure Spectroscopy of Metals and Alloys, Glasgow, Scotland, September 1971 (to be published).

<sup>9</sup>J. P. Walter and M. L. Cohen, Phys. Rev. B 4, 1877 (1971).

<sup>10</sup>D. Brust, Phys. Rev. Lett. 23, 1232 (1969).

<sup>11</sup>D. Penn, Phys. Rev. 128, 2093 (1962).

## Measurement of Acoustic-Wave Dispersion in Solids

D. Huet, J. P. Maneval, and A. Zylbersztein

*Groupe de Physique des Solides, Ecole Normale Supérieure, Paris 5<sup>e</sup>, France*

(Received 3 August 1972)

By measuring the time of flight of heat pulses using superconducting tunnel junctions, we have obtained the frequency dependence of the energy velocity of acoustic waves in indium antimonide. This yields, with good accuracy, the dispersion relations for long wavelengths.

Dispersion of acoustic waves in a perfect lattice arises from the discreteness of the crystal-line array, and becomes important for wavelengths comparable to the dimension of the unit cell.<sup>1</sup> The frequency versus wave-vector relations ( $\nu$  versus  $q$ ) can, in general, be obtained from neutron or x-ray scattering data; however, the frequency dependence of the acoustic velocity has not been observed directly in a solid. We present here the measurement of group-velocity dispersion in indium antimonide by means of heat pulses at liquid-helium temperatures.

Acoustic phonons, having energies of a few Kelvin ( $10^{11}$  to  $10^{12}$  Hz), can be generated in a crystal by Joule heating of a metallic film evaporated on the surface. The phonons, thus produced in a wide spectrum, propagate ballistically and

reach the crystal boundary—each with its own velocity. If the film is excited with a current impulse, the front edge of the heat pulse will signal the arrival of those phonons that have the largest group velocity, and this usually corresponds to the lowest frequency, while later times will correspond to the arrival of dispersed phonons. The available detectors are quadratic and are mainly of two kinds: (i) superconducting bolometers,<sup>2</sup> sensitive to the energy flux irrespective of the frequency content of the heat pulse, and (ii) superconductor-barrier-superconductor tunnel junctions,<sup>3</sup> which are only sensitive to the phonons able to break the Cooper pairs. Therefore, they have a threshold of detection equal to the superconducting energy gap  $2\Delta$ , which usually stands inside the spectrum emitted by the metallic film.

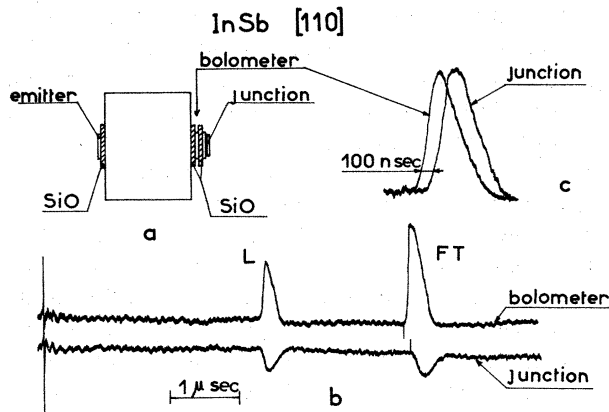


FIG. 1. (a) Sample arrangement. (b) Heat pulses detected by an aluminum bolometer (upper curve) and a tin junction (lower curve). (c) Magnification of the FT pulses showing the delay of the junction signal.

If the group velocity of the phonons, of energy  $2\Delta$ , shows dispersion, the time of flight of heat pulses will appear different when measured with a bolometer or with a tunnel junction.

The experimental arrangement for the pure single InSb sample used is shown in Fig. 1(a). The gold emitter has a thickness of 500 Å and is excited by electrical pulses of  $10^{-7}$  sec duration. On the opposite face of the sample, an aluminum film, 300 Å thick, and a Sn/oxide/Sn junction are superposed for simultaneous reception of the heat pulses. All three elements, emitter, bolometer, and junction, have an equal overall area of  $1 \times 1$  mm<sup>2</sup>, and are separated from one another and from the substrate by an SiO layer, 4000 Å thick. The bolometer has such a shape that it does not screen more than 50% of the heat flux incident on the junction.

On the recordings [Fig. 1(b)], corresponding to a 1.30-cm path along [110], the time which separates the fast transverse (FT) heat pulses, as detected by the bolometer and by the junction, is quite appreciable. It is much smaller in the case of longitudinal (L) waves, because, at a given frequency, dispersion is a decreasing function of the velocity of sound. The slow transverse heat pulse was too small to allow accurate time-of-flight measurements. Figure 1(c) represents the magnification of the FT pulses, where the two signals have been made equal by adequate attenuation of the bolometer signal, which is the largest, so as to provide all instruments with the same amplitude. The delay measured was 100 nsec, or 1.8% of the transit time, independent of

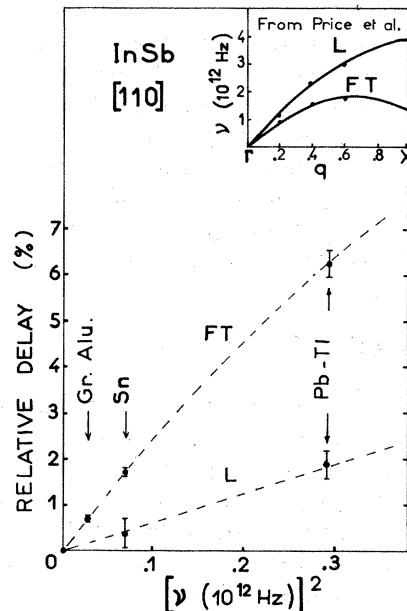


FIG. 2. Relative delays  $(t_J - t_B)/t_B$ , measured with various junctions, versus  $(2\Delta/\hbar)^2$ . Dashed lines, best fits according to the expansion of Eq. (1) (see text). Inset, taken from Ref. 7.

the emission power from 10 to 100 W/mm<sup>2</sup>. On the other hand, the energy gap of tin, as deduced from an  $I$ - $V$  characteristic curve taken at the working temperature, 1.4 K, was found to be  $2\Delta = 1.145$  meV.

The accuracy of time measurements is determined by the response times of the detectors and of the signal integrator. But, since here the signal-to-noise ratio was high, the delay could be obtained with a much better precision ( $\pm 5$  nsec) than the overall response time ( $\approx 40$  nsec) would indicate. In the case of the tunnel junction, the sharp rise of the heat-pulse signal is particularly interesting: It shows that the detection occurs at a well-defined energy threshold and confirms that the mechanism is pair breaking,<sup>4</sup> at least in the limit of low temperatures and relatively small bias currents.

We have applied this time-of-flight spectrometry to the determination of the dispersion relations along the [110] direction in InSb. We have measured the difference between the apparent arrival times  $t_B$  and  $t_J$  at the bolometer and at the junction, respectively, for several junctions made of granular aluminum ( $2\Delta = 0.72$  meV), tin ( $2\Delta = 1.145$  meV), or a lead-thallium alloy ( $2\Delta = 2.25$  meV). With the superconducting energy gap deduced, each time, from the  $I$ - $V$  character-

istics, we have plotted (Fig. 2) the relative delay  $(t_J - t_B)/t_B$  versus the square of the detection threshold frequency  $2\Delta/\hbar$ . The origin corresponds to the measurement at the bolometer, which serves then as a calibrator.

The time of flight  $t_J$  is inversely proportional to the energy velocity  $v_g = 2\pi d\nu/dq$  at the frequency  $\nu = 2\Delta/\hbar$ . If we approximate the dispersion relations, for wave vectors along [110], by a development

$$2\pi\nu = v_s q (1 - a^2 q^2 + b^4 q^4 + \dots), \quad (1)$$

we can deduce from our measurements the parameters  $v_s$  and  $a$ . We find, for the FT mode (polarization vector along [001]),  $v_s(\text{FT}) = 2.35$  km/sec ( $\pm 1\%$ ),  $a(\text{FT}) = 1.03 \text{ \AA}$  ( $\pm 5\%$ ), and can tentatively propose for  $b$  the value  $0.9 \text{ \AA}$ . For the L polarization, we find  $v_s(\text{L}) = 3.85$  km/sec, and  $a = 0.83 \text{ \AA}$ . The dashed lines of Fig. 2 represent the function  $v_s/v_g - 1$  obtained from Eq. (1) with this choice of the parameters.

We could observe the dispersion of acoustic waves because heat pulses provide phonons of large wave vectors, up to about one fourth of the Brillouin zone, far above the domain of ultrasonic waves. But the applicability of our method is limited, on the high-frequency side,<sup>5</sup> by the largest available energy gap (2.6 meV for Pb-Pb junctions). The complete dispersion curves of InSb have been obtained (with decreasing accuracy at small wave vectors, however) from studies of x-ray scattering<sup>6</sup> and, more recently, from studies of neutron scattering<sup>7</sup> at room temperature (see inset of Fig. 2). Although the first data of Ref. 7 are for  $q = 0.2$  times the distance between the  $\Gamma$  and the  $X$  point ( $1.37 \text{ \AA}^{-1}$  in InSb), that is, above our range of investigation, we substituted the neutron data at  $q = 0.2$  and  $q = 0.4$ , for the purpose of comparison, into a sine-type dependence of  $\nu$  versus  $q$ . We obtained  $v_s(\text{FT}) = 2.25$  km/sec,  $a(\text{FT}) = 0.89 \text{ \AA}$ , and for the L mode,  $v_s(\text{L}) = 3.4$  km/sec and  $a(\text{L}) = 0.78 \text{ \AA}$ . Let us recall that the velocities of sound determined by ultrasonic pulse techniques<sup>8</sup> at 0 K are  $v_s(\text{FT}) = 2.33$  km/sec and  $v_s(\text{L}) = 3.82$  km/sec.

We will discuss at last about the effect of the SiO layer between the bolometer and the junction [Fig. 1(a)]. It is not obvious *a priori* that this layer does not introduce any delay of the heat pulses, thus reducing the accuracy of our mea-

surements. To this objection, we can give two answers: (i) In the course of our experiments, on InSb along [100] and [110], and on germanium along [110], the measured time differences ranged from 290 nsec down to 15 nsec, and we have never met any inconsistency which could have been traced to SiO. (ii) A superconductor-oxide-superconductor diode has been formed alone. At low temperature, the diode was used as a tunnel junction with its detection threshold at  $2\Delta$ ; at a temperature equal to the midpoint of the superconducting transition, one arm of the diode was used as a bolometer. This very simple method, which eliminates the intermediate SiO layer, was successfully applied to the case of granular aluminum, giving exactly the same result as the first arrangement.

Differential measurements of the time of flight of heat pulses appear to be a valuable tool for investigating the onset of dispersion in solids. The essence of the heat-pulse technique is the separation of modes according to their velocities. By improved time resolution, it applies not only to the separation of different polarizations, but also, to a certain extent, to the separation of the different frequencies. This "time-domain" spectrometry makes use of broad-band, rather than monochromatic, emission, and of dispersion, which is an intrinsic property of solids.

We are indebted to B. J. Elliott for helpful discussions.

<sup>1</sup>L. Brillouin, *Wave Propagation in Periodic Structures* (McGraw Hill, New York, 1946).

<sup>2</sup>R. J. Von Gutfeld and A. H. Nethercot, Jr., *Phys. Rev. Lett.* **17**, 868 (1966).

<sup>3</sup>W. Eisenmenger and A. H. Dayem, *Phys. Rev. Lett.* **18**, 125 (1967).

<sup>4</sup>A. H. Dayem, B. I. Miller, and J. J. Wiegand, *Phys. Rev. B* **3**, 2949 (1971).

<sup>5</sup>Heat-pulse propagation at higher frequencies is practically forbidden in InSb because of the important rate of isotopic phonon scattering in this material.

<sup>6</sup>J. Jouffroy, *C. R. Acad. Sci., Ser. B* **265**, 67 (1967). However, this work does not give the dispersion relations along [110].

<sup>7</sup>D. L. Price, J. M. Rowe, and R. M. Nicklow, *Phys. Rev. B* **3**, 1268 (1971).

<sup>8</sup>L. J. Slutsky and C. W. Garland, *Phys. Rev.* **113**, 167 (1959).