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Elastic constants and anisotropic internal frictions of decagonal $Al_{72}Ni_{18}Co_8$ single quasicrystal at low temperatures

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We studied the complete set of elastic constants C_{ij} and internal frictions tensor Q_{ij}^{-1} of anisotropic decagonal $Al_{72}Ni_{18}Co_8$ single quasicrystal by electromagnetic acoustic resonance at low temperatures, to 5 K. Most $C_{ij}(T)$ showed usual stiffening upon cooling and their temperature behaviors are well characterized by an Einstein-lattice-vibration model. The average Grüneisen parameter estimated from the bulk modulus is 2.4, almost identical to crystalline Al. Ambient temperature Q_{ij}^{-1} show a linear relationship to the elastic-constants temperature derivatives $|dC_{ij}/dT|/C_{ij}$ with a slope of 1.28 K^{-1}, suggesting that lattice anharmonicity plays a dominant role for Q_{ij}^{-1}. The longitudinal modulus for the decagonal axis C_{33} and corresponding internal friction Q_{33}^{-1}, however, showed significant deviation from such ideal behavior, suggesting additional low-frequency phonon-excitation modes along the decagonal axis. © *2010 American Institute of Physics*. [doi:10.1063/1.3457898]

I. INTRODUCTION

Since their discovery in 1984, quasicrystals (QCs) continue to receive much study, both measurement and theory. The main problem is to relate their physical properties to their crystal structures, prohibited by traditional crystallographic theory.[1,2] Elastic constants are well-chosen physical properties for study for five principal reasons: (1) one can measure them accurately, typically with less than 1% error; (2) being a tensor property, they reflect full symmetry, stiffness, and anisotropy; (3) their imaginary part (internal friction) reveals any dissipative mechanisms; (4) they relate closely (often simply) to the interatomic potential; and (5) they correlate with a wide variety of physical-mechanical properties. Until now, a variety of QCs have been studied with fivefold rotational symmetries (icosahedral type), eightfold (octagonal type), 10-fold (decagonal type), and 12-fold (dodecagonal type).[1,3–5] Many studies report the atomistic structures of QCs from the view of higher-dimensional crystallographic theory[6,7] and transmission-electron-microscopy observations.[8,9] On the other hand, mechanical properties of QCs are also interesting issue because they include a new kind of lattice defect, called a phason, which is analogous to a dislocation in a conventional crystal and therefore governs plastic deformation. Experimental and theoretical studies on the dynamics of phasons also appear in the literature.[10,11] However, only a few studies have reported the elastic constants C_{ij} and internal friction Q^{-1} of QCs.[12–19] Part of this deficiency can be placed on available specimen size; single QCs are usually obtained in mm-size so that conventional mechanical tests cannot be used with sufficient accuracy, especially for internal-friction-tensor Q_{ij}^{-1} measurements.

The free-vibration acoustic resonance method, including both resonance ultrasound spectroscopy (RUS) and electromagnetic acoustic resonance (EMAR), is state-of-the-art for C_{ij} and Q_{ij}^{-1} measurements because it provides complete sets of them from a mm-sized single-crystal specimen.[20–25] Spoor *et al.*[12] performed precise C_{ij} measurements on isotropic icosahedral Al_6CuLi_3 QC by RUS at ambient temperature. Chernikov *et al.*[13] carried out low-temperature RUS measurements for anisotropic decagonal $Al_{71}Ni_{16}Co_{13}$ QC and reported $C_{ij}(T)$ from ambient temperature to 5 K. However, internal friction has not been reported in these studies. Agosta *et al.*[18] carried out RUS measurements for isotropic icosahedral Ti–Zr–Ni QCs and reported temperature dependence of C_{ij} and internal friction Q^{-1}. In this study, they suggested the existence of a thermal-activation process. This process could be related to phason jumps,[16] although the detailed mechanism remains unclear. To extend previous studies, it is worthwhile to study the internal-friction tensor Q_{ij}^{-1} as well as C_{ij} of other anisotropic QCs.

In the present study, we measured all independent elastic constants C_{ij} and internal-frictions tensor Q_{ij}^{-1} of anisotropic decagonal $Al_{72}Ni_{18}Co_8$ single QC by EMAR method from 5 to 297 K. We focused on the anisotropy of internal friction and its relationship with the temperature derivatives of elastic constants dC_{ij}/dT. Construction of this paper is as follows. In Sec. II, we present details of the QC specimen, elastic symmetry, and a brief explanation of the EMAR measurement method. Measurement results of C_{ij} and Q_{ij}^{-1} are presented in Sec. III. Section IV gives a theoretical analysis based on an Einstein-lattice-vibration model and quasiharmonic thermodynamics. Some concluding remarks appear in Sec. V.

II. MEASUREMENTS

A. Material and elastic symmetry

The material used in this study is decagonal-type $Al_{72}Ni_{12}Co_{16}$ single QC prepared by the Bridgman method.

It had a rectangular parallelpiped shape with dimensions $x=2.86$, $y=2.57$, and $z=2.82$ mm, where the z axis is set to be the decagonal direction within an accuracy of $\pm 1°$. Mass density was determined to be 3970.6 kg/m^3 from the dimensions and mass of the specimen.

The QC has a sequential periodic structure of quasiperiodic two-dimensional layers along the decagonal axis. Thus, it has transverse-isotropic-symmetry (or hexagonal-symmetry) elastic constants. In matrix notation, C_{ij} of the QC can be expressed as follows:

$$C_{ij} = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ C_{12} & C_{11} & C_{13} & 0 & 0 & 0 \\ C_{13} & C_{13} & C_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{66} \end{bmatrix}. \quad (1)$$

Here $C_{66}=(C_{11}-C_{12})/2$. Thus, there are five independent C_{ij}: C_{11}, C_{33}, C_{44}, C_{12}, and C_{13}. The bulk modulus B is given by

$$B = \frac{(C_{11}+C_{12})C_{33}-2C_{13}^2}{C_{11}+C_{12}-4C_{13}+2C_{33}}. \quad (2)$$

The internal-friction tensor Q_{ij}^{-1} is defined as the imaginary part of the complex elastic constants $\tilde{C}_{ij}$ (Ref. 25)

$$\tilde{C}_{ij} = C_{ij}(1+iQ_{ij}^{-1}). \quad (3)$$

Here i denotes imaginary index. Q_{ij}^{-1} also has five independent components, like the C_{ij}.

B. Low-temperature EMAR

The EMAR method determines C_{ij} and Q_{ij}^{-1} from free-vibration resonance frequencies and free-decay curves. Details of measurement procedure are presented in a monograph so that here we need only provide a brief description.[23] A specimen is inserted into a solenoidal coil in a magnetic field 0.2 T. A tone-burst current (~1 MHz in frequency and 80 μs in duration) is fed to the coil, which induces an eddy current on the surface area of the specimen. The eddy current and static magnetic field interact to generate ultrasound vibration in the specimen through the Lorentz-force mechanism. The reverse process is responsible for detection of ultrasound vibration. Frequency sweep of the input current provides a free-vibration resonance spectrum for A_g, B_{1g}, B_{2g}, and B_{3g} modes. Vibration modes are identified unambiguously from surface-displacement-distribution measurements by laser-Doppler interferometry.[26]

For low-temperature measurements, specimen and solenoid coil were set into a cryogenic chamber and we applied the magnetic field from outside the chamber. The specimen chamber was filled with He gas as a cooling medium and we carried out the EMAR measurements from 5 to 297 K for every 5 K. Temperature fluctuation of the measurements was approximately ± 1 K.[27] During the measurements, possible crossing of resonance frequencies at lower temperatures was not studied. Thermal-contraction effects were not considered.

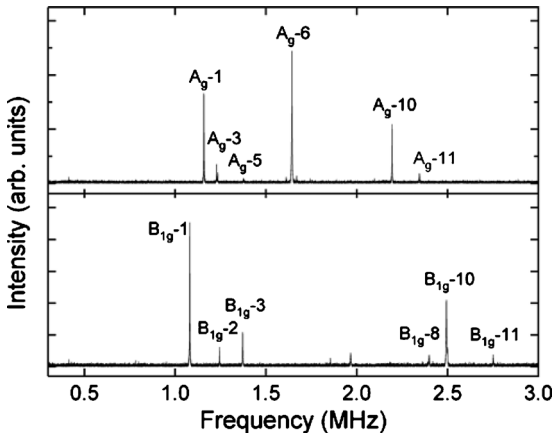

FIG. 1. A_g-group and B_{3g}-group free-vibration resonance spectra obtained by EMAR at 290 K. Indices in the figure denote vibration modes identified by surface-displacement-distribution measurements by laser-Doppler interferometry.

III. MEASUREMENT RESULTS

A. Resonance frequency and ring-down curve

Figure 1 shows A_g and B_{1g} group free-vibration resonance spectra measured separately by EMAR at 297 K. Indices in the figure represent vibration modes. Figure 2 shows a ring-down curve for the $B_{1g}-1$ mode showing free decay of ultrasound vibration amplitude with respect to time. To determine the time attenuation coefficient α_i, we fit a curve to the measurements. Internal friction of the ith mode Q_{ij}^{-1} is obtained from $Q_{ij}^{-1}=\alpha_i/\pi f_i$, where f_i represents the resonance frequency. From many Q_i^{-1}, one obtains the internal-friction tensor Q_{ij}^{-1} from algebraic calculations.

B. Elastic constants and internal frictions

Figure 3 shows temperature dependence of elastic constants obtained from the QC specimen. Ambient-temperature and low-temperature elastic constants C_{ij}, bulk modulus B, acoustic Debye temperature Θ_D are also summarized in Table I. Here, Θ_D is calculated from the following equation:[28]

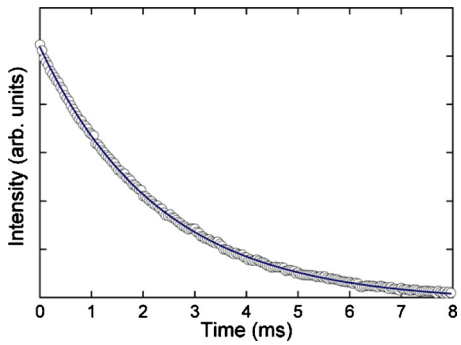

FIG. 2. (Color online) A ring-down curve for the $B_{1g}-1$ mode obtained by EMAR at 290 K. Solid line in the figure shows a least-squares fitting result of an exponential function to the measurements to determine the internal frictions of the mode Q_i^{-1}.

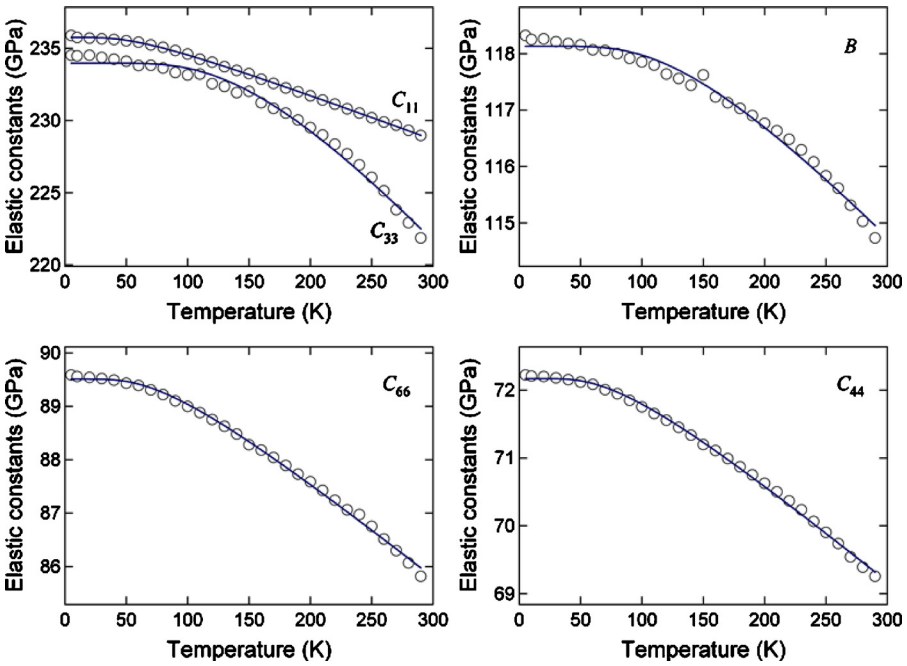

FIG. 3. (Color online) Temperature dependence of elastic constants C_{ij} and bulk modulus B of decagonal $Al_{72}Ni_{12}Co_{16}$ single QC. Open circles represent measurement points and solid curves show least-squares fitting results to Eq. (6).

$$\Theta_D = \frac{h}{k_B}\left(\frac{3N_a\rho}{4\pi m}\right)^{1/3} v_m, \tag{4}$$

where h, k_B, N_a, and m denote Planck constant, Boltzmann constant, Avogadro constant, and average atomic mass, respectively. v_m denotes the mean sound velocity (in the Debye-model sense) calculated by numerically solving the Christoffel equation.[28] The rms error between measured and calculated resonance frequencies is 0.97% throughout the temperature range, and measurement errors at 290 K are 0.6%, 0.4%, 6.0%, 3.4%, and 0.5% for C_{11}, C_{33}, C_{12}, C_{44}, and C_{66}, respectively.

As seen from the figure, all independent C_{ij} components increase monotonically as the temperature decreases. Elastic anisotropies for the diagonal components are $C_{33}/C_{11}=0.97$ and 0.99 at 290 and 5 K, and $C_{44}/C_{66}=0.81$ both at 290 and 5 K. Thus, the QC shows near isotropy in longitudinal components while shear anisotropy is significant throughout the temperature range. These temperature behaviors of C_{ij} agree quantitatively well with previous study by Chernikov *et al.*[13] Acoustic Debye temperatures obtained from pure crystalline Al, Ni, and Co are 408, 452,[28] and 441 K,[29] notably smaller than the present QC. It is worthwhile to note that mean sound velocity v_m is obtained as a reciprocal-cube average of one longitudinal-wave and two shear-wave velocities.[28] Thus, the high shear bonding within the quasiperiodic plane, C_{66}, causes the high Θ_D in the QC.

TABLE I. Elastic constants C_{ij} (GPa), bulk modulus B (GPa), and acoustic Debye temperature Θ_D (K) of the decagonal $Al_{72}Ni_{12}Co_{16}$ single QC obtained at 5 and 290 K.

Temperature (K)	C_{11}	C_{33}	C_{44}	C_{66}	C_{13}	B	Θ_D
5	235.9	234.5	72.2	89.6	61.3	118.3	612
290	229.0	221.9	69.3	85.8	59.6	114.7	(599)

Table II shows internal frictions Q_{ij}^{-1} of the QC obtained at 290 and 5 K. As seen from the table, most internal frictions Q_{ij}^{-1} take an order of 10^{-4}, which is typical in metallic systems and agrees with previous RUS work by Agosta *et al.*[18] However, only the longitudinal component to the decagonal z-axis direction, Q_{33}^{-1}, is notably high by one order of magnitude. Such a strong anisotropy of Q_{ij}^{-1} has not been observed in previous studies; we shall discuss the possible origin in the next section.

IV. DISCUSSION

A. Elastic constants and lattice anharmonicity

Let us first analyze the $C_{ij}(T)$ behaviors on the basis of an Einstein-lattice-vibration model. According to thermodynamics, internal energy E can be divided into two parts; $E=E_0+E_{vib}$, where E_0 and E_{vib} represent static and vibrational parts, respectively. For the case of an Einstein model, E_{vib} is given by[30]

$$E_{vib} = 3N\left[\frac{1}{2}h\nu_E + \frac{h\nu_E}{\exp(h\nu_E/k_BT)-1}\right]. \tag{5}$$

Here h, ν_E, k_B, and $3N$ represent Planck constant, Einstein frequency, Boltzmann constant, and degrees of freedom of the system. Leibfried and Ludwig expressed elastic constants of a system by $C_{ij}=C(1-D\varepsilon)$, where C and D are propor-

TABLE II. Internal-friction-tensor components Q_{ij}^{-1} ($\times 10^{-4}$) of the decagonal $Al_{72}Ni_{12}Co_{16}$ single QC obtained at 5 and 290 K. The longitudinal component in the z-direction, Q_{33}^{-1}, shows a notably high value, by about one order of magnitude, throughout the studied temperature range.

Temperature (K)	Q_{11}^{-1}	Q_{33}^{-1}	Q_{44}^{-1}	Q_{66}^{-1}
5	0.4	9.6	2.0	2.3
290	2.7	26.5	3.2	5.4

tionality constants and ε represents oscillator energy.[31] Inserting $E_{vib}/3N$ for ε, we obtain the following relation:

$$C_{ij} = C_{ij}(0) - \frac{s}{1 - \exp(\Theta_E/k_B T)}, \quad (6)$$

a result obtained by Varshni.[32] $C_{ij}(0)$ represents the zero-temperature elastic constants including the zero-point lattice-vibration contribution. Θ_E can be regarded as the Einstein temperature of the corresponding modes and s reflects lattice anharmonicity. Generally, a $C_{ij}-T$ curve shows linear behavior at high temperatures and a plateau below a "transition temperature" T_P. The T_P depends on average phonon energy, and quantum zero-point-vibration effects start to appear below T_P. Recently, Tarumi *et al.*[33] found a linear relationship between T_P and Θ_E; T_P=4.5Θ_E. Thus, Θ_E can be interpreted as the average phonon frequency for the corresponding C_{ij} mode.

Solid curves in Fig. 3 show least-squares-fitting results of Eq. (6) to measurement points. Equation (6) fits well to most of the $C_{ij}(T)$ measurements, while the longitudinal modulus along the decagonal direction $C_{33}(T)$ and bulk modulus show departure from typical behavior. Einstein temperatures estimated for the diagonal moduli C_{11}, C_{33}, C_{44}, and C_{66} are 170, 502, 234, and 232 K, respectively, showing a notably high value for C_{33}. Previous studies for isotropic icosahedral QCs revealed that the approximation Eq. (6) works well.[18,27] It is worthwhile to note that the icosahedral QC has a three-dimensional quasiperiodic structure. On the other hand, the quasiperiodic structure spreads only within the $x-y$ plane and the z-axis is periodic in the decagonal QC. C_{33} represents longitudinal motion about the z-axis. Equation (6) assumes a single phonon frequency for each C_{ij} because it is based on an Einstein-type lattice. Thus, the marked deviation and high Θ_E for C_{33} suggest that this assumption is not appropriate and additional longitudinal phonon modes exist along the decagonal direction, perpendicular to the transverse-isotropic plane.

We also evaluate lattice anharmonicity of the QC. Ledbetter provided a useful relationship between the temperature derivative of elastic constants dC_{ij}/dT and the mode Grüneisen parameter γ_{ij}:[34]

$$\frac{dC_{ij}}{dT} = -\frac{3k_B\gamma_{ij}(\gamma_{ij}+1)}{V_a}. \quad (7)$$

Here V_a denotes the mean atomic volume. Applying this relationship to the temperature derivative of the bulk modulus dB/dT we obtain an average Grüneisen parameter for the decagonal QC γ=2.4, nearly the same as that of crystalline Al (γ=2.0) and icosahedral $Al_{72}Pd_{19.5}Mn_{8.5}$ QC (γ=2.0).

B. Internal friction from phonon interactions

One of the dominant internal-friction mechanisms in the studied frequency range is phonon–phonon interaction. As is well known, acoustic waves disturb the equilibrium phonon density of states through phonon interactions. These interactions are irreversible processes and eventually cause energy dissipation through lattice anharmonicity. This is called the Akhieser mechanism.[35] As mentioned above, the EMAR measurement does not require mechanical contact between a specimen and an ultrasound transducer. Thus, most of the internal friction could be explained by the Akhieser mechanism.

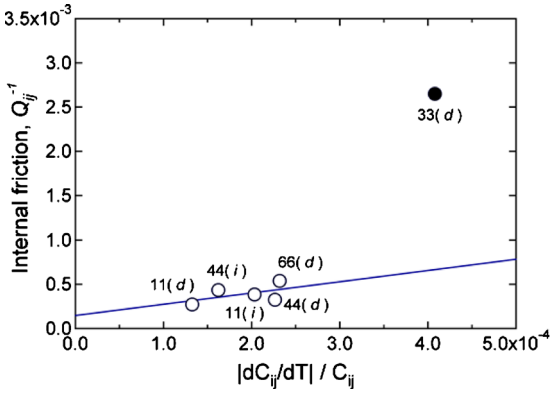

FIG. 4. (Color online) A relationship between $|dC_{ij}/dT|/C_{ij}$ and Q_{ij}^{-1}. The data plots include present decagonal $Al_{72}Ni_{12}Co_{16}$ (d) and icosahedral $Al_{72}Pd_{19.5}Mn_{8.5}$ (i), which we obtained previously (Ref. 27).

According to Mason, energy loss caused by phonon interactions is proportional to the second power of the average Grüneisen parameter, $Q^{-1} \propto \gamma^2/\rho v^2$, where ρ and v represent mass density and sound-wave velocity.[35] Ogi *et al.*[36] extended the relationship into a general form $Q_{ij}^{-1} \propto \gamma_{ij}^2/C_{ij}$. From this relation and Eq. (7), we expect a positive relationship between the temperature derivatives of elastic constants dC_{ij}/dT and internal frictions Q_{ij}^{-1}.

Figure 4 plots normalized temperature derivatives of elastic constants $|dC_{ij}/dT|/C_{ij}$ and internal friction Q_{ij}^{-1} obtained at 297 K. In the figure, we included results obtained from icosahedral $Al_{72}Pd_{19.5}Mn_{8.5}$ QC in our previous study.[27] In the figure, we see a linear relationship between $|dC_{ij}/dT|/C_{ij}$ and Q_{ij}^{-1} with the slope of 1.28 K^{-1}. However, the longitudinal component along the decagonal axis, namely $|dC_{33}/dT|/C_{33}$ and Q_{33}^{-1}, show marked deviation from the relationship. The marked correlation between $|dC_{ij}/dT|/C_{ij}$ and Q_{ij}^{-1} indicates that the Akhieser mechanism would be one of the dominant internal-friction processes in decagonal and icosahedral QCs. On the other hand, unexpectedly high Q_{33}^{-1} suggests the existence of an additional energy-dissipation mechanism. Phason motion is one of the possible mechanisms for energy dissipation. However, theoretical calculation predicts that phason motion occurs only close to the melting point in the present frequency range.[10] Also, such phason motion would contribute to in-plane Q_{ij}^{-1} components (Q_{11}^{-1} and/or Q_{66}^{-1}) because quasiperiodic structure spreads only in the $x-y$ plane. Thus, phason motion fails to explain the unusually high Q_{33}^{-1}. Another possible interpretation is weak binding of intraplane atomic motion. Such bondings would cause additional phonon excitation at low frequency and corresponding higher internal friction. Also, the additional phonon modes influence the temperature dependence of C_{33}. That is, additional phonon modes cause deviation from Einstein-model temperature behavior. To confirm the present interpretation, detailed structural analysis especially for the decagonal z-axis direction would be helpful.

V. SUMMARY AND CONCLUSIONS

In the present study, we measured the complete set of elastic constants C_{ij} and internal frictions Q_{ij}^{-1} for decagonal $Al_{72}Ni_{12}Co_{16}$ single QC from 5 to 297 K by an EMAR method. Our study produced the following principal results and conclusions:

(1) Most of the $C_{ij}(T)$ behave as expected for an Einstein-lattice-vibration model as well as our previous study for an isotropic icosahedral type $Al_{72}Pd_{19.5}Mn_{8.5}$ single QC. However, the longitudinal modulus along the decagonal axis direction $C_{33}(T)$ and bulk modulus $B(T)$ showed marked departure from expected behavior.

(2) Anisotropy of longitudinal modulus C_{33}/C_{11} is small while that of shear C_{44}/C_{66} is notable (about 20%). These tendencies do not change with temperature between 5 and 290 K.

(3) The acoustic Debye temperature Θ_D is significantly higher than those of its constitutive elements, reflecting strong interatomic bonding. Especially, strong shear bonding in the quasiperiodic $x-y$ plane accounts for the high Θ_D.

(4) The average Grüneisen parameter γ calculated from the temperature dependence of the bulk modulus is 2.4, which is close to those of crystalline Al and icosahedral $Al_{72}Pd_{19.5}Mn_{8.5}$ QC. Thus, lattice anharmonicity of the QC is similar to conventional crystalline metals.

(5) Internal frictions Q_{ij}^{-1} show usual values both at ambient and low temperatures, while Q_{33}^{-1} becomes remarkably high (by about one order of magnitude).

(6) A linear relationship between $|dC_{ij}/dT|/C_{ij}$ and Q_{ij}^{-1} suggests that the Akhieser process could be the dominant mechanisms of energy dissipation in QCs.

(7) The unusual elastic behavior and internal friction of the longitudinal mode of the decagonal z-axis direction can be explained from low-frequency phonon excitation modes caused by weak atomic bonding between the intraquasiperiodic-plane atomic motions.

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