Glasslike lattice vibrations in the quasicrystal $Al_{72.1}Pd_{20.7}Mn_{7.2}$

EunJoo Thompson, P. D. Vu,* and R. O. Pohl†
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853

(Received 13 July 2000)

The internal friction, speed of transverse sound, and thermal conductivity have been measured of a single grain of icosahedral $Al_{72.1}Pd_{20.7}Mn_{7.2}$ (composition in the melt) that was grown by the Czochralski technique at Forschungszentrum Jülich. The three sets of data could be consistently described with the tunneling model using the tunneling strength $C_t=(0.5\pm0.1)\times10^{-4}$. This value of C_t, which is close to the one obtained by speed of sound measurements by Vernier *et al.* on a sample of *i*-AlPdMn grown by the Bridgman technique, and from thermal conductivity of a sample grown by the Czochralski technique by Legault *et al.*, and of a Bridgman sample measured by Chernikov *et al.*, is close to the range observed for most amorphous solids, 10^{-4}–10^{-3}. It is concluded that the lattice vibrations of this icosahedral quasicrystal, grown from the melt in thermal equilibrium, are indistinguishable from those of amorphous solids, and of many chemically disordered crystals.

I. INTRODUCTION

The lattice vibrations of amorphous solids differ from those of perfect crystals in characteristic ways. In perfect crystals, lattice vibrations are described by plane elastic waves or phonons. In amorphous solids, localized excitations, often described as tunneling states, are needed in addition to the plane waves to describe the vibrations at frequencies below $\sim10^{11}$ Hz. They determine the specific heat, thermal conductivity, and the acoustic properties at temperatures below several Kelvin.[1–3] Furthermore, in the frequency regime above several terahertz, the phonon mean free path in amorphous solids becomes so short that the wave picture becomes inapplicable, and has to be replaced with one of localized atomic vibrations with lifetimes on the order of the vibrational period.[4,5] These short-lived excitations lead to a very small thermal conductivity, typically above ~50 K, called the minimum thermal conductivity.[6]

With the discovery of quasicrystals, the question arose whether their lattice vibrations resemble more closely those of crystals or of amorphous solids. Early results were ambiguous. In specific-heat measurements on superconducting icosahedral as well as amorphous $Mg_3Zn_3Al_2$ above 0.1 K, a linear specific-heat anomaly could not be identified due to the scatter of the data.[7] Measurements of the thermal conductivity of amorphous and icosahedral $Pd_{58.8}U_{20.6}Si_{20.6}$ between 0.15 and 2 K yielded very similar results, although neither of them showed the T^2 dependence expected for amorphous solids.[8] Here the icosahedral phase had been formed by annealing an amorphous ribbon, which resulted in quasicrystal particles of size 1500–2000 Å filling 80% of the volume, with particles ~100 Å in diameter filling the remaining space. It seems likely that such ribbons contained considerable structural disorder which might also lead to a thermal conductivity resembling that of amorphous solids, as observed, for example, in plastically deformed aluminum.[9,10] Thus the existence of glasslike excitations in *i*-PdUSi could not be concluded with certainty.

Sound velocity measurements in *i*-Al_5Li_3Cu and *i* $-Mg_3Zn_3Al_2$ between ~10 mK and 10 K on vibrating reeds revealed temperature dependencies which qualitatively resembled those observed in amorphous metals.[11] However, the tunneling strengths C, as defined in the tunneling model,[1–3] determined to be 7×10^{-6} and 1.4×10^{-5} on the ribbons of *i*-Al_5Li_3Cu and *i*$-Mg_3Zn_3Al_2$, respectively, were smaller than observed in amorphous metals, for example *a*-$Pd_{78}Si_{16}Cu_6$ ($C=8.7\times10^{-5}$ measured on a vibrating reed[12]) and $Cu_{0.3}Zr_{0.7}$ ($C=2.9\times10^{-4}$ measured through thermal conductivity[13]). Furthermore, an even smaller tunneling strength $C=3.6\times10^{-6}$ was found on a large, single-grain sample of $Al_{5.1}Li_3Cu$, which had been slowly cooled.[11] This value comes close to that observed in accidentally deformed cubic crystals (which probably contained accidental defects), and it was therefore concluded that the density of states of the tunneling defects in these icosahedral quasicrystals was more a function of accidental disorder, for instance phason strain, rather than a result of the intrinsic structure as in amorphous solids.[11] The same conclusion was reached in measurements of the internal friction of a variety of icosahedral ribbons.[14] It was suggested at that point that glasslike excitations were more likely to occur in metastable alloys, rather than in well annealed, thermodynamically stable quasicrystals. In light of the observations summarized above, the availability of large, well annealed and thermally equilibrated quasicrystals of the composition AlPdMn and AlPdRe grown either by seed pulling from the melt (Czochralski technique) or by the gradient method (Bridgman) offered the exciting opportunity of answering the question whether under these conditions their lattice vibrations resemble those of crystals or of glasses.

Measurements of either thermal conductivity[15–18] or speed of sound,[19] on such single grain samples showed a behavior similar to that of amorphous solids both in temperature dependence and magnitude. However, what has been missing so far are measurements of thermal conductivity, speed of sound, and ultrasonic attenuation all on the same sample, along with the successful description of all the data with the tunneling model (TM), using the same tunneling

 ©2000 The American Physical Society

strength C as fitting parameter. This is the purpose of the present study. We will combine the presentation of our data on a single grain sample of AlPdMn with a review of earlier studies of the thermal and elastic properties of this quasicrystal and of i-AlPdRe. Since quasicrystals contain free electrons, their thermal and elastic properties should be compared with those of amorphous metals, rather than amorphous dielectric solids. It is known that the low-energy excitations of amorphous metals depend on the thermal history.[13,20,21] This is in contrast to dielectrics, in which the effect of heat treatment is very small, as reviewed by Johari.[22] A striking sample dependence will also be seen in i-AlPdMn quasicrystals, and will be connected to their heat treatment.

As the work to be reported here was in progress, Bert *et al.*[23] reported on a search for low-energy tunneling states in single grains of $Al_{63}Cu_{25}Fe_{12}$ prepared by levitation melting and subsequent annealing. Through measurements of speed of sound and ultrasonic attenuation at 190 MHz, they reported a speed of sound varying as the logarithm of the temperature, as predicted by the TM. However, in contrast to the TM prediction, they did not observe a temperature-independent plateau of the internal friction which, furthermore, was one order of magnitude larger than predicted. They concluded that tunneling states alone were inadequate to describe the acoustic properties on this quasicrystal. We will return to these findings in Sec. VI.

II. EXPERIMENTAL MATTERS

The single grain i-AlPdMn quasicrystal studied in this investigation was grown by the Czochralski method by Dr. M. Feuerbacher at the Institut für Festkörperforschung in Jülich, Germany. The composition in the melt was 72.1 at. % Al, 20.7 at. % Pd, and 7.2 at. % Mn. The sample was cut by spark erosion with all the edges parallel to twofold axes and the long axis oriented along a fivefold direction. The surfaces were then sanded to a rod of 1.9×2.1 mm cross section and a length of 20.9 mm. The internal friction and speed of sound was measured simultaneously in torsion at 90 kHz, using a composite oscillator technique described in Ref. 24, and also in detail in Ref. 9. Measurements below 2 K were made in a dilution cryostat, and those from 1.5–200 K in an insertable ^{4}He cryostat.[25] The thermal conductivity of the same sample was measured by the standard steady-state four-probe technique using a heater, two thermometers, and a dry-clamping method as detailed in Ref. 26.

III. THERMAL CONDUCTIVITY

Our measurements on AlPdMn are shown in Fig. 1, together with previous measurements on one sample of AlPdRe, and on three samples of AlPdMn. The sample of $Al_{70}Pd_{21.4}Re_{8.6}$ was prepared by arc melting, subsequently annealed at 950 °C (2 days) and 650 °C (2 days), then rapidly cooled to room temperature.[18] Average grain size, according to scanning electron microscopy, was $\sim1\times10^{-3}$ cm. One sample of $Al_{70}Pd_{21}Mn_9$ [labeled (a) in Fig. 1] was also produced by arc melting, annealed at 800 °C (2 days), and then quenched in water.[15] Another sample of AlPdMn [of unspecified composition, labeled (b) in Fig. 1],

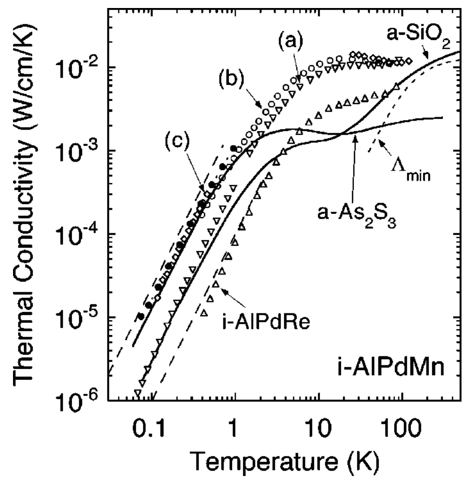

FIG. 1. Thermal conductivity of our i-AlPdMn sample, (solid circles) together with previously published data, on three differently prepared i-AlPdMn samples, (a) inverted triangles (Ref. 15); (b) open circles (Ref. 17); (c) open diamonds (Ref. 16); AlPdRe, open triangles (Ref. 18); a-As_2S_3 and a-SiO_2 for comparison, solid curves. The short dashed curve, Λ_{min}, is calculated for the AlPdMn sample and is almost identical to the Λ_{min} of a-SiO_2 and AlPdRe. The two dashed lines indicate the glassy range, the range of the thermal conductivity of all amorphous solids measured (Ref. 31).

was of a single grain, Czochralski-seed pulled, with no subsequent heat treatment.[17] The third sample (c) was a single grain grown by the Bridgman-(gradient) method, also with no information on any subsequent heat treatment.[16] Our own measurements, shown as solid circles, as performed on the sample described in Sec. II, agree very well with those obtained on (c).

The data shown in Fig. 1 are the total thermal conductivity Λ measured without corrections for heat transport by electrons, except for sample (b), for which only the lattice thermal conductivity has been published. However, since the electrical conductivity of these quasicrystals[27–29] is on the order of $10^2\ \Omega^{-1}\,\mathrm{cm}^{-1}$ (much smaller than that of a-PdSiCu,[30] given as 13 000 $\Omega^{-1}\,\mathrm{cm}^{-1}$) the electronic contribution to the heat flow is expected to be negligible, according to the Wiedemann-Franz-Lorenz law. Therefore the distinction between lattice and total thermal conductivity is concluded to be unimportant for all the quasicrystals shown. Over the entire temperature range investigated, these data resemble closely those obtained on amorphous dielectric solids with data on a-As_2S_3 and a-SiO_2 shown here for comparison. Below ~1 K, the data for all samples of i-AlPdMn lie in the range found for all amorphous solids measured to date (the glassy range, see Ref. 31) indicated by the two dashed lines.

Conduction electrons not only carry heat; they also reduce the heat flow by scattering the heat carrying phonons. In a-PdSiCu, it has been shown that phonon scattering by electrons dominates above ~1 K, while below this temperatures, the scattering is dominated by tunneling states.[30,32] Encouraged by this separation of the two scattering mechanisms acting in the amorphous metal we conclude that the observed thermal conductivity in the quasicrystals which varies as T^2 below ~1 K, and which has the same magnitude as observed in amorphous solids, is also the result of

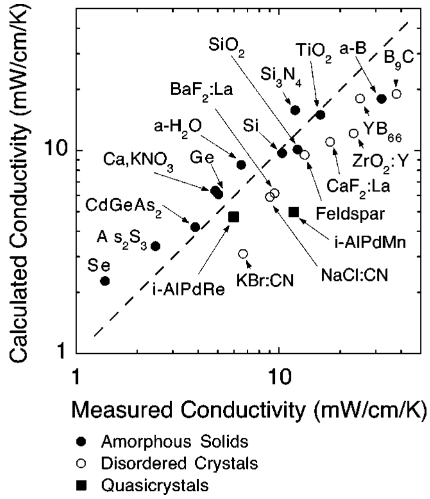

FIG. 2. Comparison of the predicted minimum thermal conductivity, Eq. (1), to that measured at 300 K on amorphous solids and of highly disordered crystals containing glasslike excitations (Refs. 31 and 33). Also shown is the comparison for the quasicrystals at the highest temperature of measurement at 100 K.

two-level systems. Any scattering by electrons will be observed only at higher temperatures, if at all.

Above ~50 K, the thermal conductivities approach that measured in a-SiO_2. In amorphous solids, the high temperature thermal conductivity has been found to be close to the minimum thermal conductivity $\Lambda_{\min}$ calculated for these solids. We calculated the minimum conductivity for $Al_{72.1}Pd_{20.7}Mn_{7.2}$ sample using the method outlined in Ref. 33 and the expression

$$\Lambda_{\min}=\left(\frac{\pi}{6}\right)^{1/3}k_B n^{2/3}\left[\sum_i v_i\left(\frac{T}{\theta_i}\right)^2\right]\int_0^{\theta_i/T}\frac{x^3e^x}{(e^x-1)^2}dx, \tag{1}$$

where the sum is taken over the three sound modes (two transverse and one longitudinal) using the speed of sound $v_t=3.8\times10^5$ cm/s (from our present measurement which agrees with Ref. 19) and $v_l=6.5\times10^5$ cm/s from Ref. 34; $n=6.73\times10^{22}$ atoms/cm^3 is the number density of atoms found from the mass density $\rho=5.08$ g/cm^3; θ_i is the cutoff frequency for each polarization expressed in K:

$$\theta_{l,t}=v_{l,t}\left(\frac{\hbar}{k_B}\right)(6\pi^2 n)^{1/3}. \tag{2}$$

In Fig. 1, we have only shown one curve for $\Lambda_{\min}$ since the minimum conductivity of i-AlPdMn and i-AlPdRe (calculated in Ref. 18) are all very similar to that of a-SiO_2. Although the measurements do not extend much above 100 K, an extrapolation to higher temperatures naturally approaches the theoretical curve of $\Lambda_{\min}$. Figure 2 shows a comparison of the predicted thermal conductivity $\Lambda_{\min}$ at 300 K to that measured in amorphous solids and in disordered crystalline solids containing glasslike excitations.[5] The data points lie close to the dashed line which indicates perfect agreement between the value predicted by Eq. (1) and the experiment. It is seen that the data for the two quasicrystals also lie close to the dashed line, thus confirming the claim that their high-temperature thermal conductivity is close to the minimum thermal conductivity.

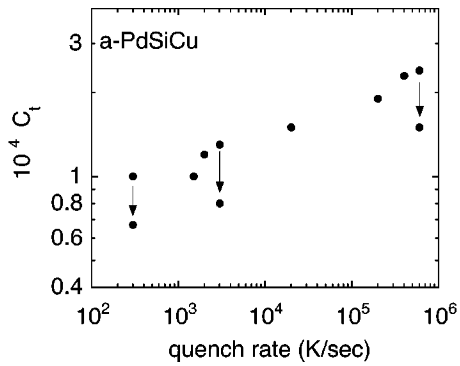

FIG. 3. The tunneling strength C_t for a-PdSiCu, as determined from the thermal conductivity in the temperature region where it varies as T^2, using Eq. (3). Data for the 300 K/s quench rate are from Ref. 21, all others from Refs. 30 and 32. The arrows point towards data obtained after anneals at 573 K (1 h for the 3×10^3-K/s sample, 6 h for the others).

On the basis of the arguments presented here, we conclude that the thermal conductivities observed on the five quasicrystals shown in Fig. 1 are very similar to those observed in amorphous dielectric solids. There is, however, one important difference: In amorphous dielectrics, the thermal conductivity (if we concentrate on the measurements below a few K) and the tunneling strength is largely independent of sample preparation and heat treatment.[22] This independence is clearly not observed in the quasicrystals, which in this respect resemble amorphous metals. As an example of the latter we show in Fig. 3 the variation of the tunneling strength C_t determined from the thermal conductivity for samples of a-PdSiCu prepared at different quench rates and after subsequent anneals (the subscript t refers to transverse waves, the dominant carriers of heat). According to the TM, the low temperature lattice thermal conductivity Λ is given by

$$\Lambda=\frac{2.66k_B^3}{6\pi\hbar^2}\frac{1}{C_t v_t}T^2=\beta_o T^2, \tag{3}$$

where v_t is the transverse speed of sound ($v_t=1.797\times10^5$ cm/s), and some simplifying assumptions have been made regarding the longitudinal phonons and their influence on the heat transport, as described, e.g., in Ref. 35. The sources for the data shown are listed in the caption of Fig. 3. Depending on the sample preparation, C_t can vary by as much as a factor of 4. Returning now to the quasicrystals, a similar variation appears to be found in i-AlPdMn, depending on its growth conditions. We list in Table I the prefactor β_o of the best fit of the form $\Lambda=\beta_o T^2$ to the five curves shown in Fig. 1, and the tunneling strength C_t derived from β_o.

To summarize: in the three single grain AlPdMn samples which were slowly grown from the melt, either by seed pulling or by the gradient method, the thermal conductivities are nearly sample independent, vary as T^2 below 1 K, lie in the glassy range and have tunneling strength of similar magnitude. Their thermal conductivities approach the minimum thermal conductivity above ~100 K. In the intermediate

TABLE I. The first and second columns list the composition of the quasicrystal measured and their growth method; column 3 is the prefactor of the T^2 thermal conductivity using Eq. (3); columns 4–6 are the tunneling strength C_t determined from the thermal conductivity Λ, from the acoustic attenuation Q^{-1}, and from the speed of sound v of the transverse waves, respectively, as indicated with the second subscript; column 7 gives the reference for each measurement. Note that the tunneling strengths for all quasicrystals grown under thermal equilibrium range within a factor of 2 regardless of the measurement from which they were determined.

Composition	Growth method	β_o (10^{-4} W/cm K^3)	$C_{t,\Lambda}$ (10^{-4})	$C_{t,Q^{-1}}$ (10^{-4})	$C_{t,v}$	Reference
$Al_{72.1}Pd_{20.7}Mn_{7.2}$ [a]	Czochralski	16	0.57	0.38	0.38 (90 kHz)	this work
$Al_{68.7}Pd_{21.7}Mn_{9.6}$ [b]	Bridgman				0.3 (28 MHz)	19
$Al_{68.7}Pd_{21.7}Mn_{9.6}$ [b]	Bridgman				0.33 (94 MHz)	19
$Al_{68.7}Pd_{21.7}Mn_{9.6}$ [b]	Bridgman				0.43 (423 MHz)	19
$Al_{70}Pd_{21}Mn_9$ [b]	arc melt [d]	4.0	2.2			15
$Al_{70}Pd_{20}Mn_{10}$ [b]	Bridgman	15	0.6			16
AlPdMn [c]	Czochralski	12	0.8			17
$Al_{70}Pd_{21.4}Re_{8.6}$ [b]	arc melt [e]	1.1	3.8			18

[a]Composition in the melt.
[b]Composition in the solid.
[c]Unspecified composition.
[d]Arc melt at 800 °C and water quenched.
[e]Arc melt at 950 °C, 600 °C and air cooled.

temperature range, around 20 K the conductivity shows a maximum as observed in disordered crystals, a point noted previously.[17] In rapidly quenched samples, the thermal conductivity is strongly depressed below ~ 1 K.

IV. SPECIFIC HEAT

The linear specific heat together with thermal relaxation ought to be a particularly clear indicator of the broad spectrum of tunneling states which occur in amorphous solids. Unfortunately, no convincing evidence has been found in either AlPdMn or AlPdRe (Refs. 18, 36 and 37) by this technique, since at low temperatures not only electrons but also magnetic effects dominate the specific heat C_p. Peaks of C_p/T^3 were observed at ~ 20 K and ~ 30 K in these solids, respectively, and also evidence for a T^3 specific heat in excess of that expected on the basis of the speeds of sound using the Debye model. Similar anomalies have also been reported for amorphous solids,[38] but neither can be viewed as convincing evidence for glasslike lattice vibrations. There appears to be little hope of a clear identification of a linear specific heat anomaly until thermodynamically stable quasicrystals become available which are nonmagnetic and superconducting. For the time being, the identification of glasslike excitations in quasicrystals will therefore have to rely on thermal conductivity and acoustic measurements.

V. ACOUSTIC PROPERTIES

The transverse speed of sound measured on our sample at 90 kHz is shown in Fig. 4. A comparison with previous measurements at higher frequencies[19] shows very similar behavior. The velocity peaks at the same temperature, regardless of the measuring frequency and resembles that of amorphous metals. In contrast, the temperature of the peak for dielectric amorphous solids would depend on the frequency $f^{1/3}$ in accordance to the TM. On the low-temperature side of the peak in Fig. 4, the velocity changes logarithmically with temperature (the solid lines), except for an anomaly observed in all curves at the lowest temperatures. By measuring the speed of sound in a magnetic field, Vernier *et al.*[19] made this anomaly disappear (open symbols), thus showing its magnetic character, and revealing a clear logarithmic dependence of $\Delta v/v_0$ to the lowest temperatures. We fit an expression to these low-temperature data of the form given by the TM for amorphous metals:[12]

$$\frac{v-v_0}{v_0}=\frac{\Delta v}{v_0}=\frac{1}{2}C_t\ln\frac{T}{T_o}, \tag{4}$$

where C_t is the tunneling strength, and v_o is the transverse speed of sound at some reference temperature ($v_0=3.8$

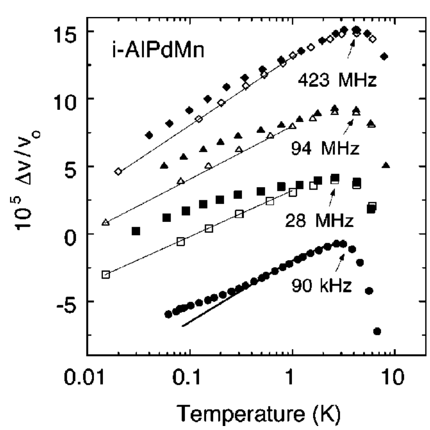

FIG. 4. Relative variation of the transverse speed of sound in single grain i-AlPdMn at 90 kHz (solid circles, this study) and at higher frequencies (Ref. 19) (open symbols: with magnetic field, closed symbols: without). The velocity maximum is independent of the measuring frequency which is also observed in amorphous metals.

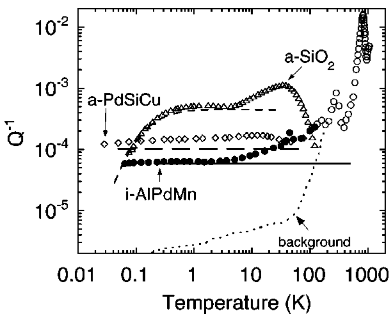

FIG. 5. Internal friction of single grain i-AlPdMn quasicrystal measured in torsion at 90 kHz in this study (solid circles) and at 3 Hz (open circles from Ref. 39). In the temperature of overlap, the agreement is excellent for the two samples. The solid line which fits the data very well below 2 K is computed from the TM using the tunneling strength C_t as determined from the speed of sound measured on the same sample (Fig. 4). Equally good agreement with the TM prediction using the parameters from speed of sound measurements is shown for a-SiO_2 (Ref. 24) at 90 kHz (dashed curve and open triangles), and for a-PdSiCu (Ref. 12) at 1038 kHz (long dashed curve and open diamonds). The measurements of a crystalline quartz sample, background, are shown as a dotted line.

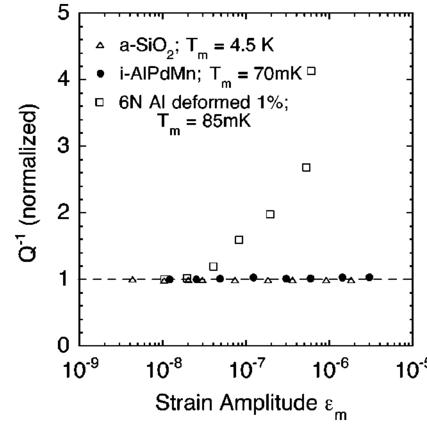

FIG. 6. Strain amplitude dependence on a normalized scale of a i-AlPdMn quasicrystal measured at 70 mK compared with a-SiO_2 (4.5 K) and a 6N aluminum deformed 1% (85 mK). Both the quasicrystal and silica show no strain amplitude dependence. Therefore dislocation tunneling as seen in the Al sample is unlikely in the quasicrystal.

$\times 10^5$ cm/s for i-AlPdMn). The tunneling strengths C_t determined are also listed in Table I. They are independent of frequency, as predicted by the TM, and show no sample dependence.

Acoustic attenuation at low temperatures has only been measured on our sample, see the solid circles in Fig. 5. Note that our data agree well with those reported above 100 K by Feuerbacher *et al.*[39] at a frequency of 3 Hz on an identically prepared but different AlPdMn sample, open circles. Between 10 K and 0.05 K, the lowest temperature of our measurement, Q^{-1} is constant, as also found in amorphous metals, and illustrated in Fig. 5 with measurements on a-PdSiCu by Raychaudhuri and Hunklinger.[12] The dropoff of the internal friction below 1 K occurring in amorphous dielectrics, (shown in Fig. 5 with measurements on a-SiO_2), can be understood in the framework of the TM with the assumption that the relaxation of the tunneling defects occurs only via one-phonon processes. In amorphous metals, the absence of a similar drop-off has been explained by the fact that conduction electrons provide an effective channel for relaxation at the lowest temperatures.[12] The same explanation is expected to apply for quasicrystals. From the magnitude of the internal friction plateau, $Q^{-1}=6\times 10^{-5}$, the tunneling strength $C_t=0.38\times 10^{-4}$ is determined, in perfect agreement with the value determined from the speed of sound; see Table I.

The physical nature of the intrinsic low-energy excitations in the i-AlPdMn is, of course, as unknown as it is in the amorphous solids. In plastically deformed crystalline aluminum, we have recently identified tunneling dislocations as the defects causing ultrasonic attenuation at low temperatures.[40] Their motion resulted in a strain-amplitude-dependent internal friction, as shown in Fig. 6 for a 99.9999% (6N) Al rod which had been annealed prior to a 1% plastic deformation by stretching. At 85 mK, the internal friction increased fourfold as the ultrasonic strain amplitude ϵ_m was increased by two orders of magnitude to 10^{-6}. Such a dependence is absent in amorphous solids, as shown for a-SiO_2 in Fig. 6. Since dislocations have been observed in plastically deformed i-AlPdMn,[41] we decided to search for their low-temperature motion by measuring our (undeformed) sample with increasing ϵ_m. As is seen in Fig. 6, the absence of any strain amplitude dependence gives no evidence for any dislocation tunneling in this solid.

VI. DISCUSSION

Low-temperature measurements of the thermal conductivity, speed of sound, and ultrasonic attenuation, were made on the same sample of a single grain of i-AlPdMn cut from a boule grown by seed pulling from the melt (Czochralski). These measurements are well described with the TM, using the tunneling strength $C_t=(0.5\pm 0.1)\times 10^{-4}$ as the only free parameter, summarized in Table I. The same C_t also describes the earlier speed of sound measurements at much higher frequencies by Vernier *et al.*[19] on a Bridgman-grown sample. Thermal conductivity measurements on a Bridgman-grown and on a Czochralski-grown sample, performed by Legault *et al.*[16] and by Chernikov *et al.*,[17] respectively, can also be described with nearly the same C_t. All these results provide convincing evidence that glasslike excitations exist in the most carefully grown icosahedral samples and are intrinsic to this thermodynamically stable quasicrystal, thus answering a long-standing question of whether the lattice vibrations of quasicyrstals resemble those of amorphous solids. We repeat that the same conclusion had been reached earlier by Vernier *et al.*,[19] Legault *et al.*,[16] and by Chernikov *et al.*,[17] based on their measurements of either thermal conductivity or speed of sound only. The present work, however, provides convincing evidence by combining the thermal with acoustic measurements. The tunneling strength determined here lies at the lower limit of that found for most amorphous solids, which span roughly the range from 10^{-4}–10^{-3}, as reviewed in Ref. 42. It is, however, close to that of annealed amorphous metals. As shown in Fig. 3, in

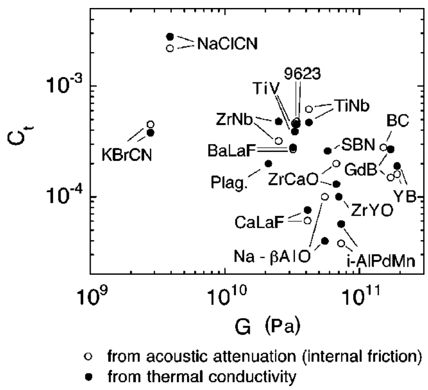

FIG. 7. Tunneling strength C_t of crystals with glasslike excitations, determined from thermal conductivity (solid circles) and from sound attenuation or internal friction (open circles). Chemical formulas of the crystals plotted along with their references are as follows: KBrCN: $(KBr)_{0.25}(KCN)_{0.75}$ (Ref. 35); NaClCN: $(NaCl)_{0.9}(NaCN)_{0.1}$ (Ref. 35); BaLaF:$(BaF_2)_{0.54}(LaF_3)_{0.46}$ (Ref. 35); CaLaF: $(CaF_2)_{0.74}(LaF_3)_{0.26}$ (Ref. 35); ZrCaO: $(ZrO_2)_{0.89}(CaO)_{0.11}$ (Ref. 35); ZrYO: $(Zr0_2)_{1-x}(Y_20_3)_x$, $0.10<x<0.18$ (Ref. 44); GdB: $GdB_{62.5}$ (Ref. 35); YB: YB_{63} (Ref. 35); TiV: $Ti_{0.67}V_{0.33}$ (Ref. 35); TiNb: $Ti_{0.63}Nb_{0.37}$ (Ref. 35); Plag.: Plagioclase Feldspar (Refs. 33,35); 9623:Corning type 9623, ceramicized, thermal conductivity (Ref. 45), internal friction (Ref. 46); SBN: strontium barium niobate (Ref. 47); Na-βAlO: sodium-β-alumina, thermal conductivity (Ref. 48) ultrasonic attenuation (Ref. 49); BC: B_9C (Ref. 42).

a-PdSiCu values of C_t ranging from 0.7×10^{-4}–2.5×10^{-4} have been observed in thermal conductivity measurements with the smaller values observed on the slower cooled and annealed samples (similar values have been reported in acoustic measurements (reviewed in Ref. 9). Thus the range of C_t observed on the different slowly grown boules of *i*-AlPdMn may have variations in sample growth as their origin. In the very rapidly cooled arc-melted sample, the large value of $C_t=2.2\times10^{-4}$ may have a similar origin (see also the large value observed for the arc-melted AlPdRe sample, Table I). However, no firm conclusion can be drawn from the arc-melted samples until acoustic measurements confirm this value of C_t, since the reduced thermal conductivity could also result from scattering mechanism other than resonant scattering by the tunneling states, e.g., dislocations or small angle grain boundaries.

On a sample of $Al_{63}Cu_{25}Fe_{12}$ produced by levitation melting followed by a 4-day anneal at 860 °C, and a rapid quench to room temperature, Bert *et al.*[23] recently measured the low-temperature speed of transverse sound at 190 MHz. From its logarithmic temperature dependence below 0.4 K, we determine $C_t=0.68\times10^{-4}$ with the use of Eq. (4). Without measurements of the thermal conductivity, no comparison with the tunneling strengths obtained on *i*-AlPdMn is possible, although the general agreement is encouraging (we mention again that the acoustic attenuation observed by Bert *et al.* did not agree with the prediction of the tunneling model, using the tunneling strength C_t determined from the speed of sound).

The existence of glasslike low-energy excitations in the thermodynamically stable icosahedral AlPdMn is now firmly established. We now turn to the question whether we can utilize our knowledge of the structure of quasicrystals in order to develop a picture of their physical nature which may also apply to amorphous solids. These quasicrystals are not the only nonamorphous solids with low-energy excitations resembling those of amorphous solids both qualitatively and quantitatively. A review of the most thoroughly studied disordered crystals with glasslike lattice vibrations has been given by Topp and Cahill,[35] see also Ref. 24. In Fig. 7, we summarize the tunneling strength C_t observed in these disordered crystals, as obtained by thermal conductivity and acoustic attenuation measurements, together with those obtained here on *i*-AlPdMn. Note that the two techniques yield values of C_t which usually differ by a factor of 2 or less. Probably the most convincing evidence that the lack of long-range order is irrelevant for the occurrence of the low-energy excitations was given with a study of internal friction and thermal phonon scattering in ion-implanted silicon.[43] Depending on the ions used, the silicon either amorphized (by Si^+ ion bombardment) or it remained crystalline (B^+ ions). Yet, in either case the tunneling strength saturated with increasing dose at the same value, 0.21×10^{-4}. In an attempt to understand this startling result, it was suggested that with increasing dose, the buildup of point defects leads to a local increase of random strains. The atoms in the neighborhood of the spikes of such strains then may become unstable, and can tunnel between different potential minima. The saturation of these tunneling centers, and with that, of the tunneling strength C_t, is reached when the stress spikes begin to overlap, i.e., when the solid locally rearranges or ''yields.'' Conceivably, the amorphous structure is determined by the same upper limit of the local stresses. By mentioning it in the present context, we want to raise the question whether enough is known about the structure and local stresses in quasicrystals to either support or refute it as an origin of the glasslike excitation in the quasicrystals.

Finally, we turn to the high-temperature thermal conductivity of the quasicrystals. As discussed in Sec. III in amorphous dielectric solids the thermal conductivity above $\sim$50 K approaches its minimum value, in which the heat flow is best described by a random walk from one atom to its neighbor, with a jump time on the order of $\frac{1}{2}$ its vibrational period. In disordered crystals with glasslike low-energy excitations, the same heat-flow mechanism appears to prevail at high temperatures, and can be understood by their atomic disorder. In quasicrystals *i*-AlPdMn and *i*-AlPdRe, the thermal conductivity at high temperatures also approaches this minimum value. However, in these solids, order is observed over many atomic distances. It is not understood why this order should not lead to larger thermal conductivities.

VII. CONCLUSION

Through thermal and acoustic measurements, we have clearly confirmed earlier suggestions that the lattice vibrations of *i*-AlPdMn are glasslike. By extending the existence of such excitations beyond chemically disordered or ion-implanted crystals to quasicrystals, we conclude that the physical nature of these excitations is becoming even more restricted, although it still defies our understanding.

ACKNOWLEDGMENTS

We thank Dr. M. Feuerbacher of the Forschungszentrum Jülich, Germany, for supplying the sample investigated and for many useful communications. We also thank Dr. C. Henley, Dr. V. Elser, and Dr. J. Sethna for fruitful discussions. We acknowledge the financial support by the National Science Foundation under Grant No. DMR-9701972 and by the Cornell Center for Materials Research, NSF Grant No. ECS-93 19 005.

*Present address: Soka University of America, Aliso Viejo, CA 92656.

†Email address: pohl@ccmr.cornell.edu

[1] *Amorphous Solids, Low Temperature Properties*, edited by W. A. Phillips (Springer, Berlin, 1981).

[2] S. Hunklinger and A. K. Raychaudhuri, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (Elsevier Science Publishers, New York, 1986), p. 265.

[3] P. Esquinazi and R. König, in *Tunneling Systems in Amorphous and Crystalline Solids*, edited by P. Esquinazi (Springer-Verlag, Berlin, 1998).

[4] D. G. Cahill and R. O. Pohl, Phys. Rev. B **39**, 10 477 (1989).

[5] P. A. Medwick and R. O. Pohl, J. Solid State Chem. **133**, 44 (1997).

[6] G. A. Slack, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1979), Vol. 34, p. 57.

[7] J. E. Graebner and H. S. Chen, Phys. Rev. Lett. **58**, 1945 (1987).

[8] J. J. Freeman, K. J. Dahlhauser, A. C. Anderson, and S. J. Poon, Phys. Rev. B **35**, 2451 (1987).

[9] E. Thompson, Ph.D. thesis, Cornell University, 2000.

[10] P. D. Vu, Xiao Liu, and R. O. Pohl (unpublished).

[11] N. O. Birge *et al.*, Phys. Rev. B **36**, 7685 (1987).

[12] A. K. Raychaudhuri and S. Hunklinger, Z. Phys. B: Condens. Matter **57**, 113 (1984).

[13] S. Grondey, H. v. Lohneysen, H. J. Schink, and K. Samwer, Z. Phys. B: Condens. Matter **51**, 287 (1983).

[14] J. E. VanCleve, N. A. Gershenfeld, K. Knorr, and P. A. Bancel, Phys. Rev. B **41**, 980 (1990).

[15] M. A. Chernikov, A. Bianchi, and H. R. Ott, Phys. Rev. B **51**, 153 (1995).

[16] S. Legault, B. Ellman, J. Strom-Olsen, and L. Tailleter, in *Quasicrystals, Proceedings of the 5th International Conference, Avignon, France 1995*, edited by J. Janot (World Scientific, Singapore, 1996), p. 592.

[17] M. A. Chernikov *et al.*, in *Quasicrystals, Proceedings of the 6th International Conference, Tokyo, Japan 1998* (World Scientific, Singapore, 1998), p. 451.

[18] M. A. Chernikov *et al.*, Europhys. Lett. **35**, 431 (1996).

[19] N. Vernier *et al.*, Europhys. Lett. **22**, 187 (1993).

[20] P. Esquinazi and J. Luzuriaga, Phys. Rev. B **37**, 7819 (1988).

[21] J. R. Matey and A. C. Anderson, J. Non-Cryst. Solids **23**, 129 (1977).

[22] G. P. Johari, Phys. Rev. B **33**, 7201 (1986).

[23] F. Bert, G. Bellessa, A. Quivy, and Y. Calvayrac, Phys. Rev. B **61**, 32 (2000).

[24] K. Topp, E. Thompson, and R. O. Pohl, Phys. Rev. B **60**, 898 (1999).

[25] E. T. Swartz, Rev. Sci. Instrum. **57**, 2848 (1986).

[26] P. D. Vu, J. R. Olson, and R. O. Pohl, J. Low Temp. Phys. **113**, 123 (1998).

[27] M. A. Chernikov, A. Bernasconi, C. Beeli, and H. R. Ott, Europhys. Lett. **21**, 767 (1993).

[28] D. N. Basov *et al.*, in *Quasicrystals, Proceedings of the 5th International Conference, Avignon, France, 1995* (Ref. 16), p. 564.

[29] A. L. Pope, T. M. Tritt, M. A. Chernikov, and M. Feuerbacher, Appl. Phys. Lett. **75**, 1854 (1999).

[30] D. M. Herlach, H. W. Gronert, and E. F. Wassermann, Europhys. Lett. **1**, 23 (1986).

[31] R. O. Pohl, in *Encyclopedia of Applied Physics* (Wiley-VCH, New York, 1998), Chap. 23, p. 223.

[32] D. M. Herlach, H. W. Gronert, E. F. Wassermann, and W. Sander, Z. Phys. B: Condens. Matter **65**, 353 (1987).

[33] D. G. Cahill, S. K. Watson, and R. O. Pohl, Phys. Rev. B **46**, 6131 (1992).

[34] Y. Amazit, M. De Boissieu, and A. Zarembowitch, Europhys. Lett. **20**, 703 (1992).

[35] K. A. Topp and D. G. Cahill, Z. Phys. B: Condens. Matter **101**, 235 (1996).

[36] M. A. Chernikov *et al.*, Phys. Rev. B **48**, 3058 (1993).

[37] C. Walti *et al.*, Phys. Rev. B **57**, 10 504 (1998).

[38] R. O. Pohl, in *Topics in Current Physics*, edited by W. A. Phillips (Springer, Berlin, 1981), p. 27.

[39] M. Feuerbacher, M. Weller, J. Diehl, and K. Urban, Philos. Mag. Lett. **74**, 81 (1996).

[40] E. Thompson and R. O. Pohl, in *Polycrystalline Metal and Magnetic Thin Films*, edited by K. P. Rodbell, D. Laughlin, O. Thomas, and B. Zhang, MRS Symposia Proceedings No. 562 (Materials Research Society, Pittsburgh, 1999), p. 183.

[41] U. Messerschmidt *et al.*, Philos. Mag. A **79**, 2123 (1999).

[42] P. A. Medwick, B. E. White, Jr., and R. O. Pohl, J. Alloys Compd. **270**, 1 (1998).

[43] X. Liu *et al.*, Phys. Rev. Lett. **81**, 3171 (1998).

[44] F. J. Walker and A. C. Anderson, Phys. Rev. B **29**, 5881 (1984).

[45] R. B. Stephens, Phys. Rev. B **13**, 852 (1976).

[46] A. K. Raychaudhuri and R. O. Pohl, Phys. Rev. B **44**, 12 233 (1991).

[47] G. Mattausch *et al.*, Phase Transit. **59**, 189 (1996).

[48] P. J. Anthony and A. C. Anderson, Phys. Rev. B **14**, 5198 (1976).

[49] P. Doussineau *et al.*, J. Phys. (Paris) **41**, 1193 (1980).