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PHONON PHYSICS: ACOUSTICS AT TERAHERTZ-FREQUENCIES

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Introduction

Mechanical or acoustical vibrations and waves in liquids and solids are quantized as are electromagnetic waves and the corresponding energy quanta are known as acoustical phonons. As consequence of the atomic structure of matter there is an upper limiting frequency for the existence and propagation of mechanical waves ranging from 10^{12} to 10^{13} Hertz or up to 10 Terahertz.

Acoustics at terahertz-frequencies consequently is an extention of the ultrasonic range to the highest possible frequencies and thus is an area for investigating wave propagation, absorption and scattering in the volume or at interfaces. Scattering and absorption are caused by resonant transitions of impurity atoms or lattice defects as for example dislocations. Investigations of this kind lie in the field of acoustic absorption spectroscopy where the resolution can exceed the sensitivity of far infrared spectroscopy in the same frequency range by several orders of magnitude.

It is well known that high frequency mechanical vibrations are generated in most energy transfer processes for example in solids as heat. Transistors, light emitting diodes or superconducting tunneling junctions are examples for energy losses besides the useful functions as switching or light emission. These energy losses in general first cause the creation of nonthermal acoustic phonons. Succeeding phonon-phonon and phonon-electron interactions lead to thermalization.

If these interactions are supressed, it is possible to directly investigate the primary emitted phonons. This supression is possible if the primary phonons are generated in thin films which are prepared on crystal substrates with weak secondary phonon interactions. The frequency analysis of these non-equilibrium phonons is equivalent to an emission spectroscopy with acoustical phonons and allows to study the most important energy loss sources causing the phonon emission. Of great importance are the primary electron-phonon processes which serve to identify the essential transition processes.

In these investigations it is important that the phonon propagation is not disturbed by trivial influences. So mostly very pure and dislocation free crystals are required as experimental techniques in the range of low temperatures of roughly 1 K in order to reduce the thermal phonon background. Propagation experiments are performed as pulse measurements where the determination of acoustical travelling times allows for the phonon mode identification.

Phonon detection

In contrast to the ultrasonic range it is not possible to detect terahertz phonons coherently by using the piezoelectric effect because of the short wave length of roughly 100 Å. For wide band phonon detection superconducting bolometers /1/ instead are useful. Tunable and frequency selective phono-detection is possible by phonon transitions in optical systems and the concomitant change of optical fluorescence /2/. Phonon detection with a low energy threshold is possible with superconductor thin film tunneling junctions /3/ where the superconducting metal films can be deposited by high vacuum evaporation on the surface of arbitrary polished crystal surfaces. Phonons with the minimum energy of the superconducting energy gap (Al = 80 GHz, Sn = 280 GHz, Pb = 680 GHz) are absorbed in the metal film by cooper pair breaking. In this process electrons or quasiparticles are excited and they can penetrate the oxid barrier by tunneling inducing a signal in the external circuit. With tunneling junctions using superconductors with different energy gap, it is possible to obtain additional frequency information of the phonon spectrum above the lower threshold energy /4/. It is also possible to analyse the emitted phonon spectrum by tunable filtering, using pressure dependent atomic resonance absorption as for example in the system Ge:Sb or Si:B.

Phonon generation

The most simple experimental technique for phonon generation in pulse experiments is the use of evaporated heater metal films /1/. The radiated phonon spectrum corresponds to the phonon equilibrium distribution in the heater film. The phonons are radiated into the substrate crystal at bath temperature. Now the radiated phonons correspond to a non-equilibrium distribution since the heater temperature is higher than the bath temperature. By variation of the heater power or correspondingly the heater temperature, the average phonon frequency can be tuned over a large range /5/.

Narrow band or quasi-monochromatic phonon spectroscopy /3,4/ is possible by the use of superconducting tunneling junctions as phonon generators. Electrons tunneling at voltages above the energy gap threshold receive a voltage dependent kinetic energy. By interaction with the lattice of the metal films in which the electrons penetrate this energy will be transformed to phonons with the maximum energy determined by the battery voltage corresponding to

$$h\nu = eU - 2\Delta$$

In this equation h is the Planck's constant, ν the phonon frequency, e the elementary charge, U_{Batt} the battery voltage, 2Δ the superconductor energy gap. The modulation /3/ of the battery voltage corresponds to the modulation of the detector signal at the (frequency ν) energy $h\nu$ with the band width of the battery voltage modulation amplitude. If the energy gap of the generator is small in comparison to the maximal phonon energy, it is possible to use the second harmonic of the detector modulation signal for an improved spectral analysis /3/. The analysis of the primary phonon emission spectrum of superconducting tunneling junctions and the qualitative agreement with all theoretically expected processes and transitions (compare /3/ and /4/) is an example how by controlled (in this case thin film technique) supression of further interaction processes (phonon-phonon, phonon-electron) thermal equilibrium can be prohibited.

Absorption spectroscopy

With respect to this application there are already known a large number of examples (compare /3/, /4/). As especially interesting appears the phonon absorption of interstitial oxygen in silicon (compare fig.1). In this experiment tunable phonons were generated with an Al-I-Al tunnel junction and measured with a Sn-I-Sn tunneling detector. The concentration of 10^{18} cm^{-3} oxygen atoms leads to a strong absorption at 878 GHz. This resonance corresponds to a very typical quantum mechanical bending vibration and rotational transition of the ^{16}O atom between two neighbouring silicon atoms. A second weaker but much sharper line at 820 GHz corresponds to the transition of the ^{18}O isotope which has a much lower natural concentration of 10^{14} cm^{-3}. The true line width of the ^{16}O vibration is as small as the ^{18}O isotope vibration as can be observed at lower concentrations in the range of 10^{14} cm^{-3}. As indicated in fig.1 many weak absorption lines are observed in the frequency range from 900 GHz to 1,25 THz. These lines show clearly up in the second harmonic modulation technique. They are caused by neighbour-neighbour-oxygen interactions which give rise to many different detuning coupling strengths which depend on the different

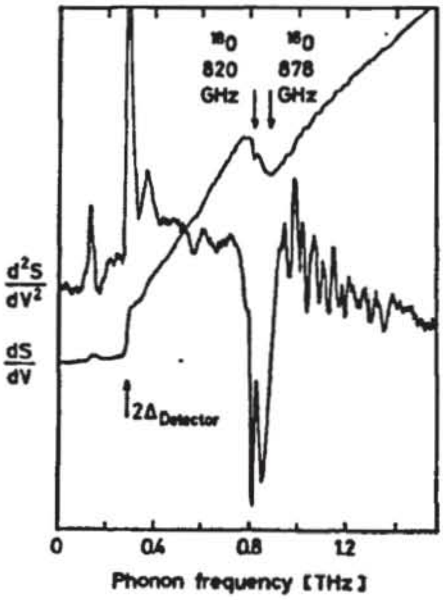

Fig.1 Phonon resonance absorption of interstitial ^{16}O and ^{18}O in Si. Satellite lines between 0.9 and 1.3 THz by ^{16}O-pair interaction are exhibited in the second derivative modulation signal. Ref /4/

neighbour position sites and distances. The concentration of these neighbour pairs is of the order of 10^{13} cm^{-3} /4/ and demonstrates the high sensitivity of phonon spectroscopy exceeding comparable far infrared measurements by several orders of magnitude.

Absorption spectroscopy with acoustical phonons meanwhile has been successfully applied to a many fold of atomic or molecular resonant systems as indicated in references /3/, /4/, /6/, /7/, /8/.

In addition to quasi-monochromatic phonon spectroscopy it has been shown that also high resolution monochromatic phonon spectroscopy /9/, /8/ with the help of the A.C. Josephson effect is possible. In all these examples acoustics has entered atomic processes in solid state physics.

Phonon propagation and quantitative phonon spectroscopy

Besides qualitative phonon spectroscopy it is much more difficult to realize quantitative phonon spectroscopy with respect to all phonon modes. So far only by superconducting tunneling junctions as phonon generator and detector absolute measurements /3/, /4/ have been tried. The comparison with simple theoretical models /3/, /4/ indicates that the phonon intensity at the detector is roughly one order of magnitude below the calculated values. Detailed investigations /10/ led to the result that the cause of losses is localized at the interface between the generator and the substrate and the detector and the substrate. Taking account of these influences, meanwhile more quantitative data on the frequency dependence of phonon absorption and scattering in the volume of solid crystals are possible. Since the phonon scattering losses at interfaces are especially important it is very interesting to investigate their properties by phonon reflection and backscattering measurements.

Phonon backscattering measurements and phonon focussing

Phonon backscattering at free Si-surfaces results in distinct and sharp echo signals /12/ as indicated in fig.2. The phonon pulses have been generated by a heater film and detected with a Sn-I-Sn tunnel junction. The sharpness of the signals generally indicates mirror reflection, but the fourth pulse can only be attributed to the travelling pathes of diffuse scattering. Further experiments with metal covering of the reflecting surface resulted in 50% reflection instead of the calculated reflection factor of 1%. This reflection factor of 50% is roughly equal to the reflection with a coverage by nitrogen or helium, despite the fact that in this case the theoretical reflection is close to 100%. Monte Carlo calculations which have been performed in taking account of the influence of the anisotropy of the propagation of ballistic phonons /12/ or of phonon focussing /11/, /7/ at the uncovered surface, demonstrated that even in the case of diffuse scattering sharp phonon backscattering pulses are obtained /12/. Phonon focussing can be easily demonstrated by experiment /7/, /11/ in accordance with calculations /13/.

In contrast to the isotropic heat conduction in cubic crystals as silicon and germanium the propagation of ballistic phonons is determined by the group velocity alone. In the case of strong anisotropy of the phonon velocity and roughly equal occupation of different K-states in a finite

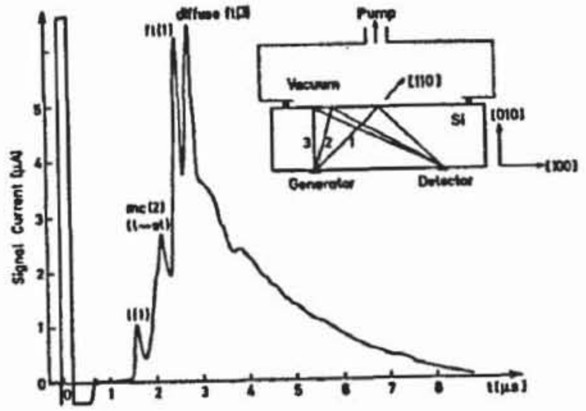

Fig.2 Phonon backscattering signal of a (010) Si surface. Signal along path No. 3 corresponds to diffuse scattering. Ref /12/

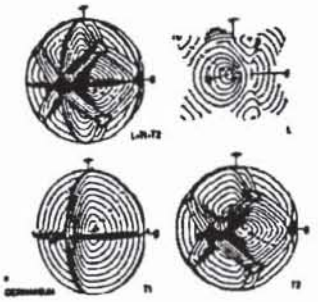

Fig.3 Phonon intensity directivity pattern in Ge for a pointlike heater. l = long.-; T_1, T_2 = fast and slow transv. modes. Ref /13/

energy range, there is a strong increase of the energy flow in definite directions /13/ (see fig.3). This pronounced anisotropic directivity distribution of energy flow can be demonstrated in a simple experiment by the phonon radiation or fountain pressure in a liquid helium film /7/, /11/. The experimental arrangement is shown in fig.4. In the temperature range of superfluid helium, i.e. below the lambda-point, the upper side of a silicon crystal partly imersed in liquid helium is covered by a thin helium film. Phonons coming from a heater on the lower side of the crystal, according to the strong directivity distribution by phonon focussing lead to a locally increased temperature distribution and corrrespondingly to a distribution of increased radiation pressure. Therefore, the helium film (compare with fig.5) buckles which is directly visible and can be photographed. The observed distributions are in agreement with the calculation. Alternative techniques /7/, /11/, /8/ allow a more accurate quantitative analysis of these directivity distributions. Recently the technique of imaging by the fountain pressure has been successfully used for identifying hot spots in measurements of the Ouantum Hall effect.

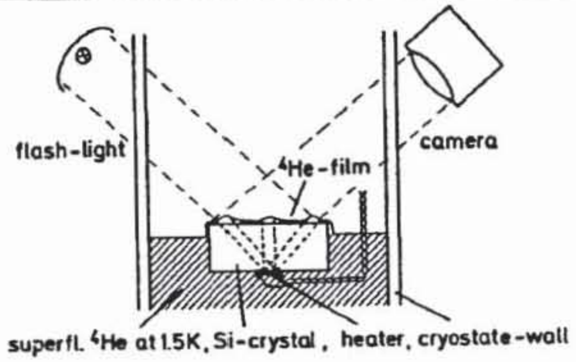

Fig.4 Experimental arrangement for phonon imaging by the fountain pressure of liquid ^{4}He. Ref /7/, /11/

Fig.5 Phonon image at the (111) surface of Si by the fountain pressure. Ref /7/, /11/

Crystal surface treatment and frequency dependence of the phonon backscattering

Since diffuse phonon scattering by focussing can appear as specular reflection, it is important that the parts of specular and diffuse scattering are carefully analysed by Monte Carlo calculations and compared with experiments. Important parameters in this respect are surface treatment and phonon frequency dependence of scattering. Earlier results for diamant polished silicon and phonon frequencies of 280 GHz indicated predominant diffuse phonon scattering /12/ according to this comparison method. Surfaces of corundum Al_2O_3 (100) showed speculär reflections for 80 GHz phonons for a polishing treatment with cubic Al_2O_3 /14/. Instead polishing with colloidal SiO_2 (Syton) resulted in dominant diffuse scattering. At phonon frequencies of 280 GHz the corundum surface polished by diamond revealed a ratio of 4 to 3 between specular and diffuse scattered phonons /14/.

Surprisingly also silicon surfaces (compare fig.6) which are polished with colloidal SiO_2 (Syton) at 80 GHz frequencies show only specular reflection /15/. Coverage with liquid Helium according to our expectation indicates no reduction of the reflection intensity /15/. Also for diamond polished silicon surfaces and 80 GHz measuring frequency we see no significant difference (compare with fig.7) beween phonon scattering at uncovered surfaces and surfaces in contact with ^{4}He, despite the fact that in this case-diffuse scattering is significantly increased. In contrast to this silicon surfaces polished with SiO_2 (Syton) show at frequencies of 280 GHz additional strong diffuse scattering contributions which disappear at Helium coverage.

This result is consistant with the model /12/ that diffuse surface scattering can be

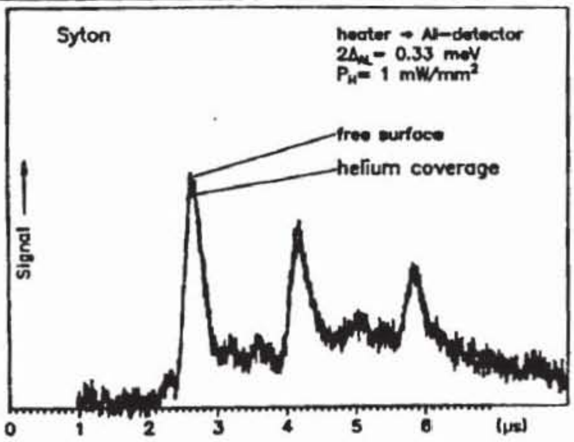

Fig.6 Phonon backscattering from a syton polished Si surface at frequencies of 80 GHz with and without liquid ^4He coverage. Ref /15/

caused as well by geometrical roughness (no coverage influence by ^4He) as well as by atomic or molecular surface states or resonance accompanied by a strong influence of coverage by Helium. The additionally present strong frequency dependence of phonon backscattering has been also observed in phonon transmission experiments /16/ into liquid ^4He with superconductor tunneling junctions. It has been observed that above the frequency of 80 GHz there is an increase of phonon transmission which can not be explained with simple acoustic models. This phenomenon is known as the Kapitza anomaly and presumably has similar causes as the anomalous high phonon backscattering rate of 50% at a crystal-metal interface, whereas the acoustical data predict almoust complete matching.

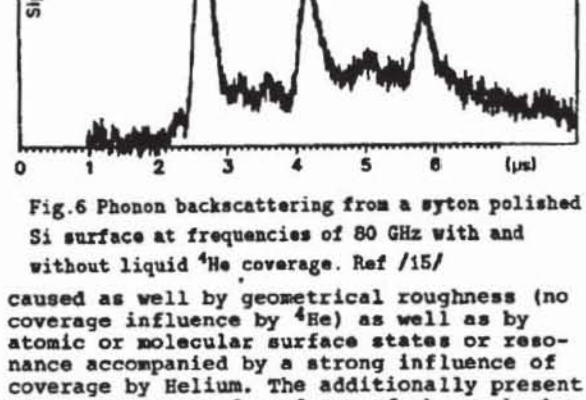

Fig.7 Phonon backscattering from a diamond polished Si surface at 80 GHz with and without liquid ^4He coverage. Ref /15/

Since the acoustical properties of real interfaces appear relatively involved but also physically interesting, there exists the question how acoustically ideal interfaces can be prepared also in the terahertz-frequency range. Possibilities in this respect are cleaving under UHV conditions /17/, the preparation of ideal interfaces by laser annealing /8/ or also the growth of layers using molecular beam epitaxy /18/. It is also important to investigate the possibilities of other simple procedures of ideal interface preparation as for example in different polishing techniques /14/, /15/. In recent experiments it has been possible to prepare acoustical ideal interfaces of silicon by chemical polishing and laser annealing up to a frequency of 280 GHz.

Phonon transmission in thin solid layers and superlattices

The phonon propagation and scattering in thin solid layers can be investigated by depositing films on crystal substrates and evaporating tunneling generators and detectors on top of the system. In this case it is again very important that the interfaces between the different layers are acoustically ideal. The crystal substrate serves for support of the film and of the generator and detector and, is necessary to distinguish between longitudinal and transverse phonons. In this way the phonon scattering of amorphous layers of Si:O, Ge:O, Si, Ge, Si:H_2 have been investigated. The layer thickness ranges from 100 Å to 1000 Å. Corresponding measurements of the frequency dependence of phonon scattering are important with respect to the phonon transport in glasses, the physical nature of the two-level-systems, and the nature of the so-called thermal conductivity plateau in the temperature range between 5 and 10 K. Recent measurements have indicated weak inelastic scattering in Si:O, Ge:O, Si:H beginning in the range of 300 to 500 GHz. Weak elastic phonon scattering has been observed in Si and Ge at higher frequencies /19/. In these experiments phonon scattering at the interfaces turned out to be negligeable. This is important for the preparation of superlattices of different amorphous systems as for example Ge/Si or also Si/SiO. Superlattices of these amorphous systems in the thickness range of roughly 50 Å show strong phonon interferences and phonon stop-bands originally demonstrated by phonon reflection in multiple crystalline and epitaxial heterostructures /18/. The results of phonon transmission measurements for a superlattice multilayer structure of amorphous Si/SiO with 40 Å thick layers in a stack of 7 show a sharp stop-band as indicated in fig.8 /20/. The

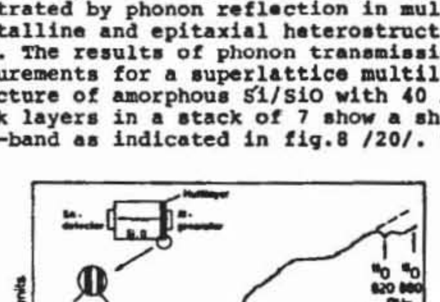

Fig.8 Phonon transmission spectrum of an amorphous Si/SiO_2 superlattice. Ref /20/

sharp stop-band structure at 400 GHz indicates that the interface properties of the layers are ideal and also the individual thickness of the multilayers is accurate within less than 1 Å. By calculation a second stop-band around 800 GHz is expected. The fact that this cannot be observed, indicates that the interference properties of the layer are disturbed at these high frequencies by elastic scattering within the film volume or at the interfaces. On the other hand, the observation of the Si:O resonance indicates that the phonons are not lost by inelastic scattering.

Phonoconductivity

It is the natural aim of phonon physics at terahertz frequencies to extend the possible range of phonon spectroscopic investigations to the upper limit of 10 THz. There are some approaches in this direction. Especially semiconductor physics and microelectronics see the importance of phonon aspects. With respect to the transitions of photons in high energetic phonons in semiconductors as Ga/As very interesting results have been obtained /21/.

In these investigations yet it is necessary to distinguish between high energetic nonequilibrium phonons and thermal phonons which are generated in parallel. In this respect new detectors with high energy thresholds are especially important. Superconducting tunneling detectors with essentially higher values of the energy gap as Nb so far are not known. Instead it has been found that doped semiconductors as Si:B (P, As, In) show in analogy to photoconductivity also a very sensitive phonoconductivity /22/, /23/. For this purpose it is possible to use the phonon excitation of charge carriers which are bound to D^--or A^+-centers. The experimental arrangement is schematically shown in fig.9. Fig.10 shows the signal increase for the system Si:In when the phonons which are radiated from the aluminum junction as generator exceed the binding energy of the system corresponding to 6,2 meV or 1,5 THz. This result gives evidence for a new and very sensitive phonon detector and also shows that quasi-monochromatic nonequilibrium phonons in thin aluminum films can be generated up to these frequences. In priciple phonons can be generated in aluminum films up to 10 THz (longitudinal) and 5 THz (transversal). Meanwhile the direct phonoionisation of neutral donors in the system Ge:Sb at 10 meV corresponding to 2,5 THz and in the system Ge:As corresponding to 3 THz have been observed. This is shown in fig.11 with a natural donor concentration of 10^{13} cm^{-3} /24/.

Future questions concern a further detailed quantitative discription of phonon emission, phonon absorption and scattering and phonon detection for all different modes and the corresponding experimental control. With respect to questions of technology in microelectronics and energy losses in electronic devices, phonon physics increasingly will be important. More fundamental research in phonon physics may concern the extension of ultrasonic inspection to the small objects as lattice vacancies, dislocations, voids or in general radiation defects characterized by phonon resonant absorption or back-scattering at very high frequencies. In fundamental research the different field of the indirect high resolution detection of nuclear particles by their phonon emission when scattered in condensed matter is another very active new field of phonon physics /25/. Phonon detection by superconducting tunneling junctions of aluminum in combination with phonon focussing in silicon crystals appears as very promising also with respect to the solution of the solar neutrino problem.

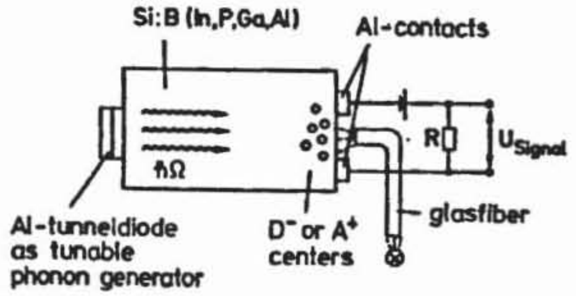

Fig.9 Experimental arrangement for phonoconductivity by phonoionisation of D^- or A^+ centers or D^0 centers. Ref /22/, /23/

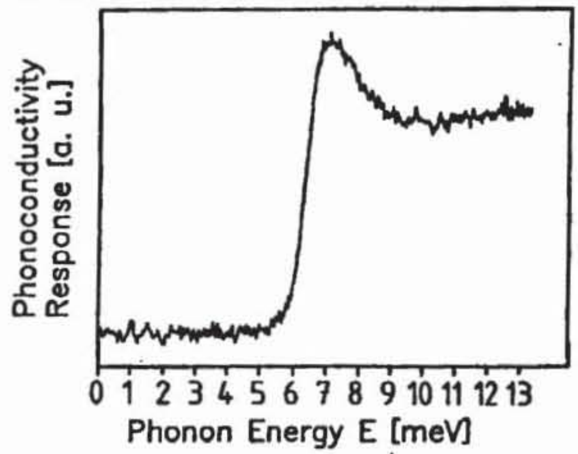

Fig.10 Phonoconductivity signal of A^+ centers in Si:In at 6.2 meV corresponding to 1.54 THz. Ref /22/, /23/

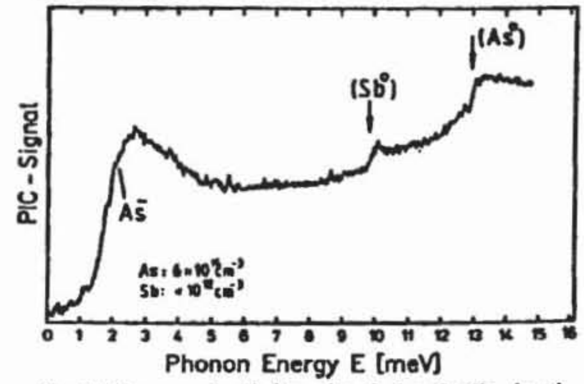

Fig.11 Phonoconductivity signal for Ge:Sb, As at 10 meV and 13 meV corresponding to 2.5 THz and 3.2 THz. Ref /24/

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