Minimal model of the phonon-phason dynamics in icosahedral quasicrystals and its application to the problem of internal friction in the *i*-AlPdMn alloy

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Different existing elastodynamical models of icosahedral quasicrystals are analyzed. The simplest minimal model of the phonon-phason dynamics is formulated. Physical restrictions and possible applications of the proposed model are considered. Several generalizations of the minimal model are discussed. It is shown that the phonon-phason coupling induces a resonant absorption peak of low-frequency sound waves in the temperature region corresponding to a thermal activation of phason excitations. The maximum value of the logarithmic decrement of the sound wave damping depends on the propagation direction and the wave polarization. The anisotropy is proportional to the square of the phonon-phason coupling constant K_3. Our estimation shows that the effect can be resolved experimentally if the relative value of K_3 is not negligible with respect to four other elastic constants of the icosahedral quasicrystal. Namely, the difference should not exceed two orders of magnitude.

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I. INTRODUCTION

Quasicrystals (QC's) discovered in 1984 (Ref. 1) combine aperiodic long-range positional order with noncrystallographic rotational symmetry. The diffractograms of these materials can be indexed with the help of linear combinations of a finite number N of basis vectors $\mathbf{b}_i$ in reciprocal space. This fact makes possible the expansion of the quasicrystal density function in Fourier series. The number N is always greater than the dimension of the physical space which results in specific symmetry properties responsible for the physical difference between quasicrystals and crystals. In particular, QC's have an additional Goldstone phason degree of freedom **w**, which is absent in a crystalline state. The inhomogeneous variation of **w** corresponds to a relative shift of incommensurate density waves which form QC's.[2] The inhomogeneous variation of **w** gives a contribution to the elastic energy of QC's.[3] As a result, the elastic properties of QC's, and especially their dynamical elastic properties, have a specific character. To date three different models of the phonon-phason dynamics in QC's have been proposed. Each model is presented as a system of coupled differential equations. Some of the equations belonging to different models are quite equivalent, but others are mutually incompatible. In our opinion, the updated experimental results and theoretical knowledge in the physics of QC's permit now to formulate the simplest minimal model of the phonon-phason dynamics, and to discuss its properties and its limitations together with the modifications of the minimal model for different applications. This task constitutes the aim of the present work. The paper is organized as follows. In Sec. II we discuss the existing models of the phonon-phason dynamics and formulate the minimal model. Section III is devoted to the limitations and some applications of the proposed model. Section IV considers the phonon-phason coupling mechanism of the resonant absorption of sound waves.

II. PHASON-PHONON DYNAMICS MODELS AND THEIR ANALYSIS

In their pioneering work Lubensky and co-workers have proposed a system of elastodynamical equations describing the behavior of an icosahedral QC:[4,5]

$$\partial_t \rho + \nabla \cdot \mathbf{g} = 0,$$

$$\partial_t g_i - \nabla_j (\eta_{ijkl} \nabla_k g_l) = -\frac{\delta F}{\delta u_i} - \rho \nabla_i \frac{\delta F}{\delta \rho},$$

$$\partial_t u_i + \Gamma_u \frac{\delta F}{\delta u_i} - v_i = 0,$$

$$\partial_t w_i + \Gamma_w \frac{\delta F}{\delta w_i} = 0, \tag{1}$$

where ρ is the density, $\mathbf{g}$ is the momentum density, η_{ijkl} is the ordinary viscosity tensor, F is the ratio of the total QC energy to its volume, Γ_u and Γ_w are the dissipative kinetic coefficients, and v_i is the velocity of the point with respect to the coordinate system. The first Eq. of system (1) is a mass conservation law. The second one has the form of a modified Navier-Stokes equation. Its right part is an elastic force vector acting upon a unit volume. This vector consists of two parts, the first of which is induced by a medium strain and the second one is caused by a density change due to a mass current. The left part of the second equation also includes two terms. The first one stands for a unit volume acceleration and the second one corresponds to the viscosity friction. The third and the fourth equations describe relaxation phenomena in QC's.

The solution of system (1) shows[4,5] that the sound velocities are isotropic but their damping reflects the icosahedral symmetry of the structure. The anisotropic part of the viscous damping is probably smaller in magnitude by a factor

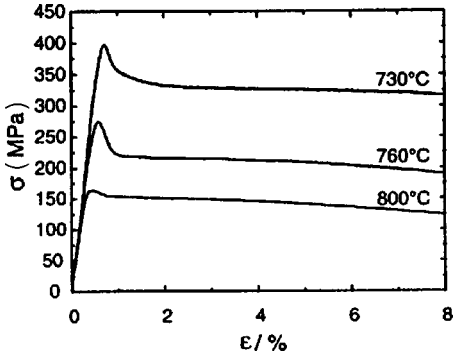

FIG. 1. Stress-strain dependences of icosahedral $Al_{70.5}Pd_{21}Mn_{8.5}$ sample stressed along twofold direction [0/0, 0/0, 0/2] at different temperatures (Ref. 8).

of 10^{10} than the isotropic part. The phason relaxation time in a sample with a size 1 cm is of the order of 3–300 000 yr.

Actually the results[4,5] mean that the sound attenuation anisotropy and the phason jumps should be experimentally nonobservable in these materials. As far as the authors know, there exists no direct experimental work reporting the observation of the sound attenuation anisotropy in icosahedral QC's. But to date the phason motion in quasicrystals is well known[6] and constitutes a contradiction with the results.[4,5] An attempt to modify system (1) has been made.[7] In the approach,[4,5] The phonon relaxation rate gives the diffusion constant of defects. The model[7] distinguishes the kinetic constant for the phonon Γ_u and that for the defect Γ_d since phonons and phasons are relaxed much more quickly than the defects. Consequently an additional defect relaxation process is considered.

As has been shown in several recent experimental works (see, for example, Ref. 8), the elastic behavior of QC's at low strain is similar to that of ordinary crystals. Even in the region of high temperatures the low strain is always elastic (see Fig. 1). Plastic deformation starts at some critical nonzero stress value. Similarly to the case of ordinary metallic systems, the critical stress value decreases with the temperature increase.

In our opinion the third equation of system (1) describing the phonon relaxation is principally applicable only to plastic solids or to liquid systems. This equation states that in the stressed QC's its strain relaxes with the velocity proportional to its amplitude at any strain value. It means that purely elastic stress in QC's is impossible even at low phonon and phason displacements **u** and **w** corresponding to linear differential equations. Actually the third equation in system (1) contradicts the well-known fact that the dissipative function of an elastic solid depends only on the spatial derivatives of the displacement rate $\nabla_i \partial_t u_j$ and not on the displacement rates $\partial_t u_j$ themselves.[9] Consequently in an elastic solid, acoustic-phonon relaxation is mainly caused by the viscosity phenomenon described by the viscosity tensor η_{ijkl}.

Another approach to the elastodynamics of QC's has been proposed in Ref. 10 and developed in Ref. 11 In this model the term of elastic energy corresponding to the density variation is neglected and the elastic energy is presented in the following form:

$$F_{el} = F_{phonon}(\lambda, \mu) + F_{phason}(K_1, K_2) + F_{coup}(K_3), \quad (2)$$

where λ and μ are the Lame coefficients, K_1 and K_2 are the phason elastic constants, and K_3 stands for the phonon-phason coupling term. According to Refs. 10 and 11 the components P_{ij} of the ordinary stress tensor can be calculated as the derivatives of the elastic energy (2) with respect to the corresponding components of the ordinary strain tensor E_{ij}. Consequently, the components P_{ij} have a linear relation with the components E_{ij} and $\partial_i w_j$ of the ordinary strain and the phason one, respectively. Here $\partial_i w_j$ is defined as the derivative of the jth component of the phason degree of freedom **w** with respect to the ith component of the radius vector. The tensor H_{ij} of the phason stress is introduced analogously to the classical tensor P_{ij}: its components are defined as the derivatives of energy (2) with respect to the corresponding strain components $\partial_i w_j$. The components H_{ij} are also linearly related to the E_{ij} and $\partial_i w_j$ tensors. Two interdependent partial differential equations of motion of a medium were proposed. The first of them has the form:

$$\partial_j P_{ij} + f_i = \rho \ddot{u}_i, \quad (3)$$

where f_i is the density of bulk forces, ρ is the mass density, and $\ddot{u}_i$ is the acceleration of the point. As is well known, this equation, which is analogous to the second Newton's law, is also valid in the crystalline case. The second equation expresses the vector of the phason acceleration and has a form similar to that of Eq. (3):

$$\partial_j H_{ij} + g_i = \rho \ddot{w}_i. \quad (4)$$

Here g_i is the density of phason bulk forces. Evidently, Eq. (4) is analogous to the second Newton's law for the so-called generalized phason force. Equations (3) and (4) were derived using the consideration of the QC motion in the six-dimensional (6D) space, where the description of QC's is similar to that of ordinary crystals in the 3D space. It explains completely ''crystalline'' and resembling forms of Eqs. (3) and (4).

We start the analysis of this model on the note that it is possible to write an equation of motion for a generalized degree of freedom in the form of the second Newton's law only if there exists a corresponding conservation law. However, the phason mode in QC's corresponds to atomic jumps or diffusion, no conservation law corresponds to the diffusion of atoms. Moreover, the direct solution of Eqs. (3) and (4) assuming that both bulk phonon and phason forces are equal to zero predicts six soundlike branches with the dispersion law $\omega \propto q$. This result is inconsistent with the experimental data (see, for example, Ref. 12) in icosahedral QC's which show only three branches of such a type. Nevertheless the theoretical idea which admits the propagation of soundlike phason modes in icosahedral QC's is used up to date. In Ref. 13 the authors try to calculate on this basis a low-temperature heat capacity of icosahedral QC's. The explicit form of energy (2) is used, the density of external forces is supposed to be zero, and the system of equations (3) and (4)

is solved. The number of resulting soundlike branches is found to be equal to six and contradicts again all available experimental data.

Reference 13 together with other theoretical works is a response to the abundant data accumulated due to recent experimental interest in low-temperature properties of QC's. One of the striking results concerning the heat-capacity $C_p(T)$ measurements is reported in Ref. 14 for $Al_{68.2}Mn_9Pd_{22.8}$ single crystals grown by the Chokhralsky method and for $Al_{70}Re_{8.6}Pd_{21.4}$ high-quality monophase QC's. For the icosahedral $Al_{68.2}Mn_9Pd_{22.8}$, the cubic (as a function of temperature) contribution to the low-temperature heat capacity $C_p(T)$ was found to be much greater (nearly the double) than the expected acoustic-phonon contribution calculated using low-temperature transverse and longitudinal sound waves data. The same result was obtained for the system AlPdRe. It is interesting to note that the vibrational contribution to heat capacity calculated using directly the experimental density function $g(E)$ of vibrational states fits quite well the experimental dependence of $C_p(T)$.[15] The result is impossible to obtain by the calculation in the framework of the Debye theory of lattice heat capacity based on the velocity of sound waves.

In order to explain this discrepancy, the notion of so-called nonacoustic localized states in QC's is usually introduced in literature (see, for example, Ref. 15). For further analysis one should clarify the relation between these states and the phason dynamics. For that goal two qualitatively different definitions of a phason can be distiguished. If phasons are considered to be collective atomic modes (surely nonlocalized in reciprocal space) there exists no direct relation of the localized states to the phason dynamics. This approach is adopted in all the models of the phonon-phason dynamics analyzed in the present work. According to the alternative definition, phasons are understood as local atomic jumps between several neighboring positions separated by a distance smaller than the atomic diameter and by a minimal potential barrier which makes possible atomic tunneling between these positions even at very low temperature. Then corresponding atomic jumps can give a contribution to the low-temperature heat capacity as is the case in glassy systems.[16] Maybe it is more convenient to call these states ''localized nonphonon'' ones because the atoms move in this case in the potential that is evidently nonharmonic.

The resumming conclusion of this comparison is that the models of QC elastodynamics based on the continuous medium approximation[4,5,10,11] cannot be used to calculate the low-temperature heat capacity of QC's. Nonpropagating collective phason modes cannot contribute to the density of vibrational states.[17] It concerns also the simplest minimal model of the phonon-phason dynamics developed in this work. However there exists a class of incommensurate structures where the collective phason modes are really similar to sound ones[18,19] and the elstodynamic model developed in Refs. 10 and 11 can be applied. These substances are the incommensurate intergrowth compounds (IIC). IIC needs two or more mutually incommensurate sublattices to characterize its structure. In IIC the above-mentioned phason momentum conservation law corresponds to the sliding of one sublattice with respect to another. Neglecting the pinning effect, one can consider the velocity of phason modes in IIC's. Thus, soundlike phason modes in IIC's are really observable. The simplest form of the equation of phason motion in IIC's is presented in the following form:

$$\partial_j H_{ij} = \rho_{eff} \ddot{u}_i , \tag{5}$$

where ρ_{eff} is the effective (or relative) density which is associated with the relative motion of sublattices, the phason friction, phason bulk forces, and the phason pinning being neglected. In contrast with the case of IIC the phason degrees of freedom in QC's do not correspond, in principle, to such kind of sliding. This fact implies the different forms of the equation of phason motion in QC's and in IIC's. To complete the analysis let us note that though the existing models of the QC elastodynamics explain several features of the phason-phonon motion, none of them is completely consistent with the experimental data.

In the following section we formulate a minimal model which tries to describe the basic features of the phason-phonon elastodynamics and can be generalized to account for more fine effects. In the quasicrystalline case, the attenuation of phason modes is essential in the dynamical theory. The simplest way to introduce this attenuation is to assume that the phason bulk force $g_i = -D\dot{w}_i$, where D is the friction coefficient. According to the previous analysis, the phason momentum in QC's is not conserved (there exist only three soundlike branches) implying $\rho_{eff} = 0$. In this case the obtained equation of the phason motion in QC's becomes equivalent to the fourth equation of system (1), if the relation between material constants is $\Gamma_w = 1/D$. In other words, in both our model and the approach developed in Refs. 4 and 5 the phason modes in QC's are considered as overdamped. The minimal model of the phonon-phason elastodynamics in icosahedral QC's can be then presented as a system of the two following equations:

$$\partial_j P_{ij} = \rho \ddot{u}_i , \quad \partial_j H_{ij} = D\dot{w}_i , \tag{6}$$

where $P_{ij} = \partial F / \partial E_{ij}$ is the ordinary (phonon) stress tensor and $H_{ij} = \partial F / \partial (\partial w_i / \partial x_j)$ is a nonsymmetrical tensor of the phason stress, the energy F being presented by Eq. (2). Actually, the minimal model (6) can be considered as a compromise between the approaches of Refs. 4 and 5 and Refs. 10 and 11, or as a simplification of the model presented in Ref. 20. Diffusion processes not related to the collective phason mode exitations are not considered in the proposed minimal model.

The solutions of the resulting system of linear differential equations can be found in the form of phonon-phason waves. These waves are characterized by a 6D polarization vector **U** and by an ordinary 3D wave vector **q**. If the constant of the phonon-phason coupling is small, then the waves with predominately phonon polarization and the waves with the predominately phason one can be distinguished. The minimal model of the phason-phonon elastodynamics is reduced to the following six equations:

TABLE I. Effective elastic constants K, δ_{ef}, and I as a function of the wave vector **q** direction and the sound wave polarization **u**.

q	**u**	**w**	K	I	δ_{ef}	N
$q\langle 1,\tau,0\rangle/N$	$u\langle 1,\tau,0\rangle/N$	$w\langle \tau,-1,0\rangle/N$	$K_1-4/3K_2$	$-2K_3$	$-6\xi_{\|\|}$	$\sqrt{\tau+2}$
	$u\langle 0,0,1\rangle$	$w\langle 0,0,1\rangle$	$K_1+2/3K_2$	0	$2\xi_{\|\|}$	1
	$u\langle -\tau,1,0\rangle/N$	$w\langle 1,\tau,o\rangle/N$	$K_1+2/3K_2$	0	$2\xi_{\|\|}$	$\sqrt{\tau+2}$
$q\langle \tau^2,1,0\rangle/N$	$u\langle \tau^2,1,0\rangle/N$	$w\langle 1,\tau^2,0\rangle/N$	$K_1+4/3K_2$	$2/3K_3$	$10/9\xi_{\|\|}$	$\sqrt{3\tau+3}$
	$u\langle 0,0,-1\rangle$	$w\langle 0,0,1\rangle$	$K_1-2/3K_2$	$4/3K_3$	$-14/9\xi_{\|\|}$	1
	$u\langle -1,\tau^2,0\rangle/N$	$w\langle -\tau^2,1,0\rangle/N$	$K_1-2/3K_2$	$4/3K_3$	$-14/9\xi_{\|\|}$	$\sqrt{3\tau+3}$
$q\langle 1,0,0\rangle$	$u\langle 1,0,0\rangle$	$w\langle 1,0,0\rangle$	$K_1-1/3K_2$	K_3	0	1
	$u\langle 0,1,0\rangle$	$w\langle 0,1,0\rangle$	$K_1+(\tau-1/3)K_2$	$\tau^{-1}K_3$	$2\tau^{-1}\xi_{\|\|}$	1
	$u\langle 0,0,-1\rangle$	$w\langle 0,0,1\rangle$	$K_1+(2/3-\tau)K_2$	τK_3	$-2\tau\xi_{\|\|}$	1

$$\sum_{j=1}^{6}[C_{kj}(\mathbf{q})U_j]=\alpha(k)U_k, \tag{7}$$

where $C_{kj}(\mathbf{q})$ is the well-known phonon-phason dynamical matrix (DM), U_k is the 6D polarization consisting of 3D phonon polarization **u** and 3D phason polarization **w**. In Eq. (7), for $k=1,2,3$ the coefficient $\alpha(k)$ is equal to $\rho\omega^2$ and for $k=4,5,6$ its value is $iD\omega$, where $i=\sqrt{-1}$. The solutions of system (7) are the dispersion relations $\omega(q)$ and corresponding polarizations. To analyze the solutions of Eq. (7) one can use the fact that a linear system with a zero right-hand side has a solution if its determinant is equal to zero. But the analytical solution of the presented system in its general form is impossible because of the enormous number of terms in the resulting equation. So we will consider analytically only three particular cases corresponding to the wave vectors which are parallel to fivefold, threefold, and twofold symmetry axes, respectively. For these directions the determinant of matrix M can be presented as a product of three determinants, which have the following form:

$$\begin{vmatrix} \nu q^2-\rho\omega^2 & Iq^2 \\ Iq^2 & Kq^2-iD\omega \end{vmatrix}=0, \tag{8}$$

where ν, I, and K are the effective constants of phonon elasticity, phonon-phason coupling, and phason elasticity. Phason friction D and density ρ are the same as in Eqs. (6). For the above-mentioned high-symmetry directions one can call the wave longitudinal (or transversal) if the phonon part **u** of its polarization vector is parallel (or perpendicular) to its wave vector **q**. For the longitudinal waves the coefficient ν in Eq. (8) should be changed to $\lambda+2\mu$ and for the transverse modes $\nu=\mu$. Other coefficients are presented in Table I as a function of the wave-vector direction (first column) and the wave polarization.

Vanishing of the determinant of system (8) defines simultaneously two dispersion relations $\omega(q)$ for the waves the phonon and phason types. Without taking into account the phonon-phason coupling, the first relation has the form $\rho\omega^2=\nu q^2$, and the second is given by $\omega=-iKq^2/D$. For an infinitely large value of the phason friction coefficient the phasons become frozen and their relaxation time diverges. The substitution in the matrix of system (8) of one of the corresponding solutions of system (8) permits to determine the two-component zero vector $\langle u,w\rangle$ of the above-mentioned matrix. The ratio of the zero-vector components is equal to the ratio of the phonon and the phason components of the polarization vector **U** (second and third columns in Table I, respectively). Taking into account the phonon-phason coupling leads to the phonon frequency $\omega_{phn}(q)$, which has both real and imaginary parts. The imaginary part determines the phonon damping coefficient (or phonon lifetime) in the minimal model. The phason frequency $\omega_{phs}(q)$ is purely imaginary, the phasons in the minimal model being characterized by their lifetime only.

The minimal model of the phonon-phason elastodynamics has several important physical consequences.

(1) Both the effective phonon-phason coupling and the effective phason elastic constant are anisotropic and they have an icosahedral symmetry. Acoustic branches decrease in different ways in different crystallographic directions.

(2) The doubly degenerated acoustic wave propagating along the fivefold axis does not interact with the corresponding phason mode.

(3) Taking into account the phonon-phason interaction breaks the transverse phonon degeneracy along the twofold axis.

Here it should be stressed that the observation of the above-mentioned effects corresponding to the deviation from an isotropic symmetry becomes possible if the phasons are not frozen, or in other words the phason friction D does not diverge.

III. LIMITATIONS AND FURTHER DEVELOPMENT OF THE MINIMAL MODEL

(1) The first limitation is related to a discrete atomic aperiodic structure of QC's. In the case of the continuous isotropic medium as well as in the crystalline case one can classify normal modes according to their frequencies and wave vectors. In the quasicrystalline case it is possible only at the beginning of the acoustic branches in the vicinity of the centers of Brillouin zones. For a mode with an essentially big frequency one cannot assign an exact value of the wave vector. The corresponding atomic motion presents a superposition of waves with close wave vectors. Consequently, the

finite width of the acoustic dispersion curves is defined not only by the finite lifetime of phonons [by the imaginary part of $\omega_{phn}(q)$], but also by the discrete atomic aperiodic structure of QC's. It is impossible to take this effect into account in the frame of the model developed in the continuous medium approximation. Therefore the conclusions of the minimal model can be applied only for the propagation of a sound wave with a wavelength essentially greater than the interatomic distances.

(2) Let us consider the second limitation. As it is well known[9] the propagation of the sound waves is accompanied by the appearance of areas of a local-density change. So the local temperature of these areas differs from the average temperature of a QC. Therefore to analyze the acoustic wave propagation in addition to the elastic stress we have to allow for the thermal one caused by the temperature variation. The above-mentioned mechanism leads to the medium heating and to the sound wave attenuation. For the cases of an icosahedral symmetry and of an isotropic medium, this mechanism works only for longitudinal waves.[20] It can be taken into account by the substitution of

$$\lambda = \lambda_T + \frac{\lambda_S - \lambda_T}{1 + \frac{i\chi q^2}{\omega C_E}} \tag{9}$$

into the solution of motion equations. In formula (9) χ is the coefficient of thermal conductivity, C_E is the specific heat of a unit volume at constant strain, λ_S and λ_T are elastic coefficients at constant entropy and at constant temperature, respectively.

(3) Another phenomenon that can be taken into account in the framework of the approach that we propose is a pinning effect, extensively studied experimentally and well understood for incommensurate phases. For example, in the IIC mentioned in the previous analysis this effect takes place if a relative shift of the sublattices leads to a restoring elastic force. The resulting elastic energy of this system depends not only on the vector **w** gradients but also on the vector **w** value. Usually the pinning effect is considered to be a consequence of structural imperfections in an incommensurate system. For vanishing **q** the frequency of the phason mode tends not to zero but to a finite nonzero value determined by the strength of the pinning effect. After a phase transition which makes initially incommensurate sublattices commensurate, the phason mode becomes a usual optical vibration, and the elastic constant of the pinning effect determines its frequency in the first Brullouin-zone center. Along the same line the pinning effect in QC's can be easily taken into account in our model. It can be done by a slight modification of the DM phason block (7). The same small constant f_s corresponding to the pinning effect strength should be added to all three diagonal elements of the phason block. Of course, the terms that are independent of the wave vector appear in the DM when the vector **w** value is taken into account in the elastic energy of the system. The resulting determinant (8) changes also very slightly: the constant f_s is added to the element in the second row and in the second column.

(4) Another important extention of the minimal model is obtained by taking into account in the DM (7) of the fourth- (or higher) degree terms in the wave vector. Even in the simplest model of a linear atomic chain with only one type of atoms, the acoustic dispersion branch is linear only in the vicinity of nodes in reciprocal space. It is related to the way the DM depends on the wave vector. In Ref. 21 it is shown that the terms of fourth degree distinguish already an icosahedral QC from an isotropic medium. For QC's the phonon block in the DM has one additional independent coefficient with respect to the isotropic case. It means that even in the low-temperature region where the phasons are frozen, the acoustic-phonon dispersions in an icosahedral QC are isotropic only in the long-wavelength limit. The fourth-degree terms in the DM lead to nine independent coefficients, namely, three phonon-phonon coefficients, three phason-phonon coefficients, and three phason-phason ones. To study the propagation of acoustic waves with long wave vectors, the terms of higher order in the wave vector should be taken into account in the phonon-phonon block in the DM. Modifying the minimal model in such a way we obtain several changes in determinant (8). The term $\mu' q^4 + \delta_{ef} q^4$ is added to the element in the first row and in the first column to account for the TA wave propagation, whereas the term $(\lambda' + 2\mu')q^4 + \delta_{ef} q^4$ is added to the same element in the LA wave case. The constants λ' and μ' correspond to the fourth-degree isotropic terms; the last column of Table I represents the anisotropic value δ_{ef} in the units of the second-order elasticity constant $\xi_{||}$. Then in the approximation of frozen phasons (i.e., in the low-temperature region), the second-order anisotropy of all the acoustic branches is determined by only one material constant $\xi_{||}$. Note also that when the second-order terms are taken into account the degeneracy of the TA modes propagating along twofold axes is lifted, though the modes propagating along threefold and fivefold axes remain degenerate. The corresponding frequency square splitting $\omega_1^2(q) - \omega_2^2(q)$ can be expressed as $(2/\tau + 2\tau)\xi_{||} q^4/\rho$. Table I shows that the maximal splitting induced by this effect is observed for the LA modes with the wave vectors parallel to threefold and fivefold axes. Its value is then given by $(10/9 + 6)\xi_{||} q^4/\rho$.

IV. RESONANT ANISOTROPIC ABSORPTION OF LOW-FREQUENCY ACOUSTIC WAVES

The minimal model presented in the preceding section permits to discuss quantitatively the peculiarities of acoustic-phonon dispersion in the long-wavelength limit. The sound attenuation mechanism caused by local temperature deviations [see point (2)] in the preceding section) applies for LA modes only. Therefore we start here from the analysis of TA modes. To calculate the anisotropy of the function $\omega_{phn}(q)$ it is necessary to take into account the effects associated with the isotropic relaxation of acoustic phonons. For that, the first equation of system (6) should be completed by a term due to usual viscosity. This term is determined by the tensor η_{ijkl}, which is isotropic in the case of icosahedral symmetry. The viscosity gives a contribution to the total stress value proportional to the velocity of strain: $\Delta P_{ij} = \eta_{ijkl} \dot{E}_{kl}$. Then,

after evident transformations, the form of determinant (8) is modified. Namely, for TA waves the term $-i\eta^{\perp}\omega q^2$ is added to the element in the first row and in the first column, whereas for LA waves the term $-i\eta^{||}\omega q^2$ is added to the same element. Here $\eta^{\perp}$ and $\eta^{||}$ are two independent coefficients of the viscosity tensor. Though the usual viscosity is isotropic in the case of icosahedral symmetry, the phason viscosity which gives a contribution of the same type but in the element (2,2) is anisotropic.[20] Thus in the following consideration, we analyze the solution $\omega_{phn}(q)$ of the equation

$$\begin{vmatrix} \mu q^2-\rho\omega^2-i\eta^{\perp}\omega q^2 & Iq^2 \\ Iq^2 & Kq^2-iD\omega \end{vmatrix}=0, \qquad (10)$$

where the effective coefficients K and I depend on the wave-vector (q) direction and on the polarization according to Table I. Phason friction coefficient D is considered to be a decreasing function of temperature. All other coefficients in Eq. (10) are constants. Let us first analyze the limit cases. If the phason viscosity D tends to infinity (and the temperature goes to absolute zero), then

$$\omega_{phn}=\frac{-i\eta^{\perp}q^2+\sqrt{4\rho\mu q^2-(\eta^{\perp})^2q^4}}{2\rho}. \qquad (11)$$

The anisotropy is evidently absent both in the real and the imaginary part of $\omega_{phn}(q)$. In the other limit case, the phason relaxation time is negligible or $D=0$. It results in

$$\omega_{phn}=\frac{-i\eta^{\perp}q^2+\sqrt{4\rho q^2(\mu-I^2/K)-(\eta^{\perp})^2q^4}}{2\rho}. \qquad (12)$$

In this case the anisotropy of the damping coefficient is completely absent and the sound velocity in the long-wavelength limit is expressed as

$$V=\sqrt{\frac{\mu-I^2/K}{\rho}}. \qquad (13)$$

Equations (12) and (13) are valid provided $D\to 0$ at a finite nonzero value of q. For TA modes of different symmetry, the value $\mu-I^2/K$ plays the role of an effective shear modulus.

To estimate the numerical values of corresponding quantities one can use the material constants of icosahedral QC's. For $Al_{70.3}Pd_{21.5}Mn_{8.2}$ the constants are known: $\rho=5100$ kg/m^3,[22] $\mu=0.65\times10^{11}$ N/m^2, $\lambda=0.75\times10^{11}$ N/m^2.[23] Following different estimations[24,25] we have chosen $K_1=0.81\times10^{11}$ N/m^2, $K_2=-0.42\times10^{11}$ N/m^2. However, for the moment there exists no reliable estimation of the phonon-phason coupling constant K_3, even of its order of magnitude. In Ref. 26 the phonon-phason coupling constant K_3 is estimated theoretically to be one order of magnitude smaller than other constants. By contrast, the analysis of the diffuse scattering profile in the vicinity of Bragg reflections[25] leads to the value of K_3 higher than the value of the phason elasticity constant K_1. In the well-known works of de Boissieu and co-workers also devoted to the diffuse scattering profile analysis, the constant K_3 is considered to be negligible (see, for example Refs. 27 and 28). If we take $K_3\approx0.1K_1$ then in the limit $D=0$ the difference of velocities between the TA wave propagating along a fivefold axis ($I^2/K=0$) and the TA wave propagating along a threefold axis ($I^2/K=16K_3^2/[9(K_1-2/3K_2)]$, see Table I) is about 1%. If K_3 is two orders of magnitude smaller than other constants then the difference of velocities is not more than 0.01%. For the cases with intermediate values of D it is necessary to estimate the isotropic viscosity value $\eta^{\perp}$. The ratio $\eta^{\perp}q^2/(\rho\omega/\pi)\approx\pi\omega\eta^{\perp}/\mu$ can be considered in the proposed model as a logarithmic decrement of free decay γ. Internal friction Q^{-1} is related to γ as γ/π. The value of the internal friction measured far from the resonance region (for the frequency 2000 Hz and the temperature of about 550 K) was reported to be $Q^{-1}=1.3\times10^{-4}$.[29] This yields $\eta^{\perp}=670$ N s/m^2. From Eq. (11) one can see that $q^2\ll4\rho\mu/(\eta^{\perp})^2$. Therefore the given estimation of $\eta^{\perp}$ value is valid only if $q\ll10^{-4}$ nm^{-1}. For smaller wavelength the dependence of viscosity upon the wavelength should be taken into account.

Usually the experimental data on the internal friction in a sample are presented as a dependence of $Q^{-1}(T)$. In the proposed model it is worth presenting it as a function $Q^{-1}(\ln[D])$. This choice is justified on the one hand by the fact that the most temperature-dependent quantity in the model is the phason viscosity D, and on the other hand by the variation of D in a wide range. The logarithmic scale is also justified by several physical considerations. Indeed, to analyze the relaxation time of thermally activated processes, the Arrhenius relation is often used :

$$\tau_{rel}=\tau_{\inf}\exp\left(\frac{H}{k_BT}\right),$$

where $\tau_{\inf}$ is the characteristic time constant, H is the activation enthalpy, and k_B is Boltzmann's constant. For an estimation one can assume that the phason relaxation time satisfies the similar equation, then the temperature is proportional to $\ln(D)^{-1}$. Several estimations based on the proposed model are given in Figs. 2 and 3. In all the cases the phason-phonon coupling constant was taken to be $K_3\approx0.1K_1$. We discuss first the correspondence between these figures and the available experimental data, and then propose a series of experiments which can clarify the considered phenomenon.

The internal friction is often measured at a fixed oscillator frequency. To satisfy this condition, Eq. (10) is solved for a given set of the parameters (including D) with a constraint $\mathrm{Re}(\omega_{phn})$ is equal to a constant value. From the practical point of view, Eq. (10) is solved using well-known analytical formulas for the solutions of the cubic equations. This procedure is repeated several times to choose the appropriate real value of q. Then Q^{-1} and V are calculated as $Q^{-1}=2\mathrm{Im}(\omega_{phn})/\mathrm{Re}(\omega_{phn})$ and $V=\mathrm{Re}(\omega_{phn})/q$. The coefficient 2 in the expression for the internal friction appears due to the above-mentioned relation between the internal friction and the logarithmic decrement of the free decay. At the value of the material constants used for the figures' construction, the difference between V as a function of $\ln(D)$ calculated at constant q and $V(\ln[D])$ calculated at constant oscillator frequency is very small in comparison with the scale of Figs. 2

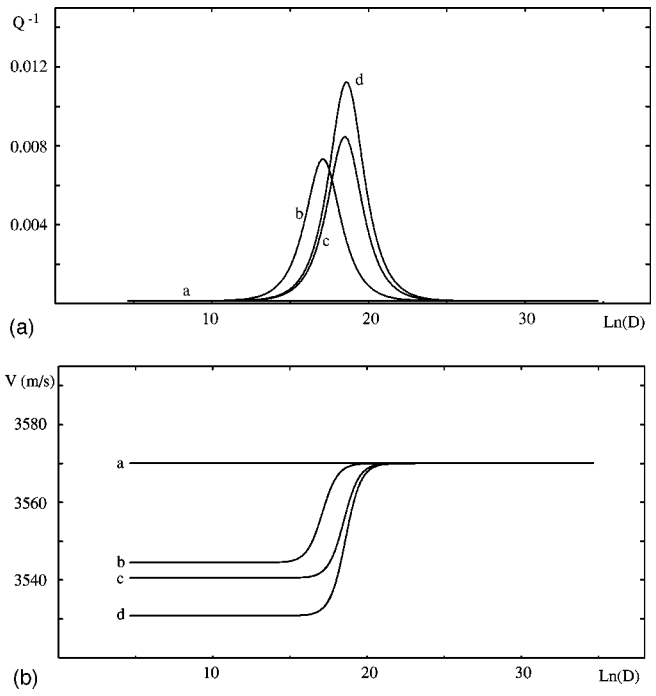

FIG. 2. (a) Internal friction Q^{-1} values as a function of natural logarithm of the phason friction coefficient for different TA modes in an icosahedral QC. Oscillation frequency is 2000 Hz. Curve *a*: Straight line, TA mode propagating along fivefold axis. Curve *b*: First TA mode propagating along twofold axis. If, for example, the wave vector of the mode is directed along [100] then the mode is polarized along [010] direction. Curve *c*: TA mode propagating along threefold axis. Curve *d*: Second TA mode propagating along twofold axis. If, for example, the wave vector of the mode is directed along [100] then the mode is polarized along [001] direction. (b) Dependence of sound velocities on natural logarithm of the phason friction coefficient for different TA modes in an icosahedral QC. Oscillation frequency is 2000 Hz. Same mode notations like in (a).

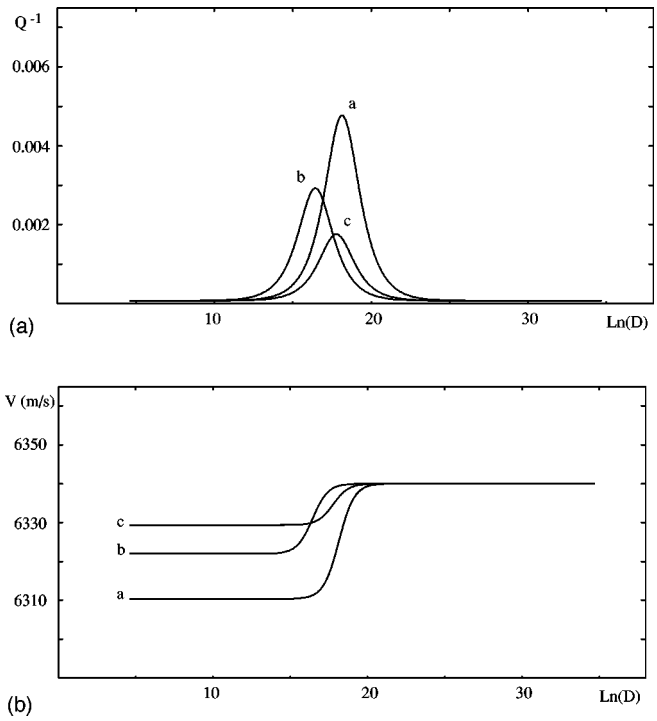

FIG. 3. (a) Internal friction Q^{-1} values as a function of natural logarithm of the phason friction coefficient for different LA modes in an icosahedral QC. Oscillation frequency is 2000 Hz. Curve *a*: LA mode propagating along fivefold axis. Curve *b*: LA mode propagating along threefold axis. Curve *c*: LA mode propagating along twofold axis. (b) Dependence of sound velocities on natural logarithm of the phason friction coefficient for different LA modes in an icosahedral QC. Oscillation frequency is 2000 Hz. Same mode notations like in (a).

and 3. The same concerns the function $Q^{-1}(\ln[D])$. This can be explained by almost linear dependence between $\mathrm{Re}(\omega_{phn})$ and q and by very small change of the sound velocity between limit cases $D=0$ and $D=\infty$. Let us stress that both the left and right parts of the dependences presented in Figs. 2(a) and 2(b) are in complete agreement with the limit cases $D=0$ and $D=\infty$ [see Eqs. (11)–(13)]. A similar analytical analysis is also possible for the limit cases of the LA modes [see Figs. 3(a) and 3(b)].

In Fig. 2(a) the ''background'' internal friction is approximately defined by the expression $\omega\eta^{\perp}/\mu$. This follows from Eqs. (11) and (12) and from the relation $\mathrm{Re}(\omega_{phn}) \approx q\sqrt{\mu/\rho}$. The analogous expression for the background internal friction of the LA modes [see Fig. 3(a)] looks like $\omega\eta^{||}/(\lambda+2\mu)$.

The maximum of the sound wave damping corresponds to the wave vector for which $\tau_{phs}\mathrm{Re}(\omega_{phn})\approx 1$, where τ_{phs} is the phason relaxation time. Using the estimations $\tau_{phs} \approx D/(Kq^2)$ and $q^2\approx[\mathrm{Re}(\omega_{phn})]^2\rho/\nu$ one can conclude that the maximal sound attenuation corresponds to a region where $D\nu\approx K\mathrm{Re}(\omega_{phn})\rho$. Using this relation one can analytically estimate the height of the absorption peak with respect to the background level as $I^2/(2K\nu)$. (As usual, for TA modes $\nu=\mu$ and for LA modes $\nu=\lambda+2\mu$.) The above analytical estimations are valid if $Q^{-1}\ll 1$. Both expressions for the peak height and for the peak position are in good agreement with the direct numerical calculations presented in Figs. 2(a) and 3(a). Unfortunately, we have not got values of some material constants that enter Eq. (9). Therefore the contribution of thermal conductivity to the attenuation of LA modes cannot be calculated. For crystalline metallic systems this contribution is not very essential. Also we have no experimental data to estimate $\eta^{||}$. In the corresponding calculation we put $\eta^{||}=\eta^{\perp}$. Let us recall that the viscosity constant variation leads mainly to the change of the background internal friction.

The experimental dependence $Q^{-1}(T)$ (Ref. 29) demonstrates two absorption peaks. The experimentally observed peak (*A*) is related to the localized atomic jumps[29] not described by the theoretical model under consideration. The authors of Ref. 29 associate the high-temperature absorption peak (*B*) with the collective atomic motions. However the data presented in Ref. 29 seem to be not sufficient to conclude if the absorption peak (*B*) height is different for the samples cut along different crystallographic directions. Therefore the experimental study of this peak may be very useful for experimental verification of the proposed model.

The values resummed in Table I permit us to conclude that in the simplest model the minimal internal friction corresponds to the transverse modes in a sample cut along the fivefold axis. In the harmonic approximation (in the case of small oscillations) the TA mode propagating along a fivefold axis is not coupled to the corresponding phason mode. As a result the resonant absorption peak is absent for the mode of this type. The absorption peak can appear, however, in a model that takes into account nonharmonic phonon-phason coupling. Possibly, the peak (B) is only partly related to the resonant phonon-phason coupling. Other collective atomic excitations (for example, the dislocation motion) can contribute to this peak. The question of whether the contributions of such excitations are anisotropic or not is still open. In any case such excitations lead to the peak broadening and change its intensity. Another possible source of the change of the peak parameters is the temperature dependence of the elastic constants that is not taken into account by the developed model. The data presented in Ref. 30 can be also compared with the conclusions of the minimal model. The measurement of internal friction at frequency 2.8 Hz at 1000 K [high-temperature edge of the peak (B)] shows that the Q^{-1} value for a sample cut along the fivefold axis is smaller than for a sample cut along the twofold axis. In addition, the authors of Ref. 30 report that in the high-temperature region, the effective shear modulus measured for the transverse modes in a sample cut along the fivefold axis is greater than the modulus measured in a sample cut along the twofold axis, the difference being equal to 2 GPa. Both these facts are in a good qualitative agreement with the results presented in Fig. 2. We think that an additional investigation of internal friction in differently orientated samples at low frequencies (less than 10 Hz) and high temperature (up to 1000 K) may be very useful to justify the contribution of the resonant phonon-phason coupling into the peak (B) origin. In this region of frequencies and temperatures, an activation of phason modes with large relaxation times τ_{inf} is expected.

Finally, let us note that if the phonon-phason coupling constant is one order of magnitude smaller than the value adopted in the previous estimation (i.e., $K_3 \approx 0.01 K_1$) then in Fig. 2(b) the maximal difference between the velocities a and d is divided by 100 and is not higher than 0.4 m/s. For the case of Fig. 3(b) this difference will be not higher than 0.2 m/s. It will be practically impossible to measure this sound velocity anisotropy experimentally. The absorption peak height with respect to the background [Figs. 2(a) and 3(a)] is also 100 times smaller in this case. However by the frequency decrease it is possible to make the background level much smaller and to resolve eventually the resonant low-frequency acoustic wave absorption induced by the phonon-phason coupling.

Also the measurement of the internal friction Q^{-1} and of the velocity V as functions of the oscillator frequency at constant temperature ($\approx$800–900 K) may be useful for the verification of the following properties of the model. In the low-frequency limit at any finite nonzero value of D the relative contribution of the term $D\omega$ into Eq. (10) is more essential. Therefore the anisotropy of sound velocities disappears and $Q^{-1} \to 0$ (similarly to the case of $D=\infty$). In the $q \to 0$ limit the phason part of the 6D polarization of the acoustic-type waves tends to zero. Indeed, in the vicinity of any fixed medium point the acoustic wave with the given amplitude and wave vector $\mathbf{q}$ produces local strain proportional to the q value. Consequently, the induced phason strain in the vicinity of the same point, being proportional to the ordinary strain, tends to zero in the long-wavelength limit. Therefore the phason effect on the sound velocities is negligible.[31] Nevertheless the finite value Q^{-1}/q is anisotropic. The ratio R between the lengths of the phason and phonon parts of the 6D polarization can be rigorously deduced from the matrix (10). For that, it is simpler to take the second line of this matrix. Then $R = Iq^2/(iD\omega_{phn} - Kq^2)$. For large enough wave vectors or high frequencies, R tends to I/K and the lattice vibration induces the corresponding phason motion.

If $\mathrm{Re}(\omega_{phn}) \gg D\nu/(K\rho)$ then the sound dispersion corresponds to Eq. (12) (similarly to the case of $D=0$). In this limit the value $\mathrm{Im}(\omega_{phn})$ is isotropic. Therefore the anisotropy of the Q^{-1} value is caused by the anisotropy of the real part of frequency. Accordingly, the phason influence is also expressed in a weak anisotropic decrease of the sound velocities. This decrease occurs essentially in the intermediate regime where $\mathrm{Re}(\omega_{phn}) \approx D\nu/(K\rho)$. The point $\omega_0 = D\nu/(K\rho)$ approximately corresponds to the maximum of $Q^{-1}(\mathrm{Re}[\omega_{phn}])$. The above-mentioned estimation for the peak maximum height with respect to the isotropic background level is still valid: $Q^{-1}_{max} \approx I^2/(2K\nu)$. The contribution of the resonant phonon-phason coupling to the dependence $Q^{-1}(\mathrm{Re}[\omega_{phn}])$ in the intermediate- and low-frequency regions is well described by the Debye function:

$$Q^{-1}(\mathrm{Re}[\omega_{phn}]) = 2Q^{-1}_{max} \frac{\mathrm{Re}(\omega_{phn})/\omega_0}{1+[\mathrm{Re}(\omega_{phn})/\omega_0]^2}.$$

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