Model for the thermal conductivity of a quasicrystalline alloy

A. Jagannathan

Laboratoire de Physique des Solides, Université Paris–Sud, 91405 Orsay, France

Abstract

We present a simple model for the observed temperature dependence of the thermal conductivity of AlPdMn (Chernikov et al, Phys.Rev.B vol.51, (1995),153). We account for the low temperature data, upto and including the plateau of the thermal conductivity, and discuss the similarities and the differences between the quasicrystal and amorphous solids.

63.50, 65.90, 66.70
A recent paper by Chernikov, Bianchi and Ott\(^1\) presents thermal conductivity data for the quasicrystal AlPdMn. The data are obtained for excellent quality samples of Al\(_{70}\)Pd\(_{21}\)Mn\(_9\), containing very few phason defects and with a structural coherence length of almost 10\(^4\)\(\AA\). The data are interesting for their similarity as well as their differences from the thermal conductivity of amorphous substances. We wish here to present a model with which to interpret the experimental observations.

As is well-known, amorphous materials show a remarkably universal behavior of the thermal conductivity \(\kappa(T)\) at low temperatures\(^2\). At the lowest temperatures, around 1K, it has a power law \(\kappa(T) \sim T^2\). Around 10K it reaches a “plateau” region where it takes on a roughly constant value. This behavior is termed universal, as it is observed in an extraordinarily diverse set of glassy materials – from polymer blends to vitreous silica to varnishes and resins. Chernikov et al have obtained a similar \(\kappa(T)\) curve for the icosahedral AlPdMn alloy: it follows a power law \(T^\alpha\) with \(\alpha \approx 2\) below 1.6K. It then rises until it reaches the plateau region between 25K and 55K. The plateau is thus attained at a higher temperature than is common in glasses, in addition, the value of \(\kappa\) in the plateau region is significantly higher as compared to the glasses.

The above observations have been made for temperatures that are around one-tenth of the Debye temperature, which as deduced from specific heat data is \(\theta_D = 362K\). We may thus safely assume that the phonons – or more cautiously speaking, the vibrational modes – responsible for thermal conduction are of large spatial extent compared to interatomic distances. At the very lowest temperatures, one may reasonably assume that phonons of long wavelength propagate in the quasicrystal, as they do in disordered media, with well-defined longitudinal and transverse sound velocities. For temperatures below a couple of Kelvin, thus, thermal transport takes place via these phonons, and \(\kappa\) will depend on the scattering of the phonons by the structure. The scattering mechanism the most frequently invoked in glasses is due to localized “tunneling systems” or TS\(^3\). In the AlPdMn quasicrystal, low temperature sound velocity data\(^4\) indicate the presence of tunneling systems. Quasicrystals may have, in fact, a natural candidate for TS in their so-called “phason” defect states.
These are local atomic displacements that preserve allowed bond angles and lengths. The quasicrystal thus could scatter phonons from these phason defects or from additional possible glassy-type TS.

In the quasicrystal, we picture the large majority of the eigenmodes of the perfect quasiperiodic solid to be not phononlike, but rather modes with a non-ballistic type of propagation. One cannot speak of a dispersion relation in this case, since there is no k-space, rather, we will speak of a mean square displacement of the mode in real space as a function of time given by $r^2(t) = \tilde{D}t^\alpha(\omega)$. Alternatively, one could write an equivalent relation for the variation of the group velocity as a function of energy (frequency). This dependence is empirically written, and it is based on the hypothesis that eigenmodes on quasiperiodic structures are in between free-particle like ballistic modes and localized modes. This hypothesis is primarily motivated by results of numerous studies of tight-binding electronic models in quasicrystals (reviewed in\textsuperscript{5}) where a similar intermediate notion exists between extended and localised electronic eigenfunctions – the critical wavefunctions. In consequence, electrons have a dynamics in-between that of free electrons and completely trapped ones. In addition to the indirect evidence for this coming from the unusual scaling properties of the energy spectrum, direct numerical evidence exists for anomalous diffusion of particles in the quasiperiodic medium\textsuperscript{6}. In the present context of the vibrational problem, Janssen and Quilichini\textsuperscript{7} have discussed numerical studies carried out for one, two and three dimensional models. The phonon spectra show the characteristic jagged shape and multifractal scaling. As for the eigenfunctions, a numerical calculation of the inverse participation ratio gives values that are intermediate between that of localized states and extended states. Although the values of $\alpha$ vary irregularly with $\omega$, one can argue that the physics will be mainly determined by the underlying smoothed out form of the spectrum. We will therefore make the simplifying assumption that one can work with a locally averaged value of $\alpha$ which will be allowed to vary smoothly with $\omega$ in the energy range of interest. It is reasonable to suppose that this averaged value in good quasicrystals is $2 > \alpha > 1$ (i.e. in the range given by the low energy phonon value of 2, down to possibly slower-than-normal-diffusion
values). Finally, the constant $\tilde{D}$ is an unknown parameter, to be eventually determined by comparison with experiment.

In a real sample, these modes will exist from a length scale given by the coherence length $\xi_{coh}$, down to atomic distances $a$. For the longer length scales, true phonons will be present. In terms of the energy $\omega$ (or temperature), small $\omega$ or low temperature corresponds to exciting majoritarily phonons, while higher temperatures will lead to the exciting of the quasilattice vibrational modes – we propose to term them “quasons” from here on. Our model supposes that the density of states is that of phonons at very low energies, crossing over to the quason form at an energy scale $\omega_\xi$ that we can estimate using the values $\xi_{coh}/a \approx 10^{-3}$ and $\theta_D=362$K. It corresponds to a crossover temperature of less than a Kelvin. One should note, however, that no abrupt changes should be expected at this crossover, since the low energy end of the quason spectrum is phononlike, with a density of states not much different from the phonon one, until one reaches somewhat higher temperatures. For the range of temperature of a few Kelvin and below, we can thus assume the density of states will be that of the phonons, while above the crossover energy one will have the density of states of the quasicrystal:

$$
(\omega < \omega_\xi) \quad N(\omega) = \frac{V}{(2\pi^2v_s^3)}\omega^2
$$

$$
(\omega > \omega_\xi) \quad N(\omega) = \frac{d_s V}{(a^3 \omega_D^{d_s})} \omega^{(d_s-1)}
$$

(1)

where $\omega_D = (6\pi^2)^{(1/3)}(\xi_{coh}/a)^{(3/d_s)}(v_s/\xi_{coh})$ taking for illustration the simplest case of a unique value of $d_s$ in the quason régime. The spectral dimension $d_s$ which is equal to 3 for phonons, is $3\alpha/2$ for the quasons, which results in the density of states in the quason-régime growing somewhat more slowly with $\omega$ than for phonons.

At the very lowest temperatures, then, we may apply the usual formula giving the phonon contribution to thermal conductivity,

$$
\kappa(T) = \frac{1}{3} \int d\omega \quad v(\omega)C(\omega)l(\omega)
$$

(2)

where $C$ is the specific heat of modes of energy $\omega$: $C(\omega) = N(\omega)\omega^2 e^{\beta\omega}/((k_B T)^2(e^{\beta\omega} - 1)^2)$. 


The mean free path $l(\omega)$ is determined by the scattering processes that enter in play. We now discuss the very low and intermediate temperature régimes, by turn.

At the lowest temperatures, phonons carry the thermal current and the TS referred to above give rise to $l \propto \omega^{-1}$. The specific heat in this limit is $C(\omega) \propto \omega^4 \exp(-\beta \omega)$. Setting the mode velocity $v(\omega)$ equal to the sound velocity, Eq.2 readily yields

$$\kappa(T) = AT^2$$  \hfill (3)

Chernikov et al have discussed the prefactor $A$ as well as the exponent of $T$ in the above law assuming a plausible set of values for the parameters entering in the explicit formula for $\kappa$. In fact, the interpretation of the data is more complex than the simple argument given above, and we refer the reader to the original paper for a detailed discussion of the behavior of $\kappa$ below 1.6K. We turn now to the expected temperature dependence of $\kappa$ at higher temperature.

At intermediate $T$, the relation in Eq.2 is replaced by an expression appropriate to the quason-dominated régime by

$$\kappa(T) = \frac{\bar{D}}{3} \int d\omega \ C(\omega)\tau^{n-1}(\omega)$$  \hfill (4)

where we have introduced a scattering time $\tau$ for the quason modes. This scattering could be due to anharmonic interactions between vibrational modes or scattering from defects in the structure. In glassy compounds Rayleigh scattering from defects is often supposed, giving rise to a frequency dependence $\tau(\omega) \sim \omega^{-4}$. In vitreous silica, Zeller and Pohl have used the dominant phonon approximation along with their thermal data to show a $\omega^{-4}$ dependence of the scattering time $\tau$. This strong scattering at higher frequencies is thought to be responsible for the failure of higher frequency phonons to propagate, as they attain the Ioffe-Regel limit of $l(\omega) \sim a$. This leads to the levelling off of the thermal conductivity at its “plateau” value.

In the quasicrystal alloy, lacking any direct experimental evidence as to $\tau(\omega)$, one can try to apply the equivalent of the dominant phonon approximation. In this approximation, the
thermal conductivity is taken to be entirely determined by the modes which have the highest heat capacity ($\omega_{\text{dom}}$ being given by the peak of $C(\omega)$ for a given temperature). Published data on specific heat gives a fit to a standard form for this quantity at low temperature. Assuming a lattice contribution that is similar to that in a glass and given the thermal conductivity data, we conclude that the scattering time $\tau$ has a similar $\omega$ dependence in the AlPdMn as in the glass system. This qualitative statement obviously needs further investigation. Although neutron scattering studies do see a significant broadening effect away from the acoustic limit, as we discuss further below. This could imply that strong Rayleigh-type scattering is found in the quasicrystal, just as in the amorphous case. The origin of this scattering could be related to inhomogeneities of the structure as in the glass. However in view of the good structural quality of the quasicrystalline sample this seems an unsatisfactory explanation. A second and interesting possibility is that this scattering lifetime is an artifact arising from our assumption that the density of states is a smoothly varying and increasing function of $\omega$ in this energy range. In fact, it has been observed in all the models studied that there are dips, or pseudogaps, in the density of states with some important ones occurring as one approaches the middle of the spectrum. If such a dip occurs at the energies just below the plateau energy, then one may attribute the dependence of $\kappa$ in this region rather to a density of states effect than to an increased scattering rate effect. Neutron scattering provides no evidence for such pseudogaps, although one should note that we are speaking here of longer length scales (several dozen Å). There is evidence from neutron scattering for a significant broadening of modes away from the acoustic limit. This favors the interpretation in terms of a lifetime effect. In any case, we will proceed on the hypothesis that it is the lifetime which is strongly energy-dependent. The alternative scenario can be easily handled by a similar type of calculation if necessary.

As in the amorphous case, the strong frequency dependence of $\tau$ then leads to the “falling off” of $\kappa(T)$ above a certain temperature. Thus the plateau appears for the quasicrystal, and for the same reason as in the glasses. The quantitative differences are important, as they arise from the fundamental difference in the dynamics of the heat carrying modes in these
two systems. This is now discussed. In this simple-minded model given by Eq.4 the thermal conductivity of quasicrystal and glass differ significantly in that the scattering lifetime enters with an exposant $\alpha - 1$ which is in all cases smaller than one (which is the exponent for the glassy case). If one takes the Rayleigh scattering $\tau$, then

$$\kappa(T) \sim \int d\omega \; C(\omega)\omega^{-(\alpha-1)}$$

(5)

Since the dominant frequency dependence is contained in the last factor, which decreases less rapidly than for the usual phonon case, the quason thermal conductivity is less strongly attenuated by the Rayleigh scattering. This implies that $\kappa$ will be slower in reaching its plateau value, and explains the observed shift of the plateau region to a higher temperature in AlPdMn. To make a quantitative estimate of the shift expected one would require further information on the prefactors entering in the scattering lifetime.

A related point concerns the relative magnitude of $\kappa$ in the glassy and quasicrystalline systems. Chernikov et al have noted that when data are plotted using variables scaled by the measured values of the Debye temperature and sound velocity, $\tilde{\kappa}(\tilde{T})$ in the glass and the quasicrystal are very similar upto the glass plateau, beyond which the quasicrystal $\tilde{\kappa}$ continues to rise to a higher value. This could be understood as follows: in terms of appropriately scaled quantities the thermal capacity of phonons and phasons are roughly equal – this is indicated by the similar values of $\tilde{\kappa}(T)$ below the plateau for glass and quasicrystal, This would then imply since modes are slightly less strongly attenuated in the quasicrystal, that $\tilde{\kappa}$ continues to rise in the quasicrystal, attaining a plateau value that is higher than for the glass. This is borne out by a little model calculation (see figure) where we have integrated Eqs.4 and 2 numerically and compared the results for $\kappa(T)$. The scattering time is taken to be identical for the two cases, namely an interpolation between a constant value (at low frequency) and $\omega^{-4}$ (at higher frequency). The density of states is pure phonon for the lower curve; for the upper curve, there is a crossover from phonon to quason density of states (with $\alpha = 3/2$).

In conclusion, we note that this discussion applies to the temperature régime $T < 50K$. 
The experimental data indicates an upturn of $\kappa$ at the upper end of this range. As in the glasses, this increase of the thermal conductivity could be explained by at least two scenarios. One is a scenario in which the Rayleigh-scattering is somehow “turned off” at higher frequencies, so that modes can propagate anew in the sample. The other would be a new type of heat conduction in which localized vibrational modes begin to hop, aided by anharmonic interactions with ordinary phonons. However this high temperature régime is outside the scope of our present paper.
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FIGURE CAPTION

Results for $\kappa$ (arbitrary units) vs $T$ from simple model calculation (see text). The upper curve corresponds to the quasicrystal, the lower curve to a glass having the same Debye temperature and scattering rate $\tau(\omega)$.
