Ballistic phonon propagation in BCS superconductors

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We present a survey report on ballistic phonon propagation within bulk, BCS superconductors. Included is a description of phonon focussing and the various phonon scattering processes such as anharmonic phonon - phonon interaction, quasiparticle interactions, and defect scattering. Experimental methods of ballistic phonon creation and detection are summarized. The results of ballistic phonon imaging experiments in superconducting Pb and Nb are described in detail, supporting the use of ballistic phonons as a probe of superconducting properties

INTRODUCTION

Phonons are the fundamental quanta of lattice vibration in a solid. They play a critical role in phenomena such as superconductivity and some phase transitions, and are the basis for the acoustic, thermal, elastic, and infrared properties of solids[1]. Phonon propagation is on the surface a trivial problem. As phonons are thought of as eigenstates of the harmonic hamiltonian, *a priori* they have infinite lifetime and a mean free path limited by the crystal boundary. In actuality anharmonic terms in the ionic hamiltonian introduce phonon-phonon interactions which limit their lifetime. In addition, scattering from defects, boundaries, and valence electrons reduce the mean free path to microscopic distances[2], recovering the fact that heat flow under ordinary conditions is a diffusive process. A phonon is said to be ballistic if all interactions that limit its lifetime are small.

Ballistic phonons represent the true sound waves within a crystal. By increasing the mean free path of single phonons to observable distances, they can be treated as a signal to detect the lowest order of scattering processes that are still present in the ballistic environment. The elastic scattering rate is a measure of frequency and distribution of defects such as impurities, ionic substitutions, or crystal boundaries within the lattice[3]. The inelastic scattering is a measure of interactions between phonons and all other quasiparticles - including anharmonic phonon-phonon interactions and scattering from excited electronic states. Ballistic phonons within a superlattice can therefore be used as a probe of superconductivity such as quasiparticle states as well as phonon anomalies and phase transitions.

THEORY

Although ballistic propagation of a lattice wave is always linear between the source and detector, the associated energy flux is inherently anisotropic. For example we see sound velocities in a crystal that are direction dependent. This is caused by an angularly dependent dispersion (frequency - crystal momentum) relation that deviates from

$$w\left(\vec{k}\right) = c\left|\vec{k}\right|$$

to something more complicated (see fig. 1). This is manifested in the group velocity for a given phonon being usually non-colinear with the

$$\vec{V}_g = \nabla w\left(\vec{k}\right)$$

wave vector.

In fact, following fig. 1 the group velocity deviates from being radially outwards from a spherical constant ω surface, to be generally collected along a few directions of zero curvature. This is called phonon focussing. The energy flux of a wave packet is colinear with the group velocity and so focussed into channels of zero curvature (caustics)[2]. Keep in mind there is no actual change of direction of propagation of the wave vector within the crystal, only the energy flux is affected. Phonon focussing is only seen with an incoherent phonon source - such as a heat pulse - which creates a spherically symmetric

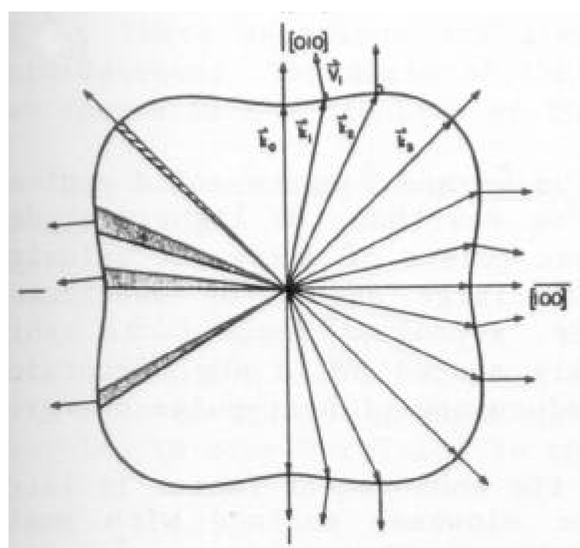

FIG.1 Section of the constant w surface for the transverse mode in Ge, plotted in the (100) plane of wave vector space. The energy flux is along the surface normal (from Northrop and Wolfe[2])

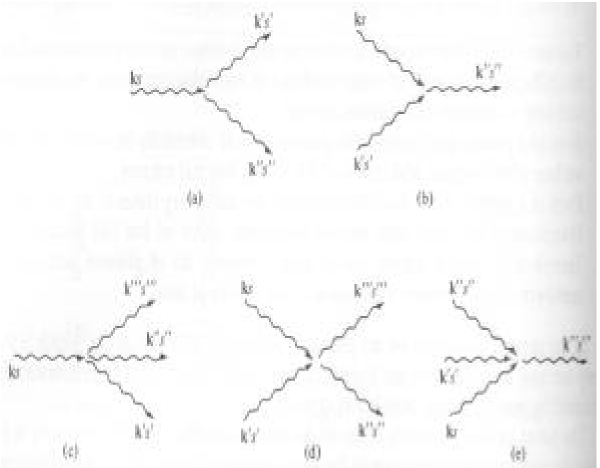

FIG. 2 Possible anharmonic phonon scattering events. a) and b) are from cubic terms, c)-e) are quartic. (from AM[4])

population of wavevectors. Coherent sources which produce a single frequency see this effect only as a retardation compared to other directions.

Phonons, as created by a harmonic lattice, have infinite lifetimes within that assumption. However anharmonic effects are always present in the crystal hamiltonian, to preserve such features as crystal expansion and contraction as well as heat transport properties. Viewing these terms as second-quantized operators, they give us the necessary creation and annihilation operators to express phonon - phonon scattering processes. These collisions have a variety of channels, as phonon number is not conserved, and the crystal momentum (wave vector) is only conserved up to a reciprocal lattice vector[4]. Fig 2 shows typical processes including cubic, and quartic scattering.

Calculating the characteristic rate of the phonon-phonon processes is non-trivial. Here we may use the low -temperature form of many equations, but must avoid the usual assumption $\hbar\omega/k_B T << 1$ as untenable for high energy phonons at low temperatures. Srivastava[1] deduced the following results for the characteristic time between collisions from both the projection operator method and time dependent perturbation theory.

$$t^{-1}(s+s' \to s'') = c_2 w^2 T^3$$

$$t^{-1}(s \to s' + s'') = c_1 w^5$$

Here s, s' s" indicate the relevant three phonon processes, the parameters c1 and c2 are unique to each material, but can be fit to steady state thermal conductivity results at low temperatures. Four phonon processes are at least two to three orders of magnitude weaker than three phonon processes.

A related process in a superconducting lattice is phonon - quasiparticle interactions. In the two BCS superconductors that will be discussed, we have an excellent view of this interaction at low temperatures[5]. In Fig 3, a typical incident phonon energy spectrum is plotted on the left hand side versus the y ordinate, and the quasiparticle density of states is on the right side. The gap energy Δ represents the energy cost per electron to excite a cooper pair from the bose ground state. We see there is no phonon scattering until $\hbar\omega > 2\Delta$, and then the cross section is finite and large as cooper pairs are broken, forming excited quasiparticles. A quasiparticle then may

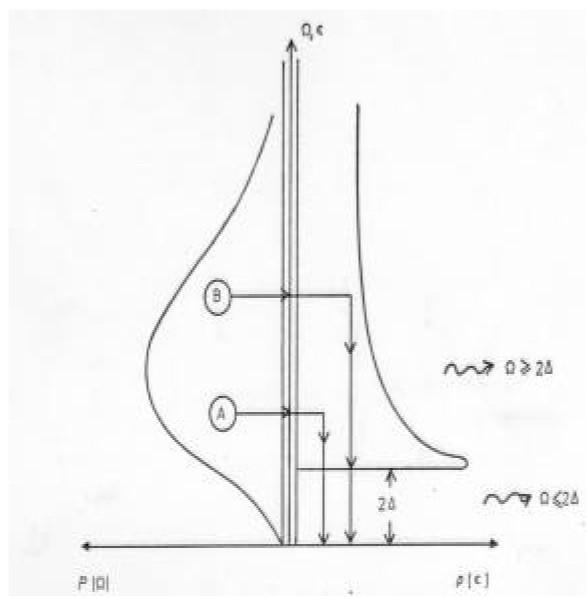

FIG. 3 On the left is a planckian distribution of phonons from an incoherent heat pulse. The right hand side is the quasiparticle density of states at the same scale. The wavy lines refer to incident and not re-emitted phonons. (from Bron[5])

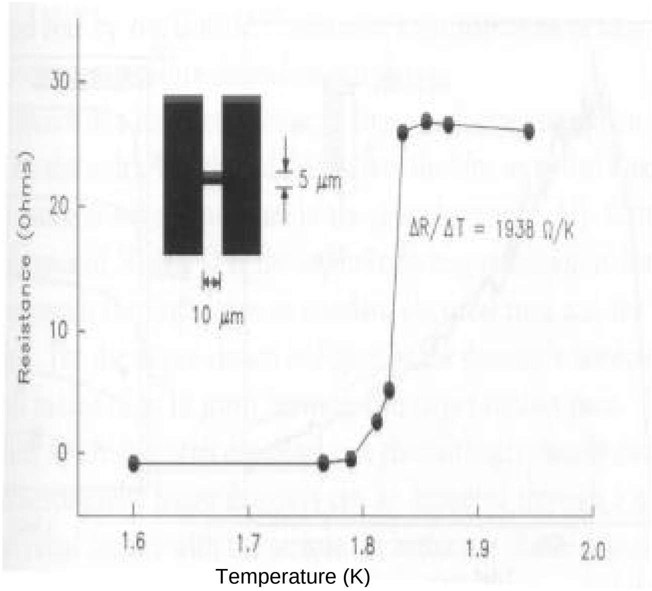

FIG. 4 Response of a granular Al bolometer of dimensions 50 µm^2. (from Wolfe, orig Msall[7])

decay to the gap edge, re-emitting a phonon of $\hbar\omega' = \hbar\omega - 2\Delta$. From the top of the gap two quasiparticles recombine at a rate τ_R^{-1} to form a cooper pair, emitting a phonon of energy exactly 2Δ. The rate of simple phonon - quasiparticle scattering is small compared to the creation / decay process at T<<Tc[5]. Quasiparticle number is conserved in a phonon scattering event, however the more common recombination and creation processes of course do not conserve this number. For a higher order example, two quasiparticles combine to form a 2Δ phonon, which then decays into two lower energy phonons that may travel ballistically across the crystal. At T ~ Tc ballistic behavior disappears and these interactions can successfully be modeled by describing the phonon and quasiparticles as coupled diffusive gasses.[6]

Phonon-defect scattering is more straightforward to quantify. Even in pure crystals there is a non-negligible percentage of ion substitution, i.e. different mass impurities, which act as fixed elastic scattering centers for phonons. Although the theory of scattering rates at point imperfections is quite complicated, Bron[5] considered Rayleigh type scattering from an isotropic distribution of local mass variations (ΔM), and found the following

$$t_{el}^{-1} = \left(\frac{n}{h}\right)\left(\frac{\Delta M}{M}\right)^2 \frac{w^4}{4pu^3}$$

where n is the defect concentration, η is the number of atoms per unit volume, and v is the average group velocity in the acoustic branches, with the overall rate to be proportional to ω^4.

Note that in all these processes, higher frequency phonons are predicted to be scattered much more strongly then lower energy. This will be important later on when creating tunable detectors, and also to support that ballistic phonons are still sensitive to these processes even at low temperatures.

EXPERIMENT

Methods

Production and detection of phonons in crystals requires the deposition of known materials directly on the surface of the subject crystal that easily excite or are excited by lattice vibration. Common incoherent phonon inducers are thin films of metal, whereas coherent sources are exclusively piezo-electric crystals. Under laser or electronic excitation these produce populations of phonons which transfer to the substrate crystal. As in the discussion on phonon focusing, both events stimulate a hemispherical distribution of wave vectors. The thin metallic film has an incoherent planckian distribution of frequencies and the piezo-electric crystal is a coherent source at a single frequency.

Phonon detection is commonly achieved through a strip of superconducting material such as Al cooled to precisely the transition temperature(bolometer). At this point the resistivity of the bolometer is extremely sensitive to small changes in temperature, such as arriving phonon pulses. A bias voltage is applied across the strip, and the current changes are monitered. Thus the thermal stability of the detection surface is critical, but certainly experimentally feasible, eg through a well regulated He bath. In some cases these detectors are granualated by growth in an oxygen rich environment to raise the transition temperature to more accessible levels. A typical design and reponse curve is in figure 4[7].

A more sensitive design of phonon detection is through a superconducting tunnel junction (STJ) deposited on the surface of the material in question. This is a superconductor / insulator / superconductor junction that measures quasiparticle tunneling across the insulating gap. The junction is typically kept at T<<Tc and a bias voltage is applied $V<2\Delta/e$ so that a small, constant current proportional to the equillibrium quasiparticle density in the STJ flows in the absence of incident quasiparticles. A phonon of $\hbar\omega>2\Delta$ creates quasiparticles on both sides of the gap, with a resulting increase in current flow across the junction. If one side of the junction is in electrical contact with the sample, incident quasiparticles from the sample will also increase the current flow.

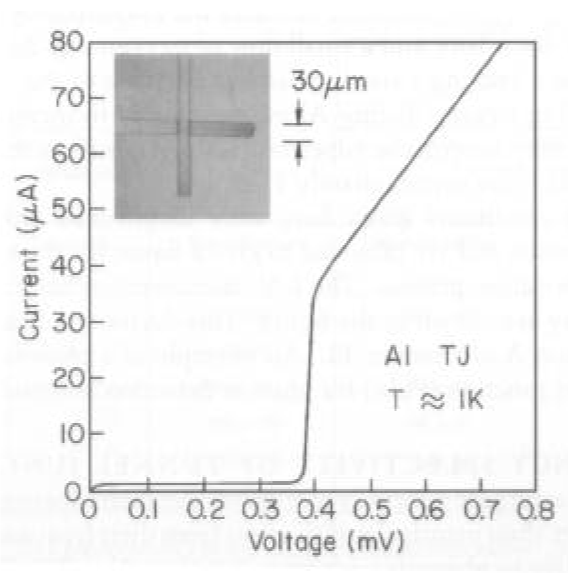

FIG. 5 Response of a superconducting tunnel junction (STJ) of Al - Al O - Al. Note there is a constant nonzero current proportional to the equilibrium quasiparticle density until energies of 2Δ are reached. From Wolfe[7]

An interesting property of these devices acting as phonon detectors arise from the frequency selectivity and sensitivity. Phonons of frequency $\hbar\omega<2\Delta$ are not registered, and past this onset frequency the detector is extremely sensitive. As we saw in the previous section, all lowest order phonon scattering processes have a scattering rate that increases sharply with frequency, so lower frequency ballistic phonons are selected by diffusion in the crystal. With a knowledge of these processes, and an adjustable doping of the STJ members - to select the bottom onset frequency - one can design a small frequency range, monochromating detector employing the crystal as the low frequency pass and the detector as the high frequency pass.

Other interesting detectors, that will not be discussed here, include a large area tunnel junction that is itself spatially scannable by laser induced precursor formation, and supercooled highly sensitive single photon STJ detectors.

The phonon source can be either static (earlier work), or scanned across a surface of deposited film (later work). The scanned heat pulse technique deserves some explanation as it will be heavily referred to later on. The detected ballistic phonon pulse from a heat source is guaranteed to travel in the straight line connecting the source spot and the detector. This selects a certain wavevector k out of the hemispherical population in Fig 1. However, it is the energy flux of the arriving wave which is detected, so if the group velocity is not colinear with this k, there is no signal. If this wavevector is representative of a phonon focussing caustic, there is a high signal. Therefore the picture is a two dimensional cut of the phonon dispersion surface, with an odd inversion - the regions of zero curvature are bright, whereas regions of high curvature are dark. It is perhaps more intuitive to view the pictures as a direct representation of the group velocity population - therefore the lines of preferred energy

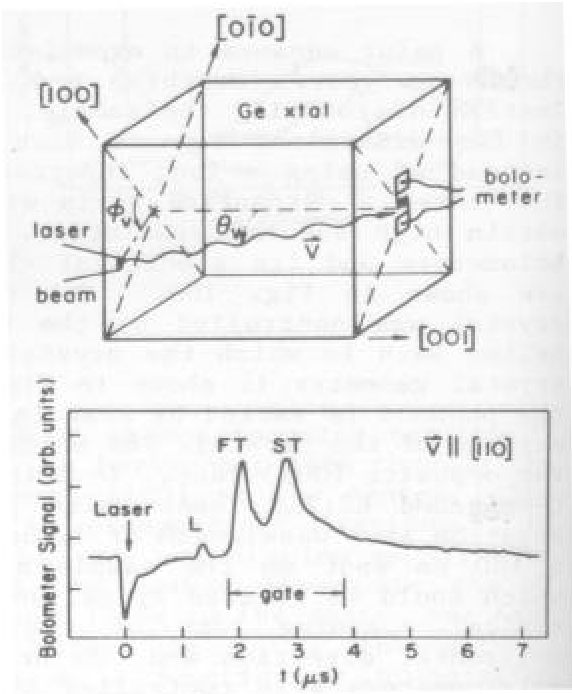

FIG.. 6 Scannable phonon imaging set up. The laser is raster scanned over the surface opposite the bolometer. The propagation direction is given explicitly by Θ_v, Φ_v. Bottom picture is the heat pulse signal, Δt averages over both slow and fast transverse modes. (from Northrop and Wolfe[2])

transfer - and not as a quantification of the $\omega(k)$ surface.

The detection of a scanning heat pulse involves recording the bolometer signal starting after a time triggered by the laser pulse t_0 for a time gate Δt. This peak intensity for each pixel is averaged over several laser pulses. The time t_0 is simply the distance from pixel to detector divided by the average group velocity. The time gate Δt can be set to record a particular mode, or as an average over all modes. Composite time dependent pictures of phonon activity (movies), are obtained by taking several complete crystal scans at varying t_0 with a small time gate Δt[7].

Results and discussion

Naryanamurti *et. al* used a combination of static techniques to probe quasiparticle and phonon properties in bulk, superconducting lead. A tunnel junction of Pb-oxide-Pb was used to detect both quasiparticles and phonons of energy $\hbar\omega > 2\Delta$, and a bolometer of granualized aluminum or $Pb_{1-x}Bi_x$ alloy was used to detect phonons below this energy. The STJ detector was deposited on one face of the lead single crystal, and the bolometer was deposited on the same face, electrically insulated from the bulk by a thin film of SiO. A fixed metallic strip on the opposite face provided the phonon generation by laser heating.

Given the high cross section within the crystal for phonons of energy $> 2\Delta_{lead}$, the tunnel junction almost exclusively detected ambient crystal quasiparticles. The bolometer was electrically insulated by the SiO and so an exclusive phonon detector. This made it possible to decouple a single heat pulse signal to four contributions - ballistic phonons, ballistic quasiparticles, diffusive phonons, diffusive quasiparticles. Figures 7 and 8 show typical detector signals. One can see all four signatures clearly. From the above we can describe the diffusive phonon pulse as an accumulation of phonon - quasiparticle - phonon channels, and phonon specific decays. The diffusive quasiparticle pulse is an accretion of phonon - quasiparticle processes with the lifetime of each modulated by the quasiparticle recombination rate.

The lineshape of both diffusive pulses was fit using coupled diffusive transport equations of interacting gasses. As the main scattering process was recombination, this gave an estimate of the quasiparticle rate τ_R^{-1} as a function of temperature that compared well with previous calculations[8]. Also, the proposed lifetime modulation of both diffusion rates via the recombination channel was justified.

The ballistic profiles were of course beyond the scope of equilibrium diffusion theory, but proved the exclusivity of the detectors. That is, the low temperature ballistic quasiparticles were invisible to the bolometer, and each of the ballistic

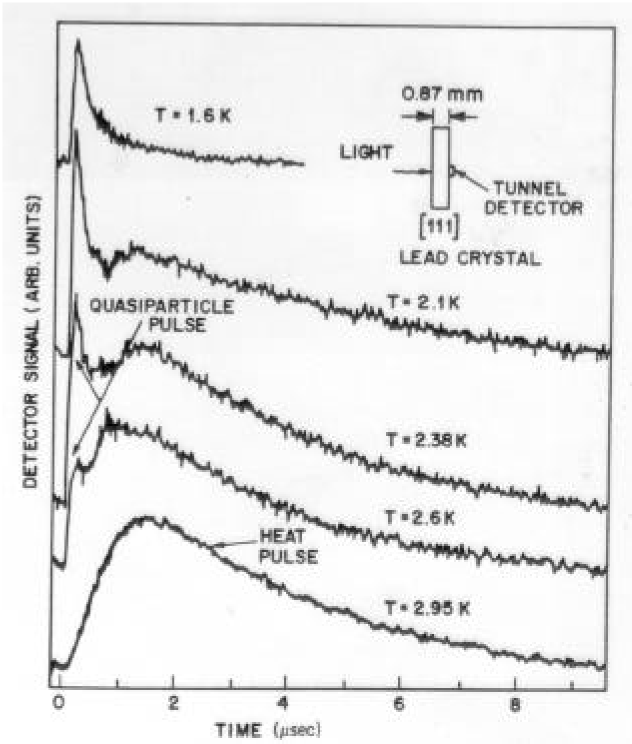

FIG.7 Tunnel junction detector on Pb single crystal. Incident quasiparticles are seen both ballistically, and in a diffusive tail. (data from Narayanamurti[6])

peaks were an order of magnitude slower to arrive at the tunnel junction and so have nothing to do with the quasiparticle lineshape.

This experiment showed how ballistic phonons can be used to probe fundamental properties of quasiparticle states within a superconductor with a relatively simple experimental set up. A more flamboyant experiment by Hauser and Wolfe[9] is the topic of the remainder of this section.

Hauser's work resulted in scanned images of ballistic phonon transport in superconducting Nb. A optically excited Cu film was deposited on the [110] face of single - crystal Nb. Detection was by a granualized Al bolometer film of dimensions 5x10 μm^2 on the opposite face. The Nb crystal was cooled to 1.8K ($T << T_c = 9.5K$), and the Cu film was raster scanned to produce the images in Fig. 9. The lower energy images show

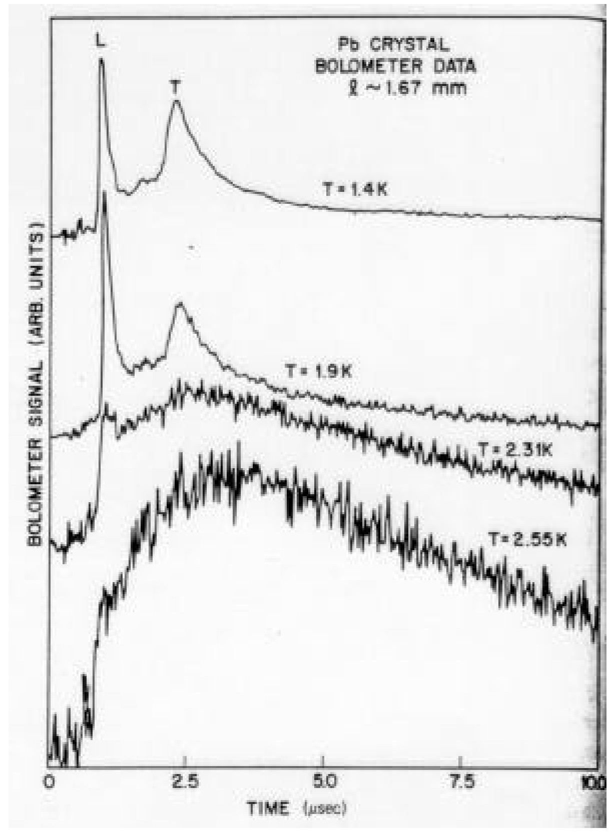

FIG. 8 Bolometer detector on Pb single crystal. Ballistic phonon pulses are clearly observed at low T, switching to a broad diffusive pulse.(data from Narayanamurti[6])

sharp caustics and well defined ballistic signal. At higher energies the lines broaden and saturate, with darker regions superimposed over the original caustic lines. The signal lineshapes as a function of temperature (not shown) follow the pattern of Naryanamurti with the ballistic pulse disappearing, and a diffusive tail becoming more prominent.

The dark regions are understood in terms of quasiparticle scattering. When the total energy of a ballistic pulse steps over the 2Δ threshold, scattering of phonons by quasiparticles changes from near zero to a very high rate. Therefore one sees dark regions appearing along the easiest routes of energy flux first as the laser power is increased. This is borne out by subtracting a very low energy picture, where the only active regions of energy flux are the caustic lines, from a higher

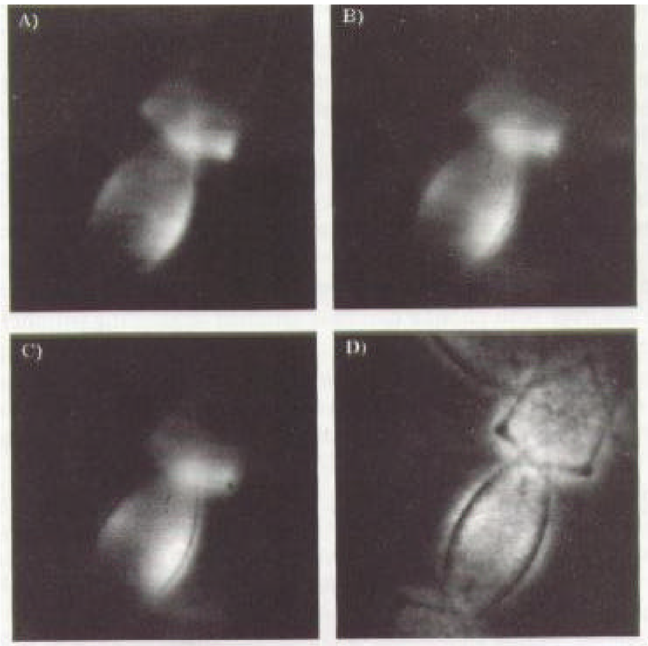

FIG. 9 Phonon images of superconducting Nb at increasing excitation energy: a) 52 nJ, b) 99 nJ, c) 156 nJ. The bright caustics increase in width due to an increase in phonon source size. Dark lines on the image are caused by quasiparticle absorption and delineate low - power caustics. d) theoretical image, subtracting a 50 μm source from a 180 μm source showing the same characteristics. (From Wolfe, orig. Hauser)

energy picture. The subtracted image pinpoints the location of theoretically high quasiparticle scatering, and corresponds well to the dark regions seen.

Also observed was an increased lifetime and spatial extent of the hot spot with increasing laser energy, as evinced by the broadened caustic lines at higher laser energy. The caustics at all three of these energies are broadened with respect to the width of the dark, superimposed lines, representing very low energy. A theoretical picture assuming a hot spot of a large spatial extent, with a smaller phonon source image subtracted, clearly corresponds to the images seen with laser energy variation. Thus the caustic smearing is due to a phonon source of a longer lifetime and larger size.

This lifetime broadening of the phonon source with increasing excitation energy is an interesting question, and worth a detailed look. The basic point is that an incoherent heat pulse generates a broad range of frequencies, as in Fig 10. The population over 2Δ encounters a microscopic mean free path due to quasiparticle scattering, whereas phonons with energy under the gap suffer almost no attenuation. This is not a simple low pass filter due to quasiparticle decay and recombination. That is, following Fig 2, after absorption of the cross hatched population of phonons the excited quasiparticles decay to the top of the gap at a certain rate, emitting one or more phonons of frequency $\hbar\omega<2\Delta$, and then fully recombine to a ground state cooper pair emitting another phonon of $\hbar\omega=2\Delta$. The decay to the top of the gap happens virtually instantaneously[5], and the $\hbar\omega<2\Delta$ phonons then propagate ballistically through the superconductor. This process in thin films is called phonon flourescence. In this thin film limit, i.e. after one round of scattering in our case, the resulting distribution of phonon energies is given by figure 10 (calculated for Pb). As every scattering event eventually produces a phonon of 2Δ, the final frequency distribution is strongly peaked at that energy.

Despite what is implied in Wolfe[7], this process does not by itself increase the lifetime of the hot spot on a large crystal. Even though the recombination time of quasiparticles to the cooper pair ground state is very long, the emitted 2Δ phonons are strongly absorbed and so only slowly leave the spatial location of the hot spot. However this does form a continuous ground state population of quasiparticles proportional to the original population of high energy phonons. By ordinary quantum mechanics, the over - populated quasiparticle ground state has a lasing effect on all thermally excited higher quasiparticle states, proportional to $N_{q\text{-}p\ gap}^2$. It is this author's conjecture that the stimulated decay of a locally excess number of quasiparticles (present perhaps

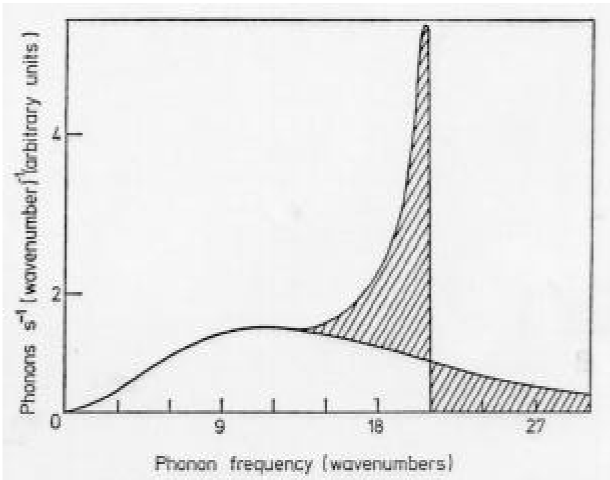

FIG. 10 Phonon generation rate as a function of phonon frequency in a thin film of superconducting Pb. This illustrates the flourescence process - all phonons in the heat pulse with $\nu > 2\Delta/h = 21 cm^{-1}$ (right hatched area) are converted to $<21 cm^{-1}$. From Bron

by phonon heating) is the mechanism for the lifetime broadening of the phonon source.

This model would predict a diffusive expansion of the hotspot proportional to the recombination rate and the original population of $\hbar\omega > 2\Delta$ phonons. With increased excitation intensity, the 2Δ quasiparticle population will be increased substantially, therefore enhancing both the spatial extent of the hot spot, and the lasing effect - thus the flux of secondary phonons -by $(N+N')^2/N^2$. The anomalous increase of both size and lifetime with optical intensity has been seen, although the exact relationship was not specified for comparison.

Returning from this digression, the experiment of Hauser produced a series of images that are indicative of many quasiparticle properties in superconducting Nb, as well as verifying the predicted phonon dispersion and caustics. Although retaining the lineshape for the heat pulse at each pixel is not feasible computationally, one assumes that recording such information in a secondary scan only over the dark caustics - where the quasiparticle signature is high - would provide a more detailed picture of electronic excited states in those regions. It seems clear that accompanying theoretical work must be done for this type of study to be a quantitative probe of the superconducting state, but the phonon focussing pictures have given us the first at-a-glance view of strong quasiparticle-phonon interactions.

In conclusion, we see in the last 10 years progress has been made utilizing ballistic phonons as a probe of superconducting properties and similar gains are expected in the near future. Phonon imaging experiments with high Tc superconductors might well reveal predicted anisotropies in the energy gap. More sensitive detectors, and producers of ballistic phonons are no doubt under development. One could easily see how micro imaging detectors, or scannable producers of coherent phonons would be of enormous benefit to the field. No matter the direction, it seems the use of ballistic phonons will develop into a powerful and versatile tool for the imaging of superconductors and other materials of interest.

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