[19]D. R. Nelson and R. A. Pelcovits, Phys. Rev. B 16, 2191 (1977).
[20]R. B. Meyer, private communication.
[21]B. D. Josephson, Phys. Lett. 21, 608 (1966).
[22]D. C. Licciardello and D. J. Thouless, Phys. Rev. Lett. 35, 1475 (1975).

Unusual Phonon Heat Transport in an Amorphous Superconducting *Pb*Cu Film

H. v. Löhneysen[a] and F. Steglich
II. Physikalisches Institut der Universität zu Köln, D-5 Köln, West Germany
(Received 11 July 1977)

We report thermal conductivity measurements of a $Pb_{0.9}Cu_{0.1}$ film before (amorphous) and after (crystalline) annealing. Below T_c, where electronic effects are negligible, we found that the phonon contribution in the amorphous phase k_{am}^{ph} was much larger than that in the crystalline phase. We also found that the temperature dependence of k_{am}^{ph} resembles that of amorphous dielectrics, thus providing evidence that this sort of "glassy" behavior does not require the presence of covalent bonds.

Amorphous insulators like glasses, polymers, and a-Se show distinctive thermal properties below 1 K. For these substances contributions to the specific heat proportional to T and T^3 are observed in addition to the Debye term.[1] Furthermore, a T^2 variation of the (phonon) thermal conductivity is found in contrast to the T^3 dependence seen in crystals.[1] This "glassy" behavior, also visible in the k vs T curves of semiconducting a-Ge,[2] can be successfully interpreted in terms of localized low-energy excitations (LEE) as assumed in the Anderson-Halperin-Varma-Phillips tunneling model.[3,4] To date, however, this model lacks a microscopic explanation. Thus, since all the mentioned materials are covalently bonded, it is not clear whether the amorphous structure itself or the presence of covalent bonds in a disordered environment is essential for "glassy" behavior.

Amorphous metals, of course, are the natural probe for such an investigation. Very recently, a phonon thermal conductivity $k^{ph} \sim T^2$ was reported for several bulk metallic glasses.[5] However, all of these materials, most of which contain metalloids with covalent bonds, are in the normal state (including $Ti_{0.5}Be_{0.4}Zr_{0.1}$ which becomes superconducting only below 0.32 K). In addition, they all contain transition metals which can act as resonance scattering centers of the conduction electrons. Therefore, the influence of the conduction electrons (as heat carriers as well as phonon scatters) is not easy to calculate, and as a result conclusions drawn for k^{ph} are, at most, only of qualitative value. To avoid these difficulties, an amorphous superconducting $Pb_{0.9}Cu_{0.1}$ (*Pb*Cu) film, prepared by quench condensation from the vapor phase, was chosen for the present investigation: Well below $T_c = 6.5$ K, the influence of the few remaining electronic quasiparticles on k is negligible. Besides, in *Pb*Cu there are no constituents with covalent bonds.

Another motivation for thermal conductivity measurements on amorphous metal films stems from the fact that in quench-condensed *crystalline* metal films k^{ph} is almost negligible as compared to the electronic contribution k^e.[6,7] It has been argued[6] that in this case the phonons are scattered strongly from spatially extended lattice defects, i.e., stacking faults, dislocations, and grain boundaries. However, since these defects should be absent in a solid without long-range order, we hoped to observe a substantial recovery of the ratio k^{ph}/k^e in amorphous (as compared to crystalline) *Pb*Cu. This should enable us to use k^{ph} as a probe of "glassy" behavior.

The amorphous film was prepared by means of the *Drehrohrofen* technique deveoped by Hilsch and Martienssen[8]: A mechanically homogenized 90-at.% Pb, 10-at.% Cu mixture was evaporated in small pellets (each producing less than one atomic layer) onto a cooled substrate. As substrate we used a 12.5-μm-thick circular Kapton foil, which sealed an additional Cu tank clamped to the cold part of a He^3 cryostat.[9] During evaporation of the metal film, this Cu tank was filled with He^4 exchange gas (5 Torr). Thus the substrate temperature could be kept below 15 K, i.e., well below the crystallization temperature (30 K) of *Pb*Cu. The pressure in the cryostat remained below 10^{-6} Torr during evaporation. Afterwards the He^4 exchange gas was pumped out and the total thermal conductance K_T of the sub-

strate-film sandwich was measured *in situ* in a stationary method with circular heat flow.[9] Before evaporation, the thermal conductance K_F of the Kapton foil had been measured.

Each datum point could be acquired with an accuracy of 1%, the uncertainties arising mainly from the calibration of the carbon thermometers. The conductance of the film, K, was obtained by subtraction: $K = K_T - K_F$. Since K/K_T varied between 0.015 at 0.5 K and 0.35 at 10 K, the maximum experimental error of K varied between 150 % and 6%, respectively. The equipment has been carefully checked by measuring the thermal conductivity of *pure* Pb films, for which—within these errors—the previously published results[6,7] were found. From K, the thermal conductivity, k, of the metal film could be determined with the known geometry factor. The film thickness d = 2800 Å was obtained by multiple-beam interferometry on a reference film evaporated right next to the sample. This reference film also served to observe electrically the transition to superconductivity and to monitor the amorphous-crystalline transition by the accompanying sharp irreversible resistivity drop. For comparison, the thermal conductivity of the crystallized film, annealed at 80 K, was also measured.

The thermal conductivity k of the *Pb*Cu film is shown in Fig. 1 as a function of temperature. For both the amorphous and the crystalline films, the transition to superconductivity (T_c = 6.5 and 7 K as measured electrically and indicated by the arrows in Fig. 1) is visible by a distinct drop of k.

For the *crystalline Pb*Cu film the measured thermal conductivity in the normal state, k_n, obeys Wiedemann's law $k_n^e = L_0 T/\rho_{cr}$, where L_0 is Sommerfeld's value and ρ_{cr} = 25 $\mu\Omega$ cm the residual resistivity determined for the reference film. Also, not too far below T_c, the thermal conductivity k_s is identified with the electronic part k_s^e which can be calculated[7] for "dirty" strong-coupling superconductors by choosing $\eta = 2\Delta_0/k_B T_c = 5.0$ (Δ_0: energy gap at $T = 0$). Deviations from this calculation at the low-temperature end are readily identified with a phonon component k_s^{ph} added to k_s^e (cf. open circles in Fig. 2): Below 2 K, k_s^{ph} is in accordance with a phonon mean free path limited by scattering from grain boundaries with average diameter $\bar{d} \simeq 50$ – 70 Å.[10] The strong depression of k_s^{ph} above 3 K presumably indicates resonant phonon scattering from dislocations[11] with very short loop lengths

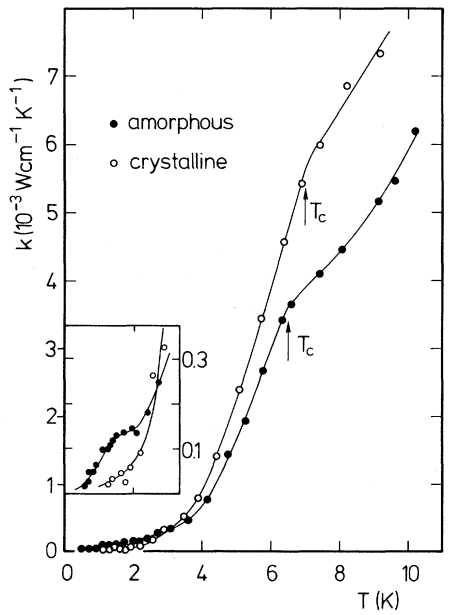

FIG. 1. Thermal conductivity k as a function of temperature for a *Pb*Cu film (d = 2800 Å) in the amorphous and crystalline state. Inset shows low-temperature data on an expanded vertical scale. Lines are intended as a visual guide.

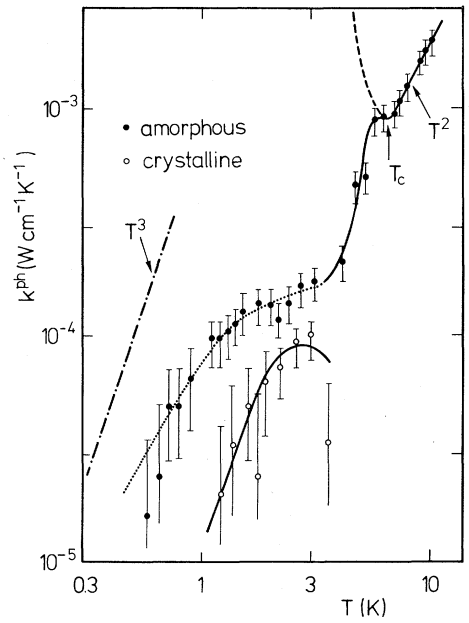

FIG. 2. Phonon thermal conductivity k^{ph} of amorphous and crystalline *Pb*Cu as a function of temperature (bilogarithmically). Solid lines are intended as a visual guide. Dashed line refers to a BCS superconductor [after P. G. Klemens and L. Tewordt, Rev. Mod. Phys. 36, 118 (1964)] and hence is a lower bound of k_s^{ph} as limited by scattering from electronic quasiparticles. The other curves are explained in the text.

$L < \bar{d}$. Above 4 K, *no* phonon contribution k^{ph} can be resolved from the high "electronic background" for the crystalline *Pb*Cu film. This fact, previously discovered for *pure Pb* films,[6,7] is attributed to the large dislocation densities (up to 10^{13} cm^{-2}) in such quenched crystalline films.[12]

For the remainder of this article, we confine the discussion to the *amorphous PbCu* film, which has an enhanced k^{ph} when compared to its crystalline counterpart. In Fig. 1 this is most easily seen for $T \lesssim 2$K, where the amorphous film exhibits higher k values. Also, at 7 K, k_{am} is smaller than k_{cr} only by a factor of 1.5, although this film has a residual resistivity $\rho_{am} = 64\ \mu\Omega$ cm $= 2.7\rho_{cr}$. Above T_c, k_n shows an upward curvature corresponding to a phonon conductivity $k_n^{ph} \sim T^2$ ($0.3k_n$ at T_c) in addition to $k_n^e = L_0 T_0/\rho_{am}$. Apparently, the absence of spatially extended lattice defects in the amorphous phase leads to the hitherto unobserved phenomenon that *the phonon conductivity of a PbCu film decreases upon annealing* (because of the introduction of a large number of grain boundaries and dislocations during crystallization). To obtain $k_s^{ph} = k_s - k_s^e$ in the superconducting state, we calculated k_s^e with $\eta = 5.0$.[13]

The resulting k^{ph} vs T curve for amorphous *Pb*Cu is shown in Fig. 2 (a second film yielded the same magnitude and temperature dependence of k^{ph}). This curve depicts a behavior quite different from k^{ph} of crystalline (bulk) metals: There, as one lowers T below T_c, the "freezing" of electronic scattering centers always causes an increase of k_s^{ph} (cf. dashed line in Fig. 2), which is finally limited by boundary scattering at low temperatures. However, for the amorphous *Pb*Cu film, k_s^{ph} levels off just below T_c and then precipitously decreases around 5 K. Passing a "plateau" at 3 K, k_s^{ph} starts again to decrease more rapidly below 1.5 K. This observation cannot be explained by boundary scattering: The dashed-dotted line in Fig. 2 represents a *lower limit* for k_s^{ph} (if it were determined by *diffuse* boundary scattering[14]), since phonon scattering from the (smooth) surfaces of an amorphous metal should be predominantly specular.

Since the experimental k_s^{ph} values fall well below this line in the whole range of available temperatures, *another* scattering mechanism must be taken into account. Below 4 K our results resemble the k vs T curves established for amorphous dielectrics[1] and a-Ge.[2] This distinct temperature dependence cannot be attributed to normal-state inclusions in the superconducting film, since precipitation of Cu in the Pb matrix is prevented by the choice of the *Drehrohrofen* technique.[8] Therefore, some type of LEE from which phonons are scattered is apparently present also in the amorphous metal. This conclusion seems to contrast with data of the lattice specific heat on amorphous superconducting films,[15,16] from which an enhancement of the cubic term, $\Delta C = C_{am} - C_{cr}$, but no linear term could be resolved.

However, we want to show that our thermal conductivity results are in accord with the assumption of a linear specific heat term $C_1 = aT$ with $a \simeq 1\ \mu J/g \cdot K^2$, which corresponds to the resolution of previous specific heat measurements,[15,16] and is of the same magnitude as that found in glasses.[17] Following Zaitlin and Anderson,[18] we assume a parabolic density of states for LEE, $n(\epsilon) = n_0[1 + \alpha(\epsilon/k_B)^2]$, where $\alpha = 0.072b'/a$ and b^1T^3 is that part of ΔC arising from LEE (after Stephens[17]). From $n(\epsilon)$ a phonon free path $l \sim [T(1 + 16\alpha T^2)]^{-1}$ is obtained in the dominant-phonon approximation ($\hbar\omega_{dom} = 4k_B T$). Adjusting the scattering strength of the LEE to agree with the experimental result at 0.7 K, we find good agreement with our data up to 4 K (cf. dotted curve in Fig. 2). Of course, below 0.7 K, where the film conductance constitutes less than 3% of the total conductance and hence is highly uncertain, no serious comparison with experimental data is possible.

For a rough estimate we infer from this fit of k_s^{ph} the excess *cubic* specific heat term, $b' \simeq 2$ $\mu J/g \cdot K^4$. This is of the same order as that found for vitreous SiO_2,[18] but it is only 10% of ΔC, as measured for the related $Pb_{0.7}Bi_{0.3}$ system.[19] Hence ΔC has to be almost entirely attributed to a softening of elastic waves. This view is supported by measurements on the glassy metal $Pd_{0.775}Si_{0.165}Cu_{0.06}$ where ΔC could quantitatively be explained by a change in sound velocity.[20]

The abrupt increase of k_s^{ph} above 4 K hints at a possible resonant scattering mechanism which is not observed in bulk (normal-state) glasses. Experiments on amorphous films of other superconductors—to resolve the origin of this new feature—are in preparation.

We conclude by stating that the apparent absence of spatially extended lattice defects in the amorphous *Pb*Cu film leads to the fact that its phonon conductivity is higher than that of its crystalline counterpart. The k_s^{ph} vs T curve in the superconducting state provides direct evidence that LEE exist also in a "simple" amorphous metal, i.e., are not confined to disordered solids with covalent bonds.

This work was performed within the research program of the Sonderforschungsbereich 125 Aachen/Jülich/Köln.

[1]R. C. Zeller and R. O. Pohl, Phys. Rev. B 4, 2029 (1971).
[2]H. v. Löhneysen and F. Steglich, to be published.
[3]P. W. Anderson, B. I. Halperin, and C. M. Varma, Philos. Mag. 25, 1 (1972).
[4]W. A. Phillips, J. Low Temp. Phys. 7, 351 (1972).
[5]J. R. Matey and A. C. Anderson, J. Non-Cryst. Solids 23, 129 (1977), and Phys. Rev. B 16, 3406 (1977).
[6]R. Hilsch and F. Steglich, Z. Phys. 226, 182 (1969).
[7]B. J. Mrstik and D. M. Ginsberg, Phys. Rev. B 5, 1817 (1972).
[8]R. Hilsch and W. Martienssen, Nuovo Cimento 7, Suppl. 2, 480 (1958).
[9]A. W. Bjerkaas, D. M. Ginsberg, and B. J. Mrstik, Phys. Rev. B 5, 854 (1972).
[10]H. Bülow and W. Buckel, Z. Phys. 145, 141 (1956).
[11]S. G. O'Hara and A. C. Anderson, Phys. Rev. B 10, 574 (1974).
[12]L. S. Palatnik, in *Grundprobleme der Physik dünner Schichten, Clausthal-Göttingen*, edited by R. Niedermeyer and H. Mayer (Vandenhoek and Ruprecht, Göttingen, 1965), p. 102.
[13]Although the "crystalline-amorphous transition" is usually accompanied by an increase in electron-phonon coupling, the assumption of an unchanged gap parameter $\eta = 5.0$ seems to be reasonable, for $\eta(a\text{-}Pb_{0.9}Cu_{0.1}) = 4.75$ [K. Knorr and N. Barth, Solid State Commun. 8, 1085 (1970)] and $\eta(a\text{-}Pb_{0.9}Bi_{0.1}) = 5.0$ [J. D. Leslie, J. T. Chen, and T. T. Chen, Can. J. Phys. 48, 2783 (1970)]. An increase of η from 5.0 to 5.4 would enhance k_s^{ph} values between 2 and 6.5 K by up to 20%, but the shape of the k_s^{ph} vs T curve (cf. Fig. 2) would remain essentially unchanged.
[14]M. P. Zaitlin, L. M. Scherr, and A. C. Anderson, Phys. Rev. B 12, 4487 (1975).
[15]G. Krauss and W. Buckel, Z. Phys. B20, 147 (1975).
[16]A. Comberg, S. Ewert, and H. Wühl, Z. Phys. B25, 173 (1976).
[17]R. B. Stephens, Phys. Rev. B 8, 2896 (1973).
[18]M. P. Zaitlin and A. C. Anderson, Phys. Rev. B 12, 4475 (1975).
[19]S. Ewert and W. Sander, Z. Phys. 247, 21 (1971).
[20]B. Golding, G. Bragley, and F. S. L. Hsu, Phys. Rev. Lett. 29, 68 (1972).

Evidence of a Large Superfluid Vortex in ^{4}He

Philip L. Marston[a] and William M. Fairbank
Department of Physics, Stanford University, Stanford, California 94305
(Received 8 September 1977)

Optical interferometry was used to measure surface profiles of He II. Depressions observed following rapid container spinup agreed with theoretical profiles of a superfluid vortex with circulation $\approx 400h/m$.

The energetically preferred state of rotating He II is a uniform distribution of vortex lines, each with a circulation Γ predicted to be $\kappa = h/m$, where h is Planck's constant and m is the ^{4}He atomic mass.[1] Photographs of vortex lines by Williams and Packard[2] show a nearest-neighbor spacing roughly consistent with that Γ. Quantization of Γ, however, does not rule out the energetically less favorable creation of a vortex having $\Gamma = N\kappa$ with integer $N > 1$. Feynman[3] noted that if a vortex with large N were created, the liquid surface above it would be noticeably depressed. This Letter describes observation of depressions in He II pools with a thickness of several microns. Depressions were accurately described by a model with superfluid flow in the bulk liquid being that of a vortex with $N \approx 400$. At the center of each depression, the substrate was covered by a thin He II film and details of flow there could not be determined.

When calculating a vortex's surface profile, it is important to include the influences of the surface tension σ and the wetting of the container. For a quasisteady profile, there is a pressure balance at each point on the surface given by Laplace's formula: $P = P_v + \sigma(R_1^{-1} + R_2^{-1})$, where P and P_v are the pressures on the liquid and vapor sides of the interface and R_1 and R_2 are principal radii of curvature. For purely azimuthal flow about the center of a cylinder of radius b, R_1 and R_2 lead to a differential equation[4] for the surface height $Z(r)$ at radius r. When linearized for $Z' \ll 1$, it becomes[5]

$$Z'' + Z'r^{-1} - a^{-2}Z = -\sigma^{-1}f(r), \quad (1)$$

$$f(r) = \int^r (\rho_s v_{s\theta}^2 + \rho_n v_{n\theta}^2) r^{-1} dr, \quad (2)$$

where differentiation with respect to r is indicated by a prime; $a = [\sigma/g(\rho - \rho_v)]^{1/2} \approx 0.5$ mm is the capillary constant; g is the acceleration of gravity; ρ, ρ_s, ρ_n, and ρ_v are the densities of the liquid, superfluid, normal fluid, and vapor, respec-