Abstract. Phonon imaging is particularly suited to measuring the quasiparticle density in superconducting Pb because the absorption of ballistic phonons by quasiparticles is highly anisotropic. The temperature dependence of the phonon absorption over the 1.4 – 2.1 K temperature range yields a superconducting gap in accord with the conventional electronic ground state of Pb.

Phonon physics has played a pivotal role in the idea that crystalline lead (Pb) may have an electronic ground state characterized by a spin density wave (SDW). Overhauser and Damen [1] forwarded this idea in 1988 to explain experiments by van der Hoven and Keesom [2] that showed a low temperature residual electronic specific heat far above the BCS prediction for superconducting Pb. This effect was not present for crystals doped with a few percent Indium. A contemporary low temperature study by Phillips, Lambert and Gardner [3], however, shows no anomalous behavior in the low temperature specific heat for pure Pb. Ultrasonic attenuation [4,5,6,7] has also lead to conflicting experimental results for Pb, likely due to dislocation scattering of elastic waves. Conventional tests such as magnetic resonance and neutron scattering have not uncovered an SDW signature in Pb. Overhauser has discussed support for SDW’s in Pb in Reference [8].

Phonon imaging [9] can provide an independent test of the SDW hypothesis in Pb because it provides a relative measure of the quasiparticle density in the superconducting state. Phonons with energies below the superconducting gap can travel ballistically across millimeter-sized crystals of pure Pb [10], occasionally scattering from thermally excited quasiparticles. By imaging the ballistic phonons, Short and Wolfe [11,12] discovered sharp lines of reduced phonon intensity due to absorption by quasiparticles, as shown in the inset of Fig. 1. The sharpness of the “absorption lines” is due to flatted regions of the Fermi surface with normals along [111] directions, greatly enhancing the absorption for phonon wave vectors in (111) planes of the crystal [13].

Overhauser and Daemen [1] argued that an SDW ground state in Pb would produce a highly anisotropic superconducting gap, $\Delta(k)$, characterized by deep valleys where commensurate SDW wave vectors roughly match $2k_{\text{Fermi}}$. Quasiparticles could be thermally excited easily into the valleys, providing an explanation for an enhanced superconducting electronic specific heat at low temperatures. Early phonon-imaging experiments seemed to support the idea of reduced-gap regions by showing a relatively weak temperature dependence of quasiparticle density over 1.45 – 2.1 K; however, later experiments [13] with different crystal thicknesses found an unexpected dependence of absorption on phonon path length. In this paper, we summarize the results of an in-depth study that takes into account the effects of non-equilibrium quasiparticles, resolving the path-length problem and yielding the conventional superconducting gap of Pb.
Non-equilibrium quasiparticles can enter a phonon-imaging experiment from two sources: the excitation point and the superconducting detector. A Planckian source of phonons is produced by laser-heating a local region of a normal metal film deposited on the crystal, also causing a local heating of the Pb crystal near the point source and a higher quasiparticle density than in the bulk of the crystal. Also, the granular-Al detector film is raised to its transition temperature (higher than the lattice temperature) with a bias current. This second cause of non-equilibrium quasiparticles is unavoidable in the variable temperature experiments essential to our SDW study.

Since we are interested in the equilibrium properties of Pb, we want to minimize the number of non-equilibrium quasiparticles created during our experiments. The phonon-source temperature decreases with the fourth-root of the incident power, so simply lowering the power of the laser pulse over our sensitivity range reduces the source temperature very little. However, a combination of longer pulse length (and detection time), larger source area, and lower power has permitted a usable dynamic range of about 5000 in power density, allowing us to significantly lessen the effect of non-equilibrium quasiparticles.

The lower power density (resulting in lower average phonon frequencies) also means less scattering from mass defects (such as Pb isotopes) in the crystal, allowing a simple analysis of the ballistic-phonon transmission. Figure 1a shows a theoretical line scan of phonon flux through the central (110) plane, showing a) the ballistic phonon flux $I_b(x)$ due to phonon focusing and b) the transmission coefficient $(1 - A(x))$ due to the fractional absorption $A(x)$ by quasiparticles. This two-component fitting function $I(x) = I_b(x)(1 - A(x))$ fits very well to experimental line scans for our data. Fits to data at three lattice temperatures are shown in Fig.1b. A rapid decline in absorption is observed as the temperature is lowered. For each helium-bath temperature $T$, we determine the peak absorption $A_o = A(x_o)$ along the [110] propagation direction.

Figure 1. a) Theoretical curves of ballistic flux $I_b(x)$ and transmission $1 - A(x)$ plotted for the scan line in the (110) plane of Pb, corresponding to the dashed line in the phonon image (inset). $I_b(x)$ is a Lorentzian that matches the theoretical line shape. $A(x) = A_o \exp(- (x - x_o)^2/2 \sigma^2)$ is an empirical Gaussian function. b) Experimental line scans for a 1.26 mm crystal fitted to $I(x) = I_b(x)(1 - A(x))$ with adjustable $A_o$, $\sigma$, and Lorentzian amplitude $I_o$. This experiment uses long pulse lengths of 0.45 \( \mu \)s and the excitation density shown.

The temperature dependence of $A_o$ as shown in Fig. 2a is modeled in terms of phonon scattering from both equilibrium (bulk) and non-equilibrium (near-surface) quasiparticles,

$$I(x_o) = I_o e^{\beta x_o} e^{-\alpha L_x},$$
where the bulk absorption coefficient $\alpha$ is effective over a distance $L$ (taken as the length of the crystal), and the near-surface absorption coefficient $\beta$ is effective over a distance $\ell \ll L$. Using $A_o(T) = 1 - R(x_o)/I_o$ and treating $\beta \ell$ as a single parameter representing the effect of all non-equilibrium quasiparticles, we find the equilibrium coefficient,

$$\alpha = - (1/L)[\beta \ell + \ln(1 - A_o)] ,$$

as a function of helium-bath temperature $T$ and the value of $\beta \ell$ (assumed-constant for a given excitation condition). The BCS form of the absorption coefficient (generally applied to ultrasonic attenuation) is $\alpha(T) = 2\alpha_0 \exp(-\Delta/k_B T)$, where $\alpha_0$ is the absorption coefficient for the non-superconducting state. Figure 2b shows the results for three different power densities, each independently determining $\Delta$ and $\beta \ell$ for a common value of $\alpha_0$. As shown in this semi-log plot, the three data sets together define a temperature dependence proportional to $\exp(-\Delta/k_B T)$ with $\Delta = (1.32 \pm 0.05)$ meV representing the superconducting gap parameter. As expected, the values of $\beta \ell$ increase with increasing power density, which likely involves both source and detector contributions.

![Figure 2](image-url)

**Figure 2.** a) Plots of $A_o(T) = 1 - R(x_o)/I_o$ as a function of $T_c/T$, where $T_c = 7.19$ K is the superconducting transition temperature of Pb. Data for three different peak power densities are shown. b) Equilibrium $\alpha(T)$ for the three data sets vs. $T_c/T$ as described in the text. Both absolute values and temperature dependences match when the contribution from non-equilibrium quasiparticles is taken into account. The dashed line represents an exponential fit to the superconducting electronic specific heat of reference [2] in the temperature range of our phonon imaging experiments.

Our complete study [14] involves both short and long excitation pulses and samples with thickness $L = 1, 1.26, 1.6, 2.5,$ and $4$ mm. The analysis described above consistently yields superconducting gaps close to the low temperature value $\Delta_s = 1.35$ meV measured by electron tunneling. [15]

The dashed line in Fig. 2b shows the temperature dependence of the electronic specific heat from the data reported in Ref. 2. In that data, it appears that the normal state specific heat changes with the addition of indium impurities rather than the superconducting specific heat [3], calling into question the interpretation of a highly anisotropic gap. With phonon-imaging data the effects of non-equilibrium quasiparticles must be taken into account, but the highly anisotropic absorption lines are directly associated with the total number of quasiparticles in the system. The results of our experiments are consistent with the conventional superconducting gap in Pb and are inconsistent with the specific-heat analysis of [2] that has been cited as evidence for an SDW ground state in Pb.
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References
[1] Overhauser A W and Daemen L L 1988 Phys. Rev. Lett. 61 1885
[2] Keesom P H and van der Hoeven B J C 1963 Physics Letters 3 360; van der Hoeven B J C and Keesom P H 1965 Phys. Rev. 137(1A) 103
[3] Phillips N E, Lambert M H and Gardener W R 1964 Rev. Mod. Phys. 36 131. This conference paper includes Discussion 19 by Keesom, which shows an unexplained anomaly in the T-dependence of the normal-state specific heat for the pure crystal [2]. This effect (not observed by Phillips et al.) could account for the residual specific heat that was cited as evidence for an SDW ground state in Pb.
[4] Fate W A, Shaw R W and Saliner G L 1968 Phys. Rev. 172 413
[5] Fate W A 1968 Phys. Rev. 172 402
[6] Tittman B R and Bömmel H E 1966 Phys. Rev. 151 189
[7] Tittman B R and Bömmel H E 1966 Phys. Rev. 151 178
[8] Overhauser A W and Giebultowicz T M 1993 Phys. Rev. B 47 14 338
[9] James P. Wolfe. 1998 Imaging Phonons: Acoustic Wave Propagation in Solids. (Cambridge University Press)
[10] Narayanamurti V, Dynes R C, Hu P, Smith H and Brinkman W F 1978 Phys. Rev. B 18 6041
[11] Short J D and Wolfe J P 2000 Phys. Rev. Lett. 85 5198
[12] Wolfe J P and Short J D 2002 Physica B 316 107
[13] J. D. Short 2001 Using Phonon Imaging to Probe Anisotropy in the Superconducting Energy Gap of Lead.( Ph.D. thesis, University of Illinois Urbana-Champaign)
[14] Head and Wolfe, submitted for publication.
[15] Ashcroft N W and Mermin N D 1976 Solid State Physics (Saunders College Publishing)