H.F. PHONON TRANSMISSION AS A PROBE OF CONDENSED MATTER

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1. INTRODUCTION

Phonons are quanta of mechanical vibrations in a solid, with a spectrum extending from audio-waves up to frequencies in the terahertz range (1 THz = 10^{12}Hz), in the infra-red lattice absorption bands. Those high frequencies which are not generated by coherent ultrasonic techniques are named H.F. by convention. To characterize the quantized energy, the following equivalences are useful : $h\nu \leftrightarrow k_BT \leftrightarrow eV \leftrightarrow hc(1/\lambda_{opt})$. Here, h, k_B, e, c are respectively the Planck constant, the Boltzmann constant, the electronic charge and the velocity of light. In numbers, we have :

$$1\ \mathrm{THz} \leftrightarrow 50\ \mathrm{K} \leftrightarrow 4\ \mathrm{meV} \leftrightarrow 30\ \mathrm{cm}^{-1} \qquad (1)$$

By nature, the phonons are virtually coupled to any departure (defect or excitation) from the ideal solid. In the range specified by Eq. 1, they are in energetic resonance with superconducting gaps, Zeeman splittings, vibronic levels, etc... Besides, the typical phonon wave-length at 1 THz being ∿ 30 Å, H.F. phonons may be put in spatial interference with, for instance, Bohr orbits of shallow impurities in semiconductors, or free electrons, or semi-macroscopic objects such as evaporated thin films. We shall show in this article how the transport of H.F. acoustical phonons can bring information about (a) crystal acoustics, (b) phonon interactions and selection rules and (c) represents a possibility for imaging, restricted to low temperatures unfortunately.

2. BALLISTIC PROPAGATION

H.F. phonons achieve sufficiently long free paths at helium temperatures to exhibit ballistic (non-diffusive) propagation over macroscopic distances. This is seen in heat pulse experiments where a packet of thermal phonons produced by Joule effect in a metal film are allowed into a single crystal ; the temperature signal is then detected at the opposite end by a thin-film bolometer (Fig. 1a). Three separate signals appear in the course of time corresponding to the three acoustic branches of the ν versus q (wavevector) dispersion relation (Fig. 1b). L is for longitudinal, T_1 and T_2 are respectively for the fast and slow transverse polarizations. The velocities of sound are thus measured to within 0,5 % accuracy.

Additional information is obtained by using quantum detectors instead of wide band bolometers. Suppose (Fig. 1b) the detector threshold is 2Δ. In the case of normal dispersion, the fastest incoherent phonons will travel with the group velocities

$V_g = \partial\omega/\partial q$ taken at this particular energy, which may be significantly different (Fig. 1c) from the velocity of sound as determined by the bolometer trace. It appears indeed under magnification that the leading edges are very clearly delayed respective to one another.

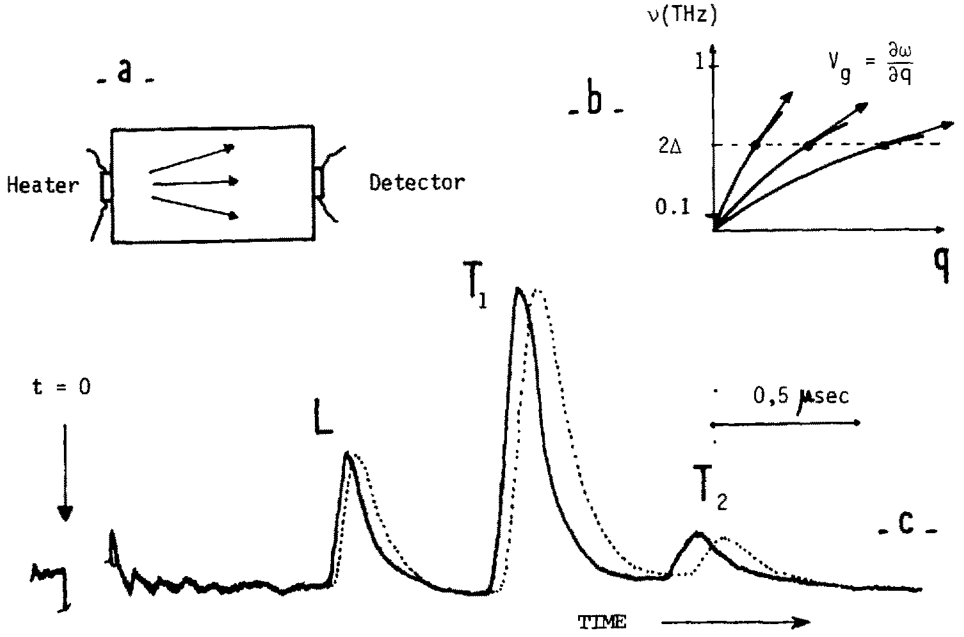


Fig. 1 : (a) InSb sample, oriented along [110], provided with thin film phonon transducers. (b) Dispersion relations for acoustic modes. (c) Bolometer (full line) and tin superconducting junction (dotted line) traces versus time as responses to a Dirac excitation. $T_o = 1.3$ K. Gap of tin : $2\Delta(\mathrm{Sn}) = 1.145$ meV.

Chromatic dispersion is in turn a useful tool for characterizing the threshold of a given detector, such as a superconducting junction in a magnetic field, or for measuring H.F. phonon frequencies when no other spectroscopy is available.

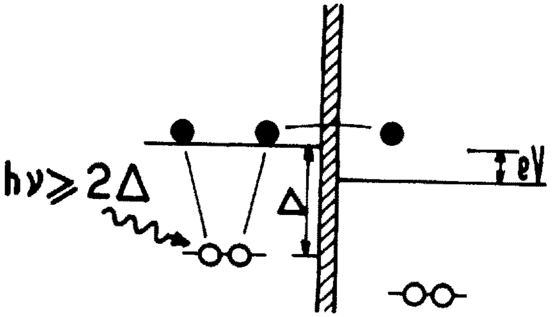
On the other hand, ballistic heat-pulse experiments provide also direct evidence of acoustic anisotropy and self-focussing effects (Ref. 1).

3. SOURCES AND RECEIVERS

- Thermal transducers. Evaporated metal films (Au, Cu, Constantan ...) are very efficient pulsed-heat radiators, with a damage limit at about 1 kW/mm^2. They can be driven by an electrical current or, alternatively, by a laser pulse. They are essentially broadband phonon generators, whose emission, in a first approximation, can be assimilated to a blackbody spectrum (Ref. 1). These sources are adequate to study the diffusion of heat taking place at ∿ 10 K and above. Second sound propagation has also been observed in exceptionaly pure NaF single crystals (Ref. 2). Superconducting bolometers, made of Al, Sn, Pb, Pb-Bi, etc..., are the most widely used wide-band detectors, featuring sensitivity and speed of response (∿ 10^{-8} sec). The main drawback is their sensitivity to the magnetic field.

- Quantum transducers. Consider the scheme of electron energies in a symmetric superconducting tunnel diode (Fig. 2).

Fig. 2 : Superconducting tunnel junction as a quantum detector. Pairs are broken by the incident phonons ; a current of quasiparticles passes through the oxide barrier.



The interelectrode oxide barrier is so formed that it is transparent to quasiparticles (normal electron tunnelling), while pair-wavefunctions on both sides remain un-correlated. The mechanism by which a H.F. phonon (or a microwave photon) is quantally detected is well established (Ref. 3). If the incident energy hν is larger than the superconducting gap 2Δ, a Cooper is broken, and a non-thermal quasiparticle current is then recorded as a signal. Fig. 1 is an example of such a process. Penetration of a smaller than critical magnetic field allows tunability to a certain extent. Fig. 3 shows how the frequency range 10 to 1000 GHz can be scanned by superconducting junctions.

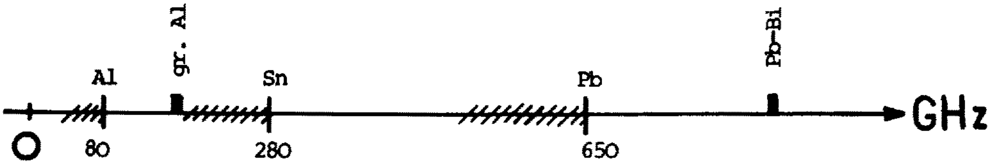


Fig. 3
Detection threshold of superconducting tunnel junctions. The hatched sections indicate the range of magnetic tunability. Granular Al and Pb-Bi are non-tunable.

As quasimonochromatic generators (Ref. 3), the superconducting junctions emit either recombination phonons ($h\nu = 2\Delta$) at a fixed frequency, or "bremstrahlung" phonons ($h\nu = eV - 2\Delta$), at a frequency depending upon the bias voltage V. This is a very attractive device since tunability can be achieved by voltage modulation. Although the quasimonochromatic power is very limited (∿ 10 μwatts for typically 5 GHz bandwidth), very fine spectroscopy could be achieved by this means. Examples are impurity levels in Al_2O_3, phonon-magnon coupling in MnF_2 (Ref. 4).

4. STIMULATED PHONON EMISSION (Ref. 5)

The three-level electronic system of V^{4+} ions in Al_2O_3 is well known (Fig. 4) : above the $E_{3/2}$ ground-state lies a first $E_{1/2}$ excited state 28.1 cm^{-1} apart, while the second excited state, which also transforms as $E_{1/2}$, is itself 24.7 cm^{-1} above. All three transitions, labelled A_{31}, A_{32} and A_{21}, are possible via photon or phonon emission ; only 3 ⟶ 2, however, is associated with L-polarized phonons along the c-axis of Al_2O_3.

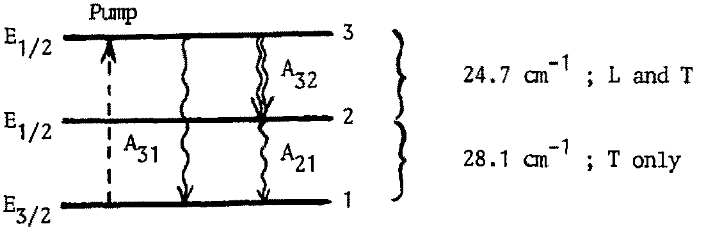


Fig. 4 : Energy diagram for the lowest three levels of the d^1 configuration of V^{4+} in Al_2O_3. Only levels 2 and 3 are coupled via an L phonon pointing along the c-axis.

A transient population inversion was produced by pumping the ground-state level by the 52.6 cm^{-1} radiation of a CH_3F laser. Stimulated emission manifests itself, at high pump powers, by an L-peak in the ballistic phonon signal. At pump intensities below threshold, the L component disappears, indicating that A_{31} is the main decay process.
To date, no volume amplification could be detected.

5. COHERENT FAR I.R. GENERATION

Limitation of coherent H.F. ultrasound stems for two main causes : the requirement of surface flatness which becomes an exceedingly difficult problem below 1000 Å, and the lack of powerful enough sources in the microwave region. On the other hand, far infrared radiation from molecular lasers overlap with the R.F. submillimeter band. The idea of exciting hypersound thanks to the electric field of an I.R. beam originated some years ago (Ref. 6).

A molecular laser beam is focussed onto the surface of a piezoelectric material (quartz), where it is partially reflected and partially transmitted (Fig. 5).

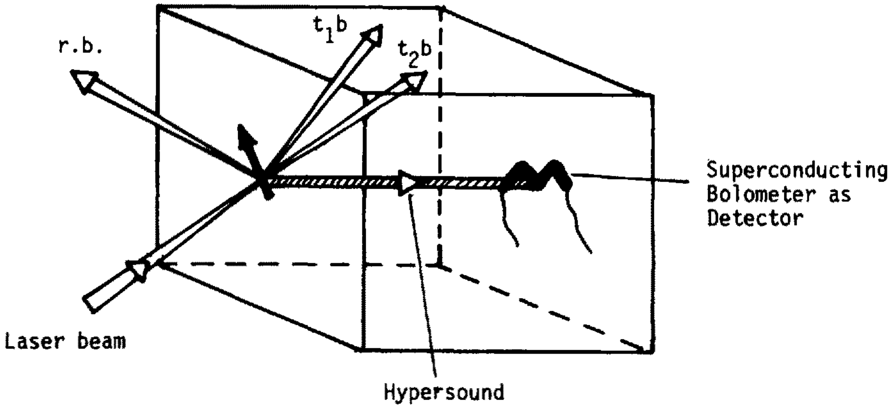


Fig. 5 : The incident I.R. beam gives rise to a reflected beam (r.b.), two transmitted beams (t_1b and t_2b), and a hypersound beam which propagates in quartz at the velocity of lattice waves.

However, the I.R. field is also coupled to mechanical motion of the quartz surface and this results in an additional beam of H.F. ultrasound, at the frequency of the incident photons, a generation process which must be distinguished from bulk absorption.

Unlike the incoherent mechanisms (thermal generation ; relaxation in a superconductor) described earlier, piezoelectric conversion is a coherent process provided the surface smoothness is perfect enough. For conditions of coherent I.R. excitation, this should produce a collimated beam of H.F. phonons. This property was unambiguously checked by moving the laser spot in front of the detector ; it was observed that no spreading at all occurs over several millimeters of propagation.

Assuming continuum mechanics, the rate of power (P) conversion reads :

$$\frac{P\ (\text{acoustical})}{P\ (\text{infrared})} = 4\ \rho\ d^2\ \frac{v_s^3}{\varepsilon_0\ c(1+\sqrt{\varepsilon})^2} \qquad (2)$$

where ρ is the density of mass, v_s the velocity of sound, and d is the appropriate piezoelectric modulus. ε_0 and ε are respectively the dielectric permittivity of vacuum and the relative permittivity of the medium. For quartz, the ratio is as low as 10^{-6} or less. In spite of this, coherent H.F. ultrasound could be produced at 0.89 THz with an HCN laser, and more recently at 2.53 and 3.4 THz.

This fascinating new technique, which boosts the domain of coherent ultrasonics by a factor of 100 (no coherent detection has been reported though), raises also some new problems such as the unexpectedly long propagation length (several millimeters) of THz waves.

6. IMAGING ?

Due to their limited mean free path, their sensitivity to imperfections and poor transmission characteristics at the interfaces, one can question the applicability of H.F. phonons in imaging. In fact, they constitute an unexcelled probe in some instances :

- image of the Fermi surface : in a collision between an acoustical phonon and a free electron, the maximum momentum imparted to the electron is $2k_F$, where k_F is the Fermi radius. This interference phenomenon between the lattice wave and the de Broglie wave of the electron is related to the famous Kohn anomaly. Spectroscopy by H.F. incoherent phonons using quantum detectors led to a value of the Fermi wavelength. $\lambda_F \simeq 280$ Å for a semiconductor containing about 4×10^{17} electrons/cm^3 (Ref. 7).
- Measurement of Bohr orbits in solids : relatively large orbits are associated with donor impurities in most semiconductors, due to the reduced effective mass and large dielectric constant. If a phonon mode is coupled to the impurity levels, the coupling constant is modulated by the structure factor $(1 + a^2q^2)^{-2}$, where a is the Bohr radius, which will cause a very rapid decrease of the phonon absorption at $q \gtrsim a^{-1}$. Absorption spectroscopy thanks to chromatic dispersion (see § 2) led to $a = (39 \pm 1)$ Å for the Bohr radius of the Sb donor in Germanium (Ref. 8).
- Thin-film resonances : although measurement of evaporated thin films is possible, it cannot compete with other standard methods. On the contrary, H.F. phonon interferences are well adapted to the thicknesses of superfluid helium films (∿ 100 Å or less). Helium film thicknesses were thus measured with good resolution thanks to the spin-phonon spectrometer (Ref. 9) made of magnetically tunable T_m^{++} ions in SrF_2.

In conclusion, spectroscopy with H.F. phonons, rather than imaging, has been exploited as yet. The latter area is likely to develop, especially if it becomes possible to handle conveniently coherent phonon beams.

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