Structural scattering of phonons in quasicrystals

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The concept of phonon scattering known as the umklapp process is generalized to quasicrystals. It is shown that such processes in quasicrystals lead to a power-law temperature dependence of the mean free path of delocalized quasilattice excitations, contrary to the exponential temperature dependence of the phonon mean free path characteristic of periodically structured crystals at intermediate temperatures. This result provides a plausible explanation for the plateau-type feature in the quasilattice thermal conductivity $\lambda_{ph}(T)$ of Al-Mn-Pd quasicrystals. [S0163-1829(96)01921-2]

I. INTRODUCTION

One of the remarkable features of quasicrystals is the apparent conflict between the high structure quality of these materials and their transport properties, which are rather reminiscent of those of highly disordered materials, such as metallic or insulating glasses. The structure quality of quasicrystals, as revealed by x-ray structure analysis or electron-microscope investigations, sets them among the best ordinary crystals. For example, high-resolution x-ray diffraction experiments performed on Al-Mn-Pd quasicrystals revealed a mosaic full width at half maximum of less than 0.001° and resolution-limited widths of the diffraction peaks.[1] At the same time, a high degree of structural perfection of icosahedral Al-Mn-Pd has been confirmed by anomalous transmission of x rays through this material.[1] On the other hand, both the electrical and thermal conductivities of quasicrystals are much lower than in periodic metallic crystals. Many transport properties, namely, the temperature and magnetic-field dependencies of the electrical and the thermal conductivity, reveal features that are commonly observed in glassy materials. Nevertheless, it seems *a priori* unjustified to claim that the transport properties of quasicrystals are influenced by the same mechanisms as those of metallic glasses. This is mainly because the structures of quasicrystals, known in great detail, differ from the structure of glasses in distinct ways.

When speaking about phonon (or electron) scattering in quasicrystals one should clearly distinguish between the scattering on defects and the structural scattering *stricto senso*. Defects are always present in quasicrystals. In view of a general lack of ''growth rules'' for quasicrystals[2] it is impossible to grow a quasicrystal with a density of defects lower than some intrinsic value. It has been suggested that at very low temperatures the temperature dependence of the thermal conductivity is most likely due to the resonant scattering of phonons on pointlike defects (tunneling states).[3] In spite of this problem and because of the fact that the structure quality of many quasicrystals may be improved by annealing procedures, it seems reasonable to investigate the transport properties of hypothetical perfect quasicrystals.

Here we shall concentrate on the scattering mechanisms which are intrinsic to the structure of quasicrystals, i.e., those that would be *present even in a hypothetical defect-free material*. Because the problems of both single-electron eigenstates and phonon eigenmodes lead to similar equations, it is worth looking first at the results obtained in the study of the behavior of noninteracting electrons in a quasicrystal. Although this problem has been extensively investigated since the very discovery of quasicrystals, there exist no reliable results in the literature, except for the one-dimensional case, for which one can solve the problem exactly.[4,5] In the two- and three-dimensional cases one can merely show that the hypothesis of the existence of quasi-Bloch states is not allowed for quasicrystals. In particular, if one tries the following ansatz for the electron eigenfunction:

$$\Psi_{\mathbf{k}}(\mathbf{r}) = \sum_n c_n \exp\left[i\left(\mathbf{k} - \sum_{j=1}^{D} n_j \mathbf{e}_j \right) \mathbf{r} \right], \tag{1}$$

where the vectors $\mathbf{e}_j$ are the basis vectors of the reciprocal lattice of the quasicrystal and D is the indexing dimension ($D=6$ for icosahedral quasicrystals), which is a straightforward generalization of the Bloch wave-function form, one immediately realizes[6] that this function cannot be normalized while keeping the coefficients c_n finite. Physically this means that while the true Bloch states in common crystals are localized in momentum space and thus resemble states occupied by free particles, the eigenstates in quasicrystals are never localized in k space. These eigenstates are always affected by an intrinsic decay rate, thus giving rise to nonzero resistivity even of perfect quasicrystals.[6] It is important to note, however, that this intrinsic decay rate is exponentially low in the limit of weak potentials. This property has important consequences when we consider the phonon propagation in quasicrystals instead of extended electronic eigenstates.

Although the equation of the phonon eigenmodes and the Schrödinger equation for one-electron states are very similar, there is one important difference. While the scale of momenta of occupied electron states is given by k_F and therefore cannot be chosen arbitrarily, in the case of phonons

there is always the long-wavelength limit, where details of the structure do not play a significant role. This limit thus corresponds to the vanishing influence of the quasiperiodicity of the potential, and that this is really the case we shall demonstrate for the one-dimensional system.

II. PHONONS IN A ONE-DIMENSIONAL QUASICRYSTAL

As has already been mentioned, for the one-dimensional model of quasicrystals (Fibonacci chain) an exact solution of the one-particle Schrödinger equation and consequently of the acoustic eigenmode problem has been found.[6,7] This solution emerges from using the very powerful transfer-matrix formalism. Transfer matrices relate two independent solutions of the Schrödinger equation, say Ψ and Ψ', or displacements of two neighboring atoms in the case of acoustic modes. The main result of the corresponding theory is that the problem is almost universal, namely, there is only one parameter whose value determines different universality classes.[5,8] The standard choice of this parameter is the half trace of the multiplicative commutator of the transfer matrices corresponding to the distinct sites of the Fibonacci chain. This parameter is never smaller than 1 and the case when it equals 1 corresponds to the universality class appropriate for the periodic chain. As an example we give the explicit expression for this parameter in the case of acoustic oscillations. We suppose that the Fibonacci chain is made of atoms with two different masses, but the elastic forces between different atomic species are the same. Then the transfer matrix relating the displacement vector $\mathbf{u}_n=(u_{n-1},u_n)$, where u_i is the oscillation amplitude of a given atom i, with the displacement vector $\mathbf{u}_{n+1}=(u_n,u_{n+1})$ for the mode with the given frequency ω is

$$T_n=\begin{pmatrix} 0 & 1 \\ -1 & 2-m_n k^{-1}\omega^2 \end{pmatrix}. \tag{2}$$

Here m_n stands for the mass of the atom on site n, and k is the spring constant. Up to the leading term, the commutator of the transfer matrices on two neighboring sites 1 and 2 reads

$$J=\frac{1}{2}\,\mathrm{Tr}(T_1T_2T_1^{-1}T_2^{-1})=1+\frac{(m_1-m_2)^2}{2k^2}\,\omega^4. \tag{3}$$

Of particular interest in this equation is the ω dependence of J. First, as expected, J approaches 1 as the frequency decreases. This reflects the fact that the short-range details of the quasiperiodic structure become less important in the long-wavelength limit. From the exact solution for the Fibonacci chain it is known that the widths of major gaps in the spectrum behave as $\sqrt{J-1}$, or as ω^2 in the case considered.[5,8] Although the spectrum has zero measure as is the case for the one-electron Schrödinger equation on the Fibonacci chain, in the $\omega\to 0$ limit the spectrum appears as continuous because the widths of gaps become smaller than the separation of the eigenmodes of a chain of finite length. In Fig. 1 we plot the frequencies of the eigenmodes of a Fibonacci chain consisting of 233 atoms with two different masses as a function of the pseudo-wave vector q. Instead of a wave vector, which strictly is an undefined quantity here because of the instability of quasi-Bloch states, the density of nodes of the eigenmode is used. This may be taken as a natural generalization of wave vectors in the case of a quasiperiodic chain because the density of zeros is proportional to the wave vector in the case of a periodic chain. In the $\omega(q)$ plot of Fig. 1 only major gaps are visible.

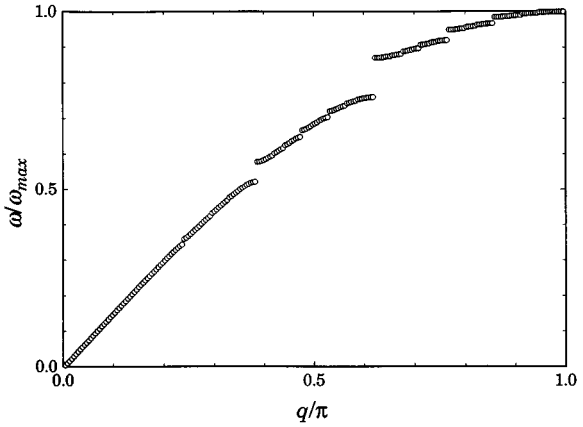

FIG. 1. Frequencies of phonon eigenmodes of a Fibonacci chain composed of 233 atoms with two different masses. The mass ratio is 1.5.

III. GENERALIZATION TO THREE-DIMENSIONAL QUASICRYSTALS

For comparisons with experiments on real quasicrystals, we have now to consider two- and three-dimensional systems. First, it is worth emphasizing that the ω^2 scaling of major gaps in the acoustic spectrum may be understood without recourse to the exact solution of the problem. The width $\delta\omega$ of the gap in the vibrational excitation spectrum formed by a density wave with a wave vector q and a relative amplitude a is of the order of av_sq, where v_s is the sound velocity. For quasiperiodic lattices there is a series of strongest harmonics in the Fourier image of the density for which the absolute value of the product $q_nq_{\perp n}$, where $q_{\perp n}$ is the component of the corresponding wave vector in the orthogonal space, reaches its minimal value and is constant, i.e., it does not depend on n. The series of the wave vectors q_n of the strongest density harmonics is a geometrical series

$$q_n\propto C^n, \tag{4}$$

with a common ratio C, i.e., $C=\tau^{-1}$, where $\tau=(\sqrt{5}+1)/2$ is the golden mean, for a Fibonacci chain as well as for face- and body-centered icosahedral lattices and $C=\tau^{-3}$ for a simple cubic icosahedral lattice.[9] The amplitudes of these harmonics scale like q, which in the limit of small q leads to the expected ω^2 scaling of the gap widths.

If at this point we assume that the use of wave vectors for describing lattice eigenmodes in quasicrystals is legitimate, then the same arguments may be applied to two- and three-dimensional quasilattices. The strongest harmonics in the Fourier image of the density are due to the faces of the ''atomic surfaces,'' used in the structure description of the considered quasicrystal. The wave-vector dependence of the amplitudes of these harmonics is determined by the structure factor. For calculating the structure factor we need the Fou-

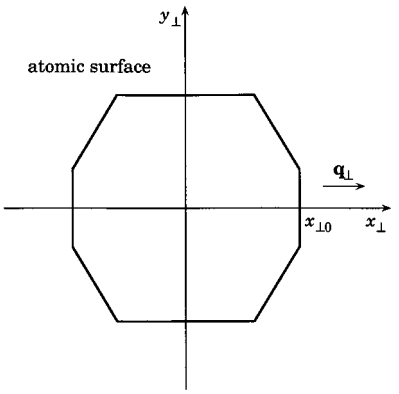

FIG. 2. Schematic representation of an atomic surface with one of its faces perpendicular to $\mathbf{q}_\perp$. The step in the density function occurs at $x_{\perp 0}$. Shown here are only two components in the orthogonal space.

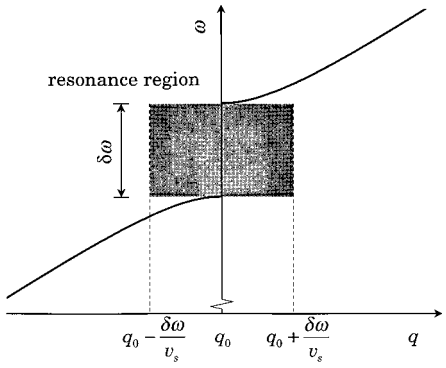

FIG. 3. Schematic representation of a gaplet in the frequency spectrum of lattice excitations in a periodic structure induced by a structure modulation with wave vector q_0.

rier transform of the step function, which is constant inside the atomic surface and zero outside. The behavior of this Fourier transform for large $q_\perp$ is dominated by the effect of the flat faces of the atomic surface (see Fig. 2). For the wave vectors $\mathbf{q}_\perp$ perpendicular to such faces, the Fourier transform scales like $q_\perp^{-1}$ and hence, for harmonics from the major series, as q. Thus the same arguments as for the one-dimensional case apply and one may expect again the same ω^2 scaling of the major gap widths.

We now return to the conflict that we avoided so far. Although we used the notion of wave vectors when speaking about gaps in the spectrum it should be kept in mind that the oscillatory eigenmodes in quasicrystals cannot be described *stricto senso* by this parameter. We note, however, that the intrinsic decay rate of the quasi-Bloch states in quasicrystals varies exponentially with the strength of the potential. Therefore, in the long-wavelength limit, the decay process is so slow that other decay mechanisms will certainly dominate over the intrinsic decay. This is the important distinction between the two problems of defining either electronic or acoustic eigenstates, because the former cannot be treated in such a long-wavelength limit.

In what follows, we shall consider the region in frequencies where the intrinsic decay rate of oscillatory quasi-Bloch states is insignificant. The phonons will never have enough time to get completely delocalized in momentum space; instead, they are scattered by other mechanisms. We shall consider, as an example, the phonon-phonon scattering due to the anharmonicity of the lattice oscillations, although other mechanisms like, e.g., scattering on electrons are not excluded.

IV. UMKLAPP PROCESSES IN QUASICRYSTALS

The phonon scattering mechanism in quasicrystals to be proposed below may seem exotic, but it is merely a generalization of umklapp processes in the usual crystals.[10] We emphasize that the scattering phenomenon known as the umklapp process is a consequence of the interplay between two scattering processes. In one of these processes, e.g., in multiphonon scattering, the momentum of lattice excitations, often called crystal momentum, is conserved. The other process, i.e., a Bragg reflection or structural scattering, violates the law of conservation of crystal momentum. The characteristic time of energy transfer to the reflected acoustic wave in this process may be estimated as $\hbar/E_g$, where E_g is the energy gap in the vibrational excitation spectrum. In periodic crystals E_g is often of the order of $\hbar\omega_{\max}$,[11] where $\omega_{\max}$ is the maximum lattice frequency, and the Bragg reflection occurs very fast. Nevertheless, for periodic crystals the important point is that neither of the two scattering processes, if considered separately, will give rise to the same physical consequences as the umklapp process does, i.e., finite thermal conductivity, etc. This is all the more true for quasicrystals where one expects a hierarchy of gaps with widths at any scale of energy less than $\hbar\omega_{\max}$, and therefore the Bragg reflections have to be considered as separate scattering processes. In addition, in quasicrystals the momentum of vibrational excitations can be transferred to the quasilattice in small portions, not limited from below in magnitude. We recall that the natural scale for umklapp processes in periodic crystals is set by the reciprocal lattice, which does not exist for quasicrystals. This also means that while in crystals the rate of umklapp processes decreases exponentially at small frequencies (temperatures), in quasicrystals it should obey some power law. As we shall see, this power-law behavior is indeed indicated by experiment.

As a first step in introducing the generalized umklapp process in quasicrystals, one may consider some intermediate case, namely, structurally modulated crystals. It is intuitively evident that although the size of the Brillouin zone changes drastically at the very beginning of superstructure formation, the rate of scattering at the superstructure wave vectors increases only gradually because the scattering probability depends on the amplitude of the modulation. To discuss this important point in more detail, we consider a region in the phonon spectrum where the density or, more precisely, acoustic-impedance wave with a wave vector q_0 has led to the formation of a gaplet at $q=q_0$ (see Fig. 3).

Qualitatively, only the modes with wave vectors close to q_0 are affected by the structural scattering. More precisely, only in the modes with $|q-q_0|<\delta\omega/v_s$ is there a significant fraction of backscattered waves. This gives the formal justification for the intuitive assumption that density modulations with small amplitudes do not much affect the umklapp processes. In the one-dimensional (1D) case only those phonons

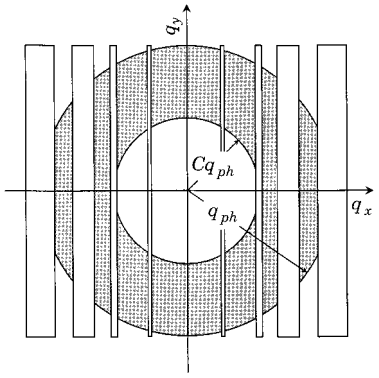

FIG. 4. The regions of resonant scattering due to the major series of density harmonics overlapping the phonon wave-vector spectrum in the shell between $Cq_{\rm ph}$ and $q_{\rm ph}$.

which are newly created in a multiphonon process and whose wave vectors are in the resonance region $|q-q_0|<\delta\omega/v_s$ are efficiently backscattered; in higher dimensions they are merely scattered by the superstructure wave vector q_0.

The situation is more complicated for quasicrystals. Instead of just one harmonic describing the density modulation as in the case of a periodic superstructure, we now have to deal with infinitely many of them. Their wave vectors densely fill the reciprocal space so that no clear distinction between resonance regions seems to be possible. As we have pointed out above, there is a major series of density harmonics described by Eq. (4), set apart from all the other harmonics by their strength. The harmonics from the major series are due to the faces of the atomic surfaces shown in Fig. 2 and, as we have outlined above, at small wave vectors the amplitudes of these harmonics scale as q. The series of their wave vectors is a geometrical series. Therefore in the spherical layer between $Cq_{\rm ph}$ and $q_{\rm ph}$

$$Cq_{\rm ph}<q<q_{\rm ph}, \tag{5}$$

where C is the common ratio of the geometrical series [see Eq. (4)], there is always a constant number of these harmonics as expected from self-similarity arguments.[12] In the following discussion we assume that no other harmonics than those from the major series have to be considered, an assumption that will be justified below.

As in the case of superstructures, we have to estimate the probability of a *newly created phonon* to be scattered by the structure. Suppose that the absolute value of the wave vector q of this phonon lies in the spherical shell described by inequality (5). We then have to estimate the total area (or volume) of the resonance regions in the reciprocal space as shown in Fig. 4. From the ω^2 scaling of the major-series gap widths it is clear that this probability P is proportional to ω, or more precisely, $P=A\omega/\omega_{\rm max}$, where A is a dimensionless constant depending on the structure of the quasicrystal. Instead of the maximum lattice frequency $\omega_{\rm max}$ one may also introduce the Debye frequency ω_D, which is close to $\omega_{\rm max}$. We briefly return to the problem of tiny gaplets not belonging to the major series and to their influence on the structure scattering. This question is closely related to the instability of the quasi-Bloch states in quasicrystals. Indeed, the instability of quasi-Bloch states is due to the fact that the number of states which are in resonance with any given one is infinite. The divergence of this number, however, is only logarithmic.[6] This means that this divergence is entirely due to the harmonics with tiny amplitudes. Therefore the same argument which we have used above to justify the irrelevance of the intrinsic instability of quasi-Bloch states may be applied to justify the irrelevance of harmonics not belonging to the major series with respect to structural scattering. Indeed, the scattering of an acoustic wave on a density modulation is not instantaneous. For the particular modulation giving rise to a gap in the acoustic spectrum of width $\delta\omega$ (see Fig. 3), the time of energy transfer to the scattered wave is of the order of $1/\delta\omega$. Thus, when the scattering is dominated by a fast nonstructural mechanism as provided, for instance, by anharmonicities, there is no time for an effective influence of the resonances that are due to the small harmonics in the structure factor.

V. THERMAL CONDUCTIVITY IN QUASICRYSTALS AT ELEVATED TEMPERATURES

We are now ready to estimate the physical consequences of the structure scattering in quasicrystals. As an example we consider the ''quasiperiodic'' umklapp processes which involve both phonon-phonon scattering and Bragg reflections. Suppose that three-phonon processes are not prohibited by the conservation laws. Then the frequency dependence of the transport cross section averaged over the phonon wave vectors within a spherical shell as defined in inequality (5) is given by

$$\langle\sigma_{\rm transp}\rangle=A\,\frac{\omega}{\omega_{\rm max}}\,\sigma, \tag{6}$$

where σ represents the full phonon-phonon scattering cross section. This formula does not depend on the particular choice of the nonstructural scattering mechanism that is responsible for the value of σ. For three-phonon scattering the full cross section is proportional to ωT^4, because the standard three-phonon matrix element is proportional to $\sqrt{\omega_1\omega_2\omega_3}$ and the phase volume rises as T^2 (six free components minus four conservation laws). Here it is supposed that only one phonon in the three-phonon process has a low frequency and that the two others are merely thermal ones with their frequency proportional to the temperature. All in all, the average transport cross section for the anharmonic and quasiperiodic structural scattering, together denoted as the quasiperiodic umklapp process, is proportional to ω^2T^4, i.e., not exponential in frequency or temperature as is valid for umklapp processes in common crystals. In the limit of low temperatures and assuming that quasiperiodic umklapp processes are the main source for the quasilattice thermal resistance $\lambda_{\rm ph}^{-1}(T)$, a Debye-type approximation leads to the expectation that $\lambda_{\rm ph}\propto T^{-3}$.

VI. COMPARISON WITH EXPERIMENT

We now discuss possible implications of the existence of quasiperiodic umklapp processes on the interpretation of experimentally determined temperature dependencies of the thermal conductivity of quasicrystals. Even in periodic crystals it is not trivial to experimentally verify the regime where

$\lambda_{ph}(T)$ is dominated by umklapp processes by observing

$$\lambda_{ph}(T) \propto T^{\xi} \exp\left(\frac{\Theta_D}{bT}\right), \tag{7}$$

where ξ and b both are of the order of unity. For a clear observation of this experimental relation the crystal needs to be of high structural quality and isotopically pure. The relevant temperature range is limited to approximately $0.03\Theta_D < T < 0.1\Theta_D$.[13] A crossover to the Casimir regime of boundary-limited thermal conductivity at the lowest temperatures leads to a maximum in the $\lambda_{ph}(T)$ dependence. Defects and isotope inhomogeneities suppress this maximum and they alter the exponential relation for $\lambda_{ph}(T)$. Even in polycrystalline and impure samples a distinct maximum of $\lambda_{ph}(T)$ is still observed, however.

As argued at the end of Sec. V, in the equivalent regime for quasicrystals, dominated by quasiperiodic umklapp processes, the thermal conductivity is expected to vary as $\lambda_{ph} \propto T^{-3}$, i.e., the variation with T is substantially weaker than in periodic crystals. At low temperatures, i.e., in the range of 1 K, $\lambda_{ph}(T)$ of real quasicrystals is limited by phonon scattering involving tunneling states and it varies approximately as T^2.[3] This scattering mechanism typically reduces $\lambda_{ph}(T)$ to values an order of magnitude below the Casimir limit. Above a few tens of degrees kelvin, quasilattice vibrational modes responsible for the heat transport cannot be described as propagating collective excitations[14] and λ_{ph} slowly increases with increasing temperature in this regime.[3,14,15]

As may be concluded from the discussion in the previous sections, quasiperiodic umklapp processes in real quasicrystals are expected to manifest themselves as a temperature region with a negative slope $d\lambda_{ph}/dT$. However, the $\lambda_{ph}(T)$ variation in this region may be substantially weaker than $\lambda_{ph} \propto T^{-3}$ or it may even be reduced to an extended plateau. Below we describe and analyze in more detail the results of $\lambda_{ph}(T)$ measurements of a bulk sample of icosahedral $Al_{70}Mn_9Pd_{21}$. Among thermodynamically stable quasicrystals, icosahedral Al-Mn-Pd is a quasiperiodically ordered material of the highest structural quality.[1,16,17] Some of these results, including details of sample preparation, experimental techniques, and low-temperature $\lambda_{ph}(T)$ data, has been given in a previous publication.[3]

In Fig. 5 the quasilattice thermal conductivity λ_{ph} of icosahedral $Al_{70}Mn_9Pd_{21}$ is shown on logarithmic scales for the temperature range between 0.06 and 297 K. Its evaluation from the measured thermal conductivity is described in Ref. 3. Below 1.6 K the temperature variation of λ_{ph} is well described by $\lambda_{ph} \propto T^{2.06}$. This variation is compatible with a dominant scattering of phonons by tunneling states, and our values of λ_{ph} are of the same order of magnitude as those reported for insulating and metallic amorphous solids. A saturation of the $\lambda_{ph}(T)$ dependence is evident above approximately 10 K. From 25 to 70 K, λ_{ph} is almost temperature independent but increases again with T above 70 K. Above 100 K, the magnitude of $\lambda_{ph}(T)$ is close to the prediction of Einstein's model,[18] suggesting that the lattice vibrations in the THz frequency range are predominantly localized, and the energy transfer between them occurs via a strong-coupling mechanism.[14] Localization of the high-

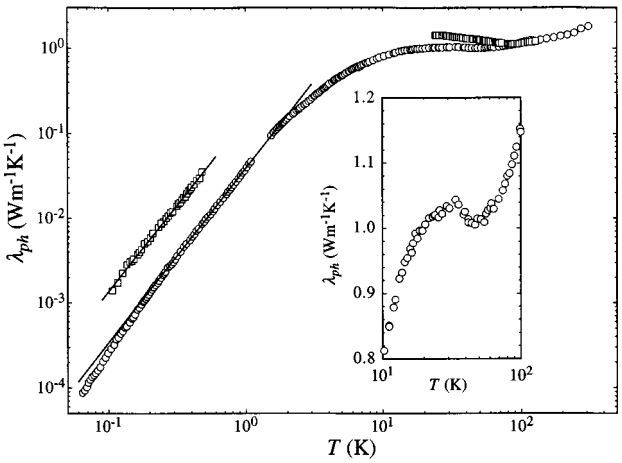

FIG. 5. Quasilattice thermal conductivity λ_{ph} of $Al_{70}Mn_9Pd_{21}$ (open circles, Refs. 3 and 14) and $Al_{70}Mn_{10}Pd_{20}$ (open squares, taken from Ref. 24) as a function of temperature T. The solid line is a power-law approximation to the data between 0.35 and 1.6 K. The inset shows $\lambda_{ph}(T)$ of $Al_{70}Mn_9Pd_{21}$ between 10 and 100 K on an expanded vertical scale.

frequency vibrational modes in Al-Mn-Pd quasicrystals has previously been claimed by de Boissieu and co-workers[16] by analyzing the results of inelastic neutron scattering experiments. They noted that true unbroadened acoustic modes are observed only for wave vectors $q \leqslant q_{max}$, where $q_{max} = 0.35$ Å^{-1}.

This overall behavior is similar to that of amorphous solids, where generally λ_{ph} monotonically increases with increasing temperature and a λ plateau is observed between approximately 2 and 10 K.[19] Nevertheless, we note some important differences in the plateau region. The λ plateau of icosahedral $Al_{70}Mn_9Pd_{21}$ is developed at higher temperatures and it extends over a wider temperature range than in amorphous solids. Moreover, λ_{ph} of the $Al_{70}Mn_9Pd_{21}$ quasicrystal in the λ plateau range is higher than that of amorphous solids in the corresponding regions.[3]

The $\lambda_{ph}(T)$ variation of amorphous solids has often been described assuming a frequency-dependent phonon mean free path $l_{ph}(\omega)$ which does not explicitly depend on temperature.[19] At low temperatures, scattering of phonons on tunneling states with $l_{ph} \propto \omega^{-1}$ gives the main contribution to the thermal resistance.[20,21] The plateau in the $\lambda_{ph}(T)$ data of amorphous solids has been claimed to be caused by an abrupt decrease of the phonon mean free path $l_{ph}(\omega)$ with increasing phonon frequency ω.[19] The available experimental evidence indicates that the mean free path of phonons in amorphous materials has a frequency dependence $l_{ph} \propto \omega^{-n}$, with $n \approx 3-4$.[22] The explicit relation $l_{ph} \propto \omega^{-4}$ does account for the λ plateau in many amorphous solids. It was argued, however, that this frequency dependence may not arise from the scattering on mass density fluctuations with a correlation length smaller than the phonon mean free path, i.e., Rayleigh scattering.[19] Two assumptions are in use as to how the mean free path of vibrational excitations in amorphous solids should be described at high temperatures. Kittel[23] has suggested that in this regime l_{ph} is a constant of the order of an average interatomic distance. Alternatively, Einstein's model of strongly coupled

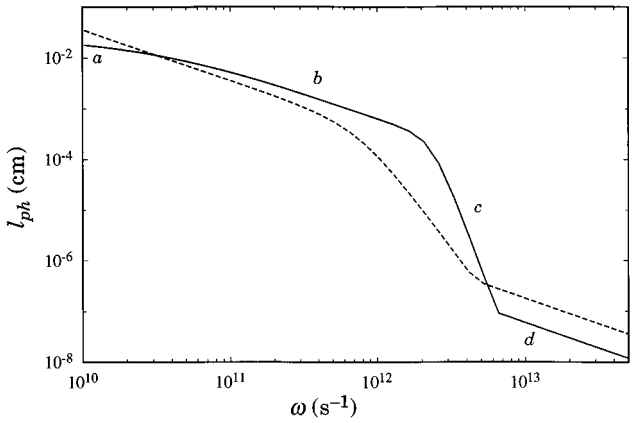

FIG. 6. Frequency dependence of the phonon mean free path $l_{ph}(\omega)$ of icosahedral $Al_{70}Mn_9Pd_{21}$ (solid line) in comparison with that following from $\lambda(T)$ of amorphous SiO_2 (broken line). The different parts (a,b,c,d) are explained in the text.

oscillators[18] is compatible with l_{ph} of the order of $\pi v_s \omega^{-1}$, i.e., half of the wavelength of the excitation.

In Fig. 6 we plot the frequency dependence of a temperature-independent $l_{ph}(\omega)$ that follows from our $\lambda_{ph}(T)$ data. In this figure, part *a* of the solid line represents the crossover region to a frequency-independent mean free path of 0.025 cm determined as described in Ref. 3. Part *b* represents a phonon mean free path $l_{ph} \propto \omega^{-1}$ to account for the nearly quadratic temperature variation of λ_{ph} in the T range between 0.35 and 1.6 K. Part *c* represents the strong decrease of the mean free path $l_{ph} \propto \omega^{-8}$ that needs to be assumed for describing the extended plateau region of the $\lambda_{ph}(T)$ data. Part *d* is the minimum conceivable mean free path for high-frequency nonpropagating quasilattice excitations of the order of $\pi v_s \omega^{-1}$. The previously published suggestion of $l_{ph} \propto \omega^{-4}$ for various amorphous materials is by far not adequate for fitting our data. The need of an exponent lower than -4 to fit our data arises from the extension of the λ plateau of icosahedral $Al_{70}Mn_9Pd_{21}$ over a distinctly larger temperature range compared to that observed for amorphous solids.

On closer look, we note a shallow maximum in the $\lambda_{ph}(T)$ variation in the plateau region at about 30 K, followed by a minimum at 50 K (see inset to Fig. 5). The appearance of the maximum in the $\lambda_{ph}(T)$ curve implies that the mean free path of phonons with a given frequency decreases with increasing temperature. Although the theoretically predicted T^{-3} temperature dependence of λ_{ph} is not really observed, we believe that the decrease of λ_{ph} with increasing T from 30 to 55 K above the maximum is due to the quasiperiodic umklapp processes introduced above. We interpret the maximum in the $\lambda_{ph}(T)$ curve as a crossover from the regime of a dominant scattering of phonons by tunneling states to the regime where quasiperiodic umklapp processes are important.

Our interpretation of the appearance of a maximum and the region of negative slope $d\lambda_{ph}/dT$ in the $\lambda_{ph}(T)$ data of icosahedral $Al_{70}Mn_9Pd_{21}$ gains further support from recent $\lambda_{ph}(T)$ measurements on a single grain of an $Al_{70}Mn_{10}Pd_{20}$ quasicrystal.[24] These data are included in Fig. 5 as open squares. The thermal conductivity of icosahedral $Al_{70}Mn_{10}Pd_{20}$ also shows an approximately quadratic T variation below 1 K, characteristic of phonon scattering on tunneling states (see Fig. 5). The absolute values of $\lambda_{ph}(T)$ are about a factor of 4 higher than for icosahedral $Al_{70}Mn_9Pd_{21}$, suggesting a lower concentration of tunneling states. The lower rate of phonon scattering on tunneling states leads to an enhancement of the height of the $\lambda_{ph}(T)$ maximum and to an extension of the temperature range where the slope of $\lambda_{ph}(T)$ is negative. For icosahedral $Al_{70}Mn_{10}Pd_{20}$ the region with a negative slope $d\lambda_{ph}/dT$ extends from 25 to 85 K at least and λ_{ph} decreases by about 25% with increasing temperature in this range, in agreement with our arguments above.

VII. CONCLUSIONS

On theoretical grounds we introduce the concept of generalized umklapp processes in condensed matter with quasiperiodic but well-ordered structures. We argue that these processes lead to a power-law dependence of the mean free path of delocalized quasilattice excitations on temperature as opposed to the exponential temperature dependence of the phonon mean free path due to conventional umklapp processes in periodically structured crystals. We compare this concept with experimental data of the quasilattice thermal conductivity $\lambda_{ph}(T)$ of a quasicrystalline material and find that it provides a reasonable explanation for the plateautype feature in $\lambda_{ph}(T)$ that seems to be a general observation for quasicrystals. This puts the frequency and hence temperature dependence of the mean free path in quasicrystals on a firm theoretical ground, quite in contrast to the situation that still prevails for amorphous materials.

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