

Sound-mode broadening in quasicrystals: A simple phenomenological model

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(Received 19 May 2003; revised manuscript received 7 October 2003; published 20 February 2004)

We propose a simple phenomenological model to analyze vibrational characteristics of quasicrystals (QCs). The interpretation of the obtained recently data is based on the existence of almost dispersionless optical modes most probably related to the specific clusters which constitute the characteristic building blocks of any QC structure. We generalize to QCs the well-known Akhiezer mechanism (responsible for the absorption of sound even in an ideal crystal), which in our case is related to a “long wave” disturbance of the quasicrystalline optical modes by the propagating sound modes. At higher wave vectors strong hybridization of acoustic and optical modes takes place, and it leads to a more steep broadening dependence on wave vectors, and besides the excitation can no longer be described as a single acoustic mode with a well defined wave vector. We show that the observed sound-mode attenuation behavior can be consistently described by these scenarios without invoking additional mechanisms.

DOI: 10.1103/PhysRevB.69.054205

PACS number(s): 61.44.Br, 63.20.Dj

I. INTRODUCTION

In most many body systems there is a fairly sharp distinction between elementary excitations (quasi-particles) and the collective modes involving a coherent mixture of quasi-particles. In a harmonic dielectric crystal these elementary excitations are phonons, having the linear dispersion law. At finite temperatures T the great majority of phonons have energies at most of the order of T due to the nature of the Bose–Einstein distribution, and the thermodynamic properties of crystals are determined almost entirely by these excitations. The wave vector associated with a phonon in a regular lattice is only defined modulo a reciprocal lattice vector, a feature which plays a crucial role in the dynamical properties of crystals. In this respect phonons in crystals are quite different from sound waves in liquids which carry true momentum. Each atom in the crystalline lattice executes only a finite amplitude motion (the oscillation about the lattice site). Evidently the mean momentum of such a motion is identically zero, and therefore, the phonon flux associated with the energy flux in a crystal is not accompanied by a transfer of mass.

QC systems are quite different from conventional solid crystals. QCs are built roughly speaking according to the following principles: Being homogeneous like crystalline alloys QCs nevertheless possess specific structural elements, or clusters (see Refs. 1–3). The resulting construction has no translational symmetry but has long-range order. In spite of

the fact that QC systems do not show lattice periodicity, the above statement on zero mean momentum of phonons should be true for QCs also. Thus one must admit that phonons in QC systems carry not momentum but only quasi-momentum, which is defined, unlike in conventional crystals, modulo a dense set of reciprocal vectors.⁴ Both systems (crystals and QCs) in this respect are different from liquids, where the phonon momentum is the actual momentum and the phonon flux does involve a transfer of mass, since in a sufficiently long time any atom or molecule can reach any point in the volume.

Thus one could say that i-QCs are quasicrystalline on the near atomic scale, and isotropic solids at macroscopic scales. Somewhere there ought to be a crossover from the local arrangement to macroscopic isotropy. If this structural crossover is sufficiently abrupt, it might also mark in reciprocal space the upper frequency end of the isotropic acoustic branches. Besides, because QC systems do not show lattice periodicity, the concept of Brillouin zone and its use for characterizing elementary excitations like phonons, strictly speaking is not adequate⁵ (see also, Refs. 6 and 7). On the other hand, at least at first glimpse, it can be argued that because in the long-wavelength limit the phonons can be described as sound waves propagating through an elastic continuum, there should be little difference between QCs and crystalline materials in this regime where the phonons are insensitive to microscopic structure. Nevertheless, experi-

mental results are fairly different in the two types of systems. The following correlation is characteristic of classical mono-crystals: The higher the quality of the diffraction pattern of a mono-crystal, the lower the line-widths of the phonons. The correlation reflects the wave nature of phonons: The better the conditions for the propagation, the smaller the scattering of the Bloch-type waves. Since QCs do not possess lattice periodicity vibrational excitations QCs are intrinsically not Bloch-type eigenmodes and, therefore, this correlation does not hold.

The same kind of problem has been discussed in the literature on electronic excitations in QCs. Some unusual electronic properties (e.g., a very low value of the electrical conductivity, a negative temperature coefficient of the resistivity, and so on) as it was shown recently⁸ are the consequences of the spectral properties of quasiperiodic Hamiltonians. It is common knowledge now that Anderson localization⁹ is a second-order phase transition between eigenstates that are spatially localized and those that are delocalized, and there are well defined conditions at which the localization–delocalization transition occurs. Many numerical investigations of quasiperiodic Hamiltonians reveal that their eigenstates are critical, i.e., they are characterized by a power-law decay of the amplitudes (see Refs. 10 and 11). The existence of the critical electronic states can be understood as related to the self-similarity of the QC structures: A given atomic cluster is repeated within a distance of the order of its size, and therefore an eigenstate from any cluster can easily tunnel to the next one which has an identical form. Thus electronic properties of QCs are critical, at least from the point of view of the second-order Anderson localization–delocalization transition.

However this analogy can not be bluntly applied to the study of vibrational modes in QCs. There are two principal differences between the Anderson electron and the vibrational problems, which are our main concern here. Firstly, when the system is mechanically stable, there are no negative eigenvalues (unlike in the electron case). Secondly in the 3d vibrational case, there are always at least 3 zero-frequency (in the long wavelength limit) Goldstone modes that can not be localized. Therefore, since the modes at the lower energy bound of the spectrum must have extended character, one can expect something similar to critical states (and to localization–delocalization transitions) only near the relatively high-frequency band edge.

The issue (elementary vibrational excitations in QCs) was intensively studied recently, and inelastic neutron scattering data (see, e.g., Refs. 11–17 and the monographs^{2,3}) reveal in the long wavelength limit well defined acoustic modes. The experimental quantity investigated in these experiments, the so-called scattering function $S(\mathbf{Q}, \omega)$, is measured close to a Bragg reflection located at a certain reciprocal vector \mathbf{G} in momentum space, and it probes phonons with frequency ω and wave vector $\mathbf{q} = \mathbf{G} - \mathbf{Q}$. To measure the largest signal one should measure $S(\mathbf{Q}, \omega)$ at high momentum transfer and close to a strong Bragg peak. Neutron scattering investigations were performed for different icosahedral^{12–15} and decagonal¹⁶ structures and it turned out that in spite of the fact that, structurally, decagonal and icosahedral QCs are

rather different, and that their physical properties (e.g., elastic moduli) might be also very different, they exhibit many common vibrational features. The purpose of our paper is to propose a simple phenomenological model to try to obtain as much theoretical information concerning vibrations in QCs as is possible on a phenomenological level without specific microscopic treatment. The rest of the paper is organized as follows: In Sec. II we give a short outline of neutron data related to vibrational mode studies in i-QCs AIPdMn and ZnMgY. Section III is devoted to the description of our phenomenological model. Finally, Sec. IV deals with miscellaneous subjects related to the vibrational dynamics in QCs.

II. EXPERIMENTAL OVERVIEW

The excitation spectrum of different QCs has been studied by inelastic neutron scattering on single grain samples. In spite of the often contradictory results of experimental investigations (see, e.g., discussions in Refs. 18 and 19), a few conclusions listed below seem inescapable. As explained above, it is only in the acoustic regime that a one to one correspondence can be made between the observed signal and a phonon, i.e., that the phonon wave vector can be defined. Generally, results are presented in an “extended” zone scheme, with a strong Bragg peak acting as the zone center (for crystallographic details see, e.g., Ref. 20). As proposed by Niizeki,^{21,22} important pseudo Brillouin zone boundaries (PZB) can be defined. They correspond to strong Fourier components of the atomic density from which acoustic modes are Bragg reflected. The positions of these pseudo zone boundaries obviously depend on the details of the atomic structure. Moreover, because of the quasi periodicity, several successive PZBs are stacked around each strong zone center.

In the following we will focus on two icosahedral phases for which detailed studies have been carried out: *i*–AIPdMn and *i*–ZnMgY. These two icosahedral phases have a very different atomic structure. Nevertheless their excitation spectra share a lot of common features. Their dynamical response can be separated into two well defined regimes: the acoustic regime for wave vectors smaller than 0.6 \AA^{-1} , and, for larger wave vectors, a regime in which the dynamical response is characterized by a broad band of dispersionless opticlike modes. In the acoustic regime, excitations are generally resolution limited for q smaller than 0.3 \AA^{-1} , and then broaden.

The way in which the broadening develops strongly depends on the atomic structure of the icosahedral phase: For instance, it is much more rapid in the *i*–AIPdMn phase than in the *i*–ZnMgY phase. The optic like spectrum generally consists of 3 or 4 broad “bands” (a few meV wide) around 7, 12, 16, and 24 meV. Although no gap opening could be observed at the PZBs, the energy of the lower optical modes roughly corresponds to the intersection of the acoustic branch with the PZBs.

Figure 1 summarizes results obtained for transverse modes propagating along a two-fold axis and polarized in a two-fold direction. The upper part is for the *i*–AIPdMn phase and the lower one for the *i*–ZnMgY one. The trans-

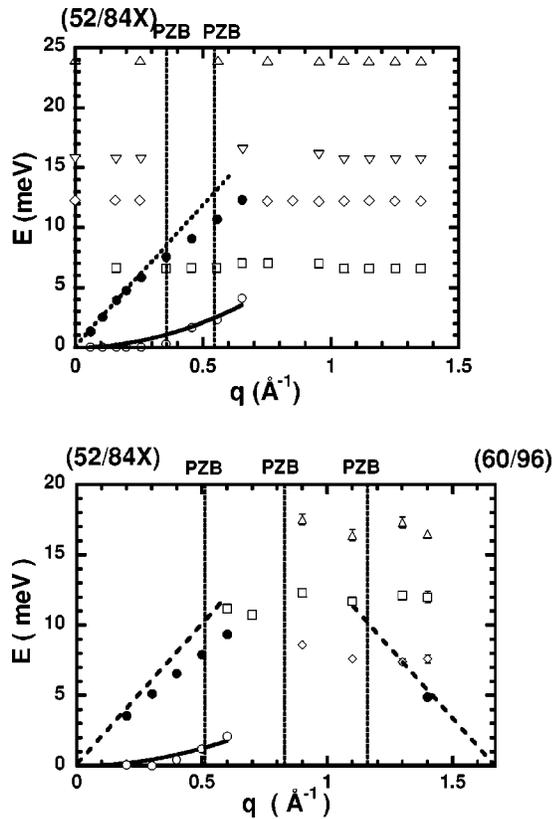


FIG. 1. Dispersion relation measured around the strong two-fold Bragg reflection with N/M indices equal to 52/84 and propagating along a two-fold axis in the i -AlPdMn (the upper figure) and i -ZnMgY phase (the lower figure). Results are presented in an extended zone scheme, the pseudo Brillouin zone boundaries are shown as vertical dashed lines. The transverse acoustic mode positions are shown as full circles and their width as open one. The dashed line is the linear dispersion relation as deduced from ultrasonic measurements and the solid line is a q^2 fit to the width increase. The other open symbols correspond to dispersionless optic like excitations, whose width is of the order 4 meV.

verse acoustic phonon dispersion relation is given by the full circles. The dashed line is the corresponding sound velocity as extracted from ultrasonic measurements. The open circles correspond to the width of the TA modes. The other open symbols correspond to the different broad optic bands, whose width is of the order 4 meV. For both phases the crossover between the acoustic and, say, hybridized regime is fairly abrupt: For the i -AlPdMn phase it shows up as a rapid broadening of the TA acoustic mode, whereas for the i -ZnMgY there is a coupling with an optic like excitation localized at 10 meV. In both cases the TA limit is given by $q=0.6 \text{ \AA}^{-1}$. Above this value the TA acoustic mode mixes up with optical excitations and the observed signal can no longer be described as a single excitation.

The broadening of TA excitations also depends on the structure: it is about two times larger in the i -AlPdMn phase than in the i -ZnMgY one. From the local slope of the TA dispersion relation and width of the excitations it is also interesting to compute the mean free path of TA phonons at the upper limit of the acoustic regime. We find a mean free

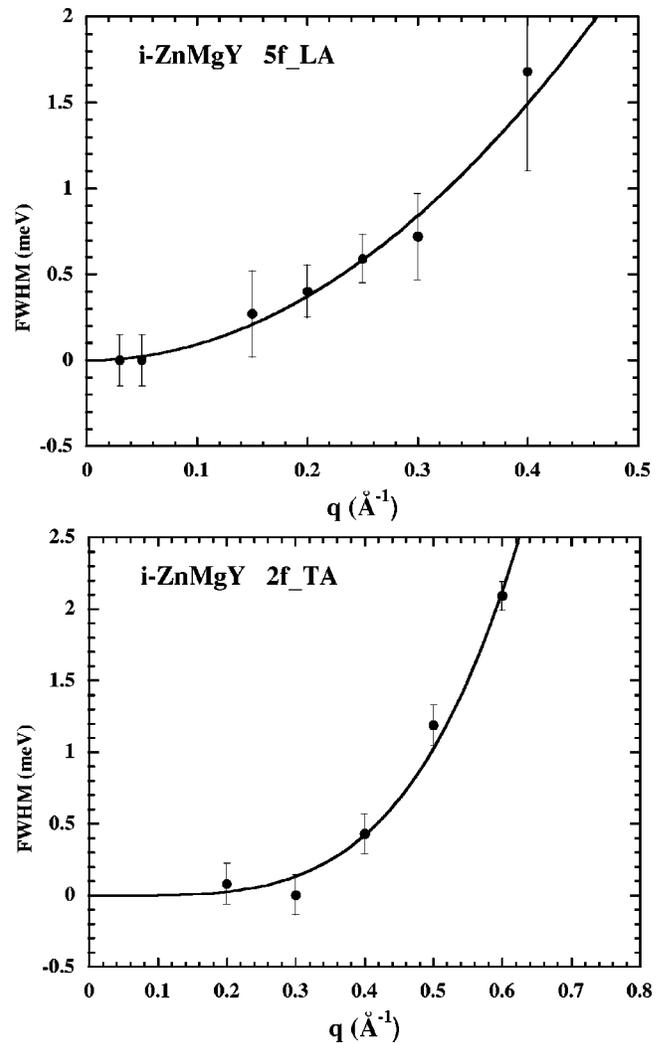


FIG. 2. Line width for longitudinal (the upper figure) modes propagating along a five-fold axis (experimental data and q^2 -fitting) and for the transversal modes (the lower figure) propagating along a two-fold axis (experimental data and q^4 -fitting) in the i -ZnMgY phase.

path of 12 and 24 \AA for the i -AlPdMn and i -ZnMgY phases, respectively. Structural x-ray studies²³ have shown one special geometric aspect of all QC structures, namely, atomic clusters, with a characteristic diameter $D_{cl}=10 \text{ \AA}$, which are part of the building blocks of QCs and their close approximants. Summarizing the experimental dynamics study, we can thus say that the crossover between the acoustic and mixed regime occurs for a phonon wavelength equal to D_{cl} . The mean free path of the TA phonon is equal to D_{cl} and $2D_{cl}$ for the i -AlPdMn and i -ZnMgY phases, respectively.

Line broadening is a nuisance in some circumstances, while in our case of QCs it provides valued physical information. We show in Fig. 2 dependences of line width for longitudinal (the upper panel) modes propagating along a five-fold axis and for the transversal modes (the lower panel) propagating along a two-fold axis in the i -ZnMgY phase, and the width for the transversal mode propagating along a

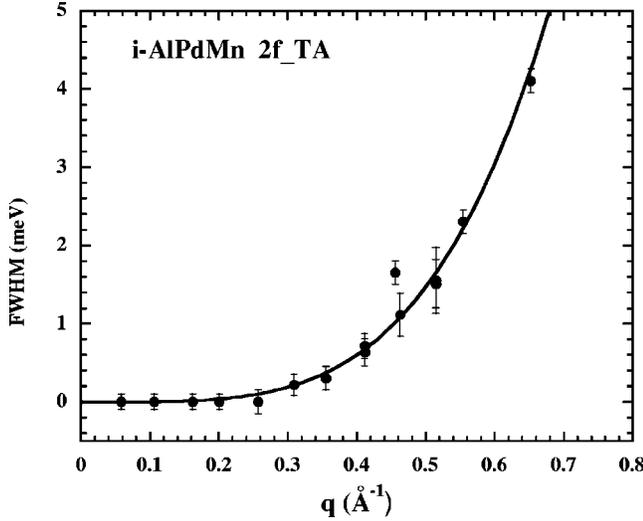


FIG. 3. Line width for the transversal mode propagating along a two-fold axis (experimental data and q^4 -fitting) in the i -AlPdMn phase.

two-fold axis for i -AlPdMn is shown in Fig. 3. The instrumental width for longitudinal modes (about 2 meV) is larger than for transverse (about 1 meV). To gain further insight into the nature of broadening we show in Figs. 2 and 3 theoretical fitting by q^2 dependence for the longitudinal waves and by q^4 for the transverse modes. We will see in the next section that these dependences correspond to broadenings dominated respectively by Akhiezer and by resonance hybridization sound absorption.

It is worth noting that very similar results are obtained for physically and structurally very different systems like i -QC CdYb or decagonal-QC AlNiCo (see Refs. 12–16). Even numerical values for the acoustic line-width are not very different, however, to be specific we restrict ourselves in this paper to the i -QCs AlPdMn and ZnMgY only for which we have more detailed and reliable data due to the high quality of the samples.

III. PHENOMENOLOGICAL AKHIEZER MODEL

Whereas the knowledge on the sound propagation in ordered crystals or disordered glass-like structures is now rapidly improving (see e.g., Refs. 24 and 25, and references therein), little is known about the behavior in systems with nonperiodic long-range order, such as QCs. The general objective of this study is to determine the structural and physical mechanisms associated with phonon line broadening in order to identify those features of QCs which differentiate them from conventional crystalline systems. Since both features (characteristic clusters and almost dispersionless optical modes) are interrelated and practically ubiquitous for all QCs it is natural to look for a model based on these widely recognized specific features. The primary aim of this section is to present such a physical model that reproduces available experimental data on vibrational eigenmodes in QC. In particular we are going to consider general phenomenological processes leading to sound absorption and dispersion, as the

detailed microscopic mechanisms governing these processes are not fully understood yet and have been heavily debated in the literature.^{18,19}

The occurrence of many broad optical, almost dispersionless modes has been, at least qualitatively, established and their origin can be easily understood. Indeed, as is well known,²⁶ the number of optical modes in a crystal is related to the number of atoms in the unit cell. Since in QC systems the unit cell size is strictly speaking infinite, it is not surprising to have in a certain energy window a dense set of optical modes. Moreover, due to natural energetic restrictions, the energy window where all the optical modes are confined is a relatively narrow one, which unavoidably leads to small mode dispersion. We shall thus include from the very beginning these modes into the model. On the other hand the dispersionless character of the optic modes means that the corresponding excitations see an almost uniform medium, the strain of which is slowly modulated by the sound wave. Due to the anharmonicity of the system the strain of the sound field causes a change of the optic mode frequencies. Despite the apparent analytic intractability of the problem, the various approaches suggest that all the complications can be subsumed by a simple phenomenological form.

In order for the discussion to proceed smoothly, we start first with a simple argument. We generalize to QCs the well-known Akhiezer mechanism of sound absorption (see, Refs. 27 and 28). In the case (we will be interested in below) when the frequency of the sound waves is much smaller than all relevant inverse relaxation times, the sound vibrations can be considered as a certain external field which modulates the QC structure, and therefore, the optical mode frequencies

$$\omega_0^{(\alpha,a)} = \omega_0^{(\alpha,a)}(1 + \gamma_{ij}^{(\alpha,a)} u_{ij}) \quad (1)$$

where α denotes the polarization of the optic mode a , $\gamma_{ij}^{(\alpha,a)}$ is the second rank tensor which characterizes the QC and depends on the direction of the optic mode (α,a) propagation. It can be viewed as the generalized Gruneisen constant tensor (see Refs. 27 and 28).

To simplify the matter (though it is not a crucial point) we assume that γ does not depend on the frequency ω . It can be shown using standard thermodynamic relations²⁹ that in this case the optical mode frequency modulation does not disturb the thermodynamic equilibrium within this branch, thus it is still proper to attribute a temperature T to it

$$\left(\frac{\Delta T}{T}\right)^{(\alpha,a)} \propto \left(\frac{\Delta \omega_0}{\omega_0}\right)^{(\alpha,a)}. \quad (2)$$

This relation (2) is valid if during the strain deformation collisions with other branches can be neglected. However, since for different modes the values of γ can be different (even could be negative or positive), after a strain deformation each branch has in general a different temperature (some branches might be cooled down, while others are heated up).

It might be useful first to separate all optical modes into two groups with different average temperatures but comparable specific heats. The average relative temperature difference between the groups, and according to (1) and (2), the average generalized Gruneisen coefficient $\hat{\gamma}_{\alpha av}$ can be then

easily calculated. Indeed one should bear in mind that although the modes contribute additively to the free energy, the Gruneisen coefficients are determined by a ratio of derivatives, i.e., a ratio of sums

$$\hat{\gamma}_{\alpha \text{ av}} = \frac{\sum_a \hat{\gamma}^{(\alpha,a)} C_a}{\sum_a C_a},$$

where C_a is the specific heat associated to the mode a .

If a sound wave propagates through a system, a periodic temperature difference will be set up between the two groups. Therefore in a certain characteristic relaxation time τ , heat exchange takes place between them, leading to entropy production, and absorption of the sound wave. The absorption coefficient per unit length α_α (in cm^{-1}) is determined by the entropy production rate and reads³⁰

$$\alpha_\alpha = \frac{C_\alpha T \gamma_{\alpha \text{ av}}^2}{\rho c_\alpha^3} \frac{\omega^2 \tau}{1 + \omega^2 \tau^2}, \quad (3)$$

where C_α is the heat capacity at constant strain corresponding to each sound mode polarization (constant volume for longitudinal waves and constant shear for transversal ones), ρ is the mass density, and $c_\alpha \equiv (c_l, c_t)$ is the longitudinal or transverse sound mode velocity.

It seems reasonable to assume that the energy exchange between the two groups takes place by means of phonon-phonon scattering processes. The relevant point is that the same kind of processes determine the heat conductivity³¹ κ

$$\kappa = \frac{1}{3} C_v c_l^2 \tau, \quad (4)$$

where C_v is the constant-volume specific heat, and c_l is the longitudinal mode velocity.

The expressions can be brought into more compact forms by taking into account that at high enough temperatures the relaxation time τ is much shorter than the sound period. Comparing (3) and (4) we end up with the following coefficients for the longitudinal (α_l) and transverse (α_t) sound absorption

$$\alpha_l \approx \frac{\kappa \gamma_{l,\text{av}}^2}{\rho c_l^5} T \omega^2, \quad (5)$$

and

$$\alpha_t \approx \frac{\kappa \gamma_{t,\text{av}}^2}{\rho c_t^3 c_l^2} \frac{C_t}{C_v} T \omega^2, \quad (6)$$

where C_t is the heat capacity at constant shear strain.

For the benefit of the skeptical reader, some comments about the applicability of the mechanism described in this section to transverse modes, seem in order. Indeed conventional wisdom claims that the Akhiezer mechanism cannot be applied directly to transverse sound modes, since only dilatation or compression deformations may provide nonuniform temperature variations and thermo-diffusive dissipation. To

resolve the contradiction we should recall that the building blocks of all QC structures are atomic clusters. These clusters are not mere geometrical constructions but real physical entities responsible for specific features in the QC vibrational spectrum (e.g., responsible for localized modes). Hence, one expects acoustic vibrations to induce stress inhomogeneities around each cluster, with a resulting strain field that will consist of a superposition of a dilatation and of a shear.

Therefore in this case the internal thermoelastic dissipation occurs even for the pure transverse vibrations. It is reminiscent of a phenomenon known in classical elasticity theory³² whereby a spherical inclusion placed in a vibrating medium containing only pure shears, introduces local strains which have dilation $\Delta(r, \theta, \phi)$ (r , θ , and ϕ are spherical coordinates) proportional to the shear deformation s at large distances from the inclusion

$$\Delta = -s \left(\frac{5\mu}{9\lambda + 14\mu} \right) \left(\frac{D_{cl}}{r} \right)^2 \sin^2 \theta \cos \phi \sin \phi, \quad (7)$$

where, for an isotropic material, μ and λ are the only two nonzero elastic constants. To find Akhiezer line broadening following from the dilation (7) one should calculate and average the corresponding Gruneisen coefficient. For one spherical inclusion in a bulk isotropic material the total effect will be zero due to angular integration. But of course this picture of one isolated cluster in a vibrating medium is not adequate for QC clusters. A complete account of the strains induced by the sound modes in real QC structures requires considerable computational work, that is beyond the scope of our purely phenomenological model. Thus we use in (6) phenomenologically the average transverse Gruneisen coefficient $\gamma_{t,\text{av}}$. However, from simple arguments presented above concerning the expression (7) we can come to the conclusion that due to the clusterlike nature of all QC structures, the Akhiezer broadening is expected to be less efficient for the transverse phonons than for the longitudinal ones. Therefore at least for the former case we have to look for another mechanisms contributing to transverse mode broadening in QCs.

QC clusters play also a crucial role for sound mode hybridization in QCs. Indeed, if we consider the vibrations of an isolated cluster of size D_{cl} , then the high-frequency modes with $\lambda \ll D_{cl}$ will not be affected by the change in the boundary conditions at scale D_{cl} , involved in partial disconnecting the cluster from the rest of the system. On the other hand, the low-frequency modes for which $\lambda \gg D_{cl}$ will disappear from the spectrum. Very qualitatively one can then think of such a material as a dense packing of connected clusters. Phonon scattering from fluctuations in the local velocity of sound $c^2(r)$, depends on the variance parameter

$$\nu(\lambda) \equiv \frac{\langle c^4(r) \rangle_\lambda}{\langle c^2(r) \rangle_\lambda^2} - 1 \approx \nu(a) \left(\frac{a}{\lambda} \right)^3,$$

where $\lambda = 2\pi/q$ is the phonon wavelength, and a is the scale at which elementary fluctuations occur (i.e., for QCs $a \approx D_{cl}$). Let us have $\nu(a) \gg 1$. Since $c^2(r)$ is always positive, the only way this can occur is by having a small number of

“hard” regions for which c^2 is larger than the average, which dominate the variance, and a much larger number of “soft” regions. If the hard scatterers (of size D_{cl}) are isolated, the only propagating modes are those of the soft background medium. The scatterers act as rigid inclusions in the medium. Roughly, the hard scatterers simply increase the effective elastic constant by a factor proportional to the small volume factor they occupy. The above argument neglects the local modes centered around the hard scatterers. For a single scatterer they would be localized and have a high frequency. When they are dilute and uncorrelated, so that they couple weakly, they can only form a relatively high-frequency optical band. On the other hand, when they are correlated and coupled sufficiently strongly, they can dominate the elastic properties and the low-frequency velocity and damping of sound. As a rule of thumb we can estimate the characteristic frequency for mode hybridization as c_α/D_{cl} . In this region phonon wavefunctions are repetitively localized on the clusters and this recurrent localization comes from the fact that, unlike for periodic crystals, it is not possible for the wavelength to be commensurate simultaneously with all inter-cluster distances in a quasiperiodic structure. Evidently this cluster vibration mode is not an exact eigenstate. It interacts with other vibration modes of similar energy and as well with acoustic phonons. This interaction affects both the cluster vibration mode and the phonons, which are for the same reason also not exact eigenstates. In other words due to the above described mode-hybridization the excitations will be broadened and shifted.

Physically these cluster vibrations are caused by a local deviation of the force constant matrix and atomic mass from the average values. The hybridization phonon broadening Γ for a phonon with wave vector \mathbf{k} , polarization j , and frequency ω_0 depends on a “concentration” [i.e., density of states $g(\mathbf{k})$] of these cluster vibrations and is proportional to the phonon-cluster mode scattering matrix \hat{t}

$$\Gamma(\omega_0(\mathbf{k},j)) = g(\mathbf{k}) \frac{1}{2} \frac{\Im\langle \mathbf{k},j | \hat{t} | \mathbf{k},j \rangle}{\omega_0(\mathbf{k},j)}. \quad (8)$$

To obtain total hybridization broadening this expression (8) has to be averaged and integrated over all cluster vibrations with frequencies approximately equal to ω_0 . If the structure of the mode does not strongly depend on its frequency, and their distribution is smooth on a scale of their width, Γ will have k -dependence coming mainly from the density of states factor in (8). Using the arguments borrowed from the standard glass theory³³ we can conclude that $g \propto k^4$.

IV. DISCUSSION AND CONCLUSION

Let us summarize now the experimental results and the theoretical interpretation presented in this paper. The physical essence of the model is as follows. If the wavelength λ of the sound is larger than the characteristic cluster size D_{cl} we can assume that each cluster is subject to a uniformly distributed stress. However, due to not spherical cluster shape and boundary conditions at their surfaces, the resulting deformation is not uniform. It varies considerably over the dimension

of a cluster (not over sound wavelength as in a uniform bulk body). Hence sound broadening due to thermal conduction (Akhiezer mechanism) is essential for the case. To be specific this scenario can be rationalized as sound modes (mainly longitudinal but as we have seen also transverse) somehow perturb the manifold of optic modes or QC cluster vibrations (the characteristic features of QC materials taken into account by the model). These perturbations in terms of Gruneisen parameters are reduced to temperature variations, and the latter ones lead to thermo-diffusional relaxational sound absorption. It might at first sight seem that the model contains no QC-specific features: But in fact all QC properties are hidden in the material parameter values entering the formula.

The discussed above (in the previous Sec. III) second broadening mechanism—resonance hybridization of sound and cluster vibration modes—can compete with the Akhiezer absorption even for the longitudinal modes in the relatively high q -region. All the more it is true for the transverse waves where the Akhiezer broadening is expected to be less efficient. Fortunately they do have different q -dependencies that allow us to disentangle them analysing experimental data. To span a wide range of possibilities let us discuss first the Akhiezer mechanism. To compare the expressions with neutron data it is more convenient to use:

- (i) the absorption coefficient $\Gamma [s^{-1}] = \alpha [cm^{-1}] c_\alpha$;
- (ii) the wave vector q instead of ω .

Thus we come from (5) to

$$\Gamma = \frac{\kappa \gamma_{av}^2 T}{\rho c_\alpha^3} q^2. \quad (9)$$

The proposed model can be checked against experimental data along three lines: The wave vector dependence of the acoustic mode broadening which should be proportional to q^2 , its temperature variation and its order of magnitude.

The q^2 increase of the acoustic phonon width was generally observed for longitudinal acoustic excitations. For TA modes the width increase seems to be more abrupt, (for a wave vector larger than 0.3 \AA^{-1}) and goes rather as a q^4 law as shown in Figs. 2 and 3. Presented by the solid line in Fig. 1 a q^2 fit to the observed widths, illustrates this statement, and besides, can be used to extract some quantitative information on the magnitudes of q^2 and q^4 parameters. As expected, the coefficient of the fit is twice as large in the i -AlPdMn phase as in the i -ZnMgY one (9.8 and 4.9 meV/\AA^{-2}). In the i -ZnMgY, we also found that the LA mode broadens twice as fast as the TA modes. More accurate inspection of the data presented in Fig. 1 clearly shows that for a region close to the mixed regime (i.e., to $q \approx 0.6 \text{ \AA}^{-1}$) the broadening is more rapid than a q^2 law. It can be attributed to Rayleigh scattering arising from local fluctuation in sound velocity due to either mass density or elastic constant fluctuations (in own turn related to the cluster structure of all QCs) or to acoustic-optic mode hybridization. In mathematical form both phenomena lead to q^4 broadening,³³ though of course the Rayleigh scattering seems generally too weak in homogeneous materials like QCs.

The next test concerns temperature dependences. Since at high enough temperatures $\kappa \propto 1/T$, in the present approximation the effect of the temperature dependent factors cancels, and therefore, the absorption due to Akhiezer mechanism can only weakly depend on temperature. It is worth noting also that the very possibility to characterize the system by only one Gruneisen function is based on the assumption that the dependence of the energy levels on the volume (or more generally on the deformations u_{ik}) is expressible in terms of a single characteristic energy. This assumption is correct for Einstein or Debye models, partly also for conventional (with relatively small elementary cell) crystalline solids but not evident at all for QC systems where a number of different contributions to the free energy can control the thermodynamic properties (see discussion in Refs. 18 and 19).

Thus in this approximation (namely, in the high temperature regime where $T > \Theta_D$, Θ_D being the Debye temperature, 400–500 K, within the classical Debye approach valid in this region, $\kappa \propto 1/T$ (all other parameters determining Γ do not depend noticeably on T) and Γ is temperature independent in agreement with experimental data. Of course it is only an approximation and we are aware that QCs are not classical Debye insulators, and besides we are not in the regime $T \gg \Theta_D$, but it identifies correctly the characteristic scales in the problem. A detailed temperature dependence of the lattice dynamics has only been carried out in the i -AlPdMn phase. At $T = 1050$ K the slope of the transverse acoustic mode displays only a 10% decrease as compared to room temperature and the broadening of the modes did not show any significant variation. This is thus in agreement with the model since the Debye temperature of the i -AlPdMn phase is about 500 K (see, e.g., Ref. 18).

Quantitative comparison of our model predictions with experimental data is more difficult since there are only scarce data available for the model input parameters and their temperature dependences (there are no systematic measurements of all needed material parameters, and experimental data are still not very accurate). Thus from here on in this section we shall not attempt to maintain numerical accuracy, but only indicate the form of the answer. Our model can be used to estimate the q^2 coefficient in the phonon line broadening. Namely, from Refs. 18, 19, and 34

$$\kappa \approx 1 \frac{W}{mK} = 10^5 \frac{\text{erg}}{\text{scmK}},$$

$$\rho = 5 \text{ g/cm}^3; \quad \gamma_{\text{av}} \approx 1,$$

and, besides, $T \approx 300$ K. Using also the literature (Refs. 13 and 14) data for i -QC materials (ZnMgY $c_l = 4.8 \times 10^5$ cm/s, $c_t = 3.1 \times 10^5$ cm/s), and neutron measurements (Refs. 13, 14, and our data shown in Figs. 1–3), one can find that at $q \approx 0.5 \text{ \AA}^{-1}$, for the longitudinal phonons, $\Gamma_l \approx 1.5$ meV, and for the transverse modes, depending on propagation directions $\Gamma_t \approx 1$ meV; for AlPdMn ($c_l = 6.3 \times 10^5$ cm/s, $c_t = 3.5 \times 10^5$ cm/s), at the same $q \approx 0.5 \text{ \AA}^{-1}$, $\Gamma_t \approx 2$ meV (no data for longitudinal mode line-widths). Putting all values together we find for the q^2 coef-

ficient about 10 meV/\AA^2 in reasonable agreement with the experimental value quoted above.

One more interesting line of thought is to apply our results to estimate the anisotropic Gruneisen parameters, introduced in (1). The observed difference in width increase rate between transverse and longitudinal excitations means that a different Gruneisen parameter should be used in each case. Comparing experimental data for longitudinal and transverse modes in the same material (for the moment such data are available only for i -ZnMgY) we can conclude that the volume Gruneisen parameter entering the longitudinal phonon broadening (5) should be four times larger than the shear anisotropic Gruneisen coefficient which determines the transverse phonon line broadening (6).

These Gruneisen parameters can be seen as a phenomenological way of taking into account the interaction between LA or TA modes and optical branches. Various phonon interaction processes result when anharmonic terms of the third (and higher) order in the displacements are taken into account. The first anharmonic term corresponds to the decay of one phonon into two or to the coalescence of two colliding phonons into one. For conventional crystals, usually the main contribution comes from processes within the same optical branch. For QCs, the dispersionless character of the optical branches could lead to a situation where the conditions for phonon decay or recombination are satisfied above a finite threshold wave vector (corresponding to the crossing with the lowest frequency optical branch), and not for a discrete set of wave vectors but for a whole spherical shell in q space.

The coefficient at q^4 for the resonance hybridization broadening, is a model dependent quantity. However, it can be always presented in the following form:³³

$$\alpha_{\alpha, \text{res}} = \Lambda_{\alpha} \frac{\omega}{c_{\alpha}} \left(\frac{\omega}{\omega_{cl}} \right)^3, \quad (10)$$

where Λ_{α} is a model and mode polarization dependent coefficient, and ω_{cl} is a characteristic frequency for cluster vibrations. To illustrate this issue we show in Figs. 2 and 3 q -dependences of line width for longitudinal and transversal modes in the i -ZnMgY and i -AlPdMn QCs. These results are clearly indicating that for the longitudinal modes the broadening are governed mainly by the Akhiezer mechanism, whereas for the transversal waves q^4 fitting leads to a reasonable agreement with the data. Moreover the coefficient at q^4 found from such a fitting is conformed with (10) describing resonance hybridization sound absorption.

Of course the imperfect knowledge of the parameters in a large temperature interval makes our predictions only qualitatively or semiquantitatively correct, and at this stage, a number of open questions must be stressed. For example, no forbidden gaps have been observed experimentally. There is also some inconsistency between thermodynamic and inelastic neutron scattering data.^{18,19} A possible origin of this inconsistency may be related to contributions, say to the specific heat, from the cluster vibrations. It is worth noting, however, that all scattering experiments measure a dynamic structure factor $S(q, \omega)$, which is the Fourier transform of

the displacement correlation function, and therefore does not carry direct information about propagation or nonpropagation of modes. But to put these speculations on a firm footing, further experimental and theoretical efforts are required. Nevertheless for a model with such a small amount of physical input, our results show quite good agreement with experiments (the order of magnitudes together with its evolution with T and q). In addition we can give some qualitative predictions concerning Gruneisen parameters and anharmonicity in different QC materials. It is worth noting in this respect one very recent theoretical prediction³⁵ that Gruneisen parameters should diverge close to a quantum critical point. In a certain sense QC phonon and electron states are critical over the whole region of the QC state stability. However, it should be noted that dynamics of QC is still a developing field and much of the excitement arises from the possibility of discovering novel physics beyond say the classical paradigms discussed here.

As this paper was being written for publication we became aware of parallel efforts^{24,25} to investigate the cross-

over from propagating to strongly attenuated acoustic modes in densified silica glasses. Although physically the two systems (QCs and glasses) are very different, acoustic mode broadening reported in these papers and its interpretation (Akhiezer attenuation due to optical mode perturbations and acoustic-optic modes scattering and hybridization) are quite similar to our results. Of course the nature of these optical modes is different in each case: In glasses it is related to disorder (in silica glasses it corresponds to librations of SiO₄ tetrahedra), in QCs the optic modes are related to cluster vibrations or, in other words, to the translational or orientational frustrations intrinsic to all QCs.

ACKNOWLEDGMENTS

One of us (E.K.) acknowledges support from INTAS (under Grant No. 01-0105). We thank T. Janssen for thought-provoking discussions.

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