

Density-functional theory of elastic moduli: Icosahedral quasicrystals

Marko V. Jarić

Center for Theoretical Physics, Texas A&M University, College Station, Texas 77843-4242

Udayan Mohanty

Department of Chemistry, Boston College, Chestnut Hill, Massachusetts 02167

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A recently developed density-functional method for calculating elastic moduli of crystals is generalized to include phason elasticity in incommensurate crystals. The theory is applied to hypothetical icosahedral quasicrystals based on interactions embodied in the pair correlation functions of a dense-random-packing model. It is found that the usual (phonon) elastic moduli are not dramatically different in quasicrystals and crystals. However, we show that a sufficiently strong phonon-phason coupling, which is very sensitive on the pair correlations at long wavelengths, may cause an elastic distortion with nonzero phason as well as phonon components.

I. INTRODUCTION

Ever since the experimental discovery of icosahedral quasicrystals,¹ one of the important theoretical questions has been understanding the origins of their stability or metastability. A frequently pursued objective of the theoretical investigations was the identification of these features of many-body interactions which are favorable for the formation of icosahedral structures. The two main approaches to this question are based on computer simulations and density-functional-type theories. While computer simulations are still in a preliminary stage, mostly dealing with small systems and two dimensions,^{2,3} numerous density-functional calculations⁴⁻⁹ have been reported.

Density-functional calculations can be divided into three categories. In the first category are numerous papers which sought the features favoring icosahedral ordering in the higher harmonics^{4,6} or higher-order terms⁵ in Ginzburg-Landau-type models of solidification based on the Alexander-McTague theory^{10,11} of fcc structure formation. Unfortunately, these approaches were unsuccessful: the free-energy terms which manifestly favored the icosahedral or pentagonal symmetry were always insufficient to *generically* stabilize an icosahedral structure relative to a periodic crystalline structure.^{9,12,13} No simple cause of quasicrystal stability could be identified. The best that could be done, within this framework, was to show that icosahedral quasicrystals are stable with respect to small commensurate lock-in elastic distortions.¹⁴

In the second category are the density-functional theories which, besides the density, also include an orientational order parameter. Clear evidence that a tendency for short-range icosahedral orientational ordering is an important feature which promotes not only the long-range icosahedral orientational ordering, but also the icosahedral positional ordering, was given in a density-functional theory which couples the positional and orientational order parameters.⁷ It was shown that a large

portion of the orientational phase diagram is occupied by an icosahedral phase¹⁵ and that in this portion of the phase diagram a coupling with the positional order parameter can induce an icosahedral quasicrystalline positionally ordered phase.

The density-functional theory of solidification developed by Ramakrishnan and Yusouff¹⁶ (RY) and its generalizations form the third category. The RY theory was very successful in a number of recent applications.^{8,17-36} For example, predictions based on this theory and its extensions are in excellent agreement with results of computer simulations for freezing of hard-sphere¹⁷⁻²² and Lennard-Jones liquids.²²⁻²⁴ The theory is based on a functional expansion of the interaction part of the Helmholtz potential around a reference liquid state, while the noninteracting, entropy part is treated exactly. The expansion is usually carried to second order so that the information about the interparticle interactions is embodied in the direct pair correlation function of the reference liquid.

Using the direct pair correlation functions experimentally measured for amorphous cobalt³⁷ and iron modeled by dense random packing (DRP),³⁸ the RY theory was recently generalized to investigate stability of icosahedral quasicrystals.⁸ In all cases the icosahedral structures were found more stable than the reference uniform structure, albeit less stable than the fcc or even bcc structures. Since icosahedral quasicrystalline structure cannot be stabilized for hard-sphere or Lennard-Jones correlations²² which, unlike cobalt and DRP, do not have the split second peak, a feature of the direct pair correlation function which stabilizes the icosahedral quasicrystalline order can be identified with this splitting. The split second peak is indeed a consequence of the emergence of the short-range icosahedral clustering.³⁹ A long-range icosahedral structure is, however, usually destabilized because of the frustration expressed by the presence of energetically costly, arbitrarily long wavelengths.

Like all incommensurate crystals, quasicrystals also have phase degrees of freedom (phasons). Therefore, in

addition to the elastic energy associated with the usual strains (shear and compression), one can define phason strains and elastic energies associated with them.^{40–42} Therefore, for a quasicrystalline structure to be stable, it is necessary that it is stable not only with respect to the usual (phonon) elastic distortions, but also with respect to the new, phason elastic distortions.

In this paper we calculate the static elastic-modulus tensor for icosahedral quasicrystals by extending the theory we previously developed for crystals (hereafter referred to as I).³⁵ The elastic-modulus tensor is calculated by evaluating and minimizing the RY functional of the strained solid and expanding the resulting thermodynamic potential to second order in the strain matrices. In the absence of necessary information about real quasicrystals, we applied this theory to hypothetical icosahedral quasicrystals whose structure was determined in Ref. 8 using the RY theory.

We find that the usual, phonon part of the elastic-modulus tensor for icosahedral quasicrystals is positive and not dramatically different from the elastic-modulus tensor calculated for corresponding fcc or bcc structures. Also, we find that the phason part of the elastic tensor is stable. For a sufficiently strong phonon-phason interaction term the total elastic-modulus tensor may still become unstable. Using the same DRP direct pair correlation function as in Ref. 8 we found that the interaction term is not strong enough to render the icosahedral structure elastically unstable.

However, stability of icosahedral structure with respect to the supercooled liquid, as well as its elastic stability, are sensitive to the large-wavelength behavior of the direct pair correlation function. Indeed, different interpolation schemes between the lowest experimentally available wave vectors and the zero wave vector lead to different answers, leading in one case to a sufficiently strong interaction term which causes an elastic instability with both phonon and phason components present.³⁴ As shown in Ref. 34, such elastic deformations cause distortions in the diffraction pattern which are not unlike the distortions actually observed in real samples.⁴³ The icosahedral elastic moduli calculated here were used elsewhere to illustrate a calculation of the thermal and quenched diffuse scattering from quasicrystals.⁴⁴

This paper is organized as follows. Section II contains a derivation of the RY theory for incommensurate crystals and quasicrystals. In Sec. III we derive the RY theory of elastic moduli for incommensurate crystals. The results for icosahedral quasicrystals are summarized in Sec. IV. Section V is devoted to discussion and a summary. Appendix A proves the connection between incommensurate crystals and crystals in higher dimensions. Equivalence between RY energies of incommensurate crystals and hypercrystals is proved in Appendix B. Elastic distortions of incommensurate crystals are defined in Appendix C while the elastic moduli are explicitly defined in Appendix D.

II. RY THEORY FOR INCOMMENSURATE CRYSTALS

The RY theory was developed originally to explain freezing of a liquid into a periodic, crystalline solid.

However, the approach was later adopted to deal with a variety of other problems, such as glass formation,²⁶ liquid-crystal structure,^{31,33} adsorption,²⁵ liquid-solid interface,^{29,30} crystal defects,^{27,28} elastic properties of crystals and liquid crystals,^{32–36} etc. In particular, the theory was extended to describe icosahedral quasicrystals.⁸ We shall generalize this work here and derive a RY theory of incommensurate structures.

A detailed derivation of the RY theory was presented in I and will not be repeated here. The result is that for a given chemical potential μ , volume V , and temperature T , the difference between the grand-canonical potentials G , or pressures P , of a (“solid”) state s and a reference (“liquid”) state l is given by

$$\begin{aligned} -V(P_s - P_l) &= G_s[\mu_s, V, T] - G_l[\mu_l, V, T] \\ &= \min_{n(\vec{x})} \{ \Delta W[n(\vec{x}); \Delta\mu, V, T] \}, \end{aligned} \quad (2.1)$$

where the variational potential

$$\begin{aligned} \Delta W &= \int_V \left[n(\vec{x}) \ln \left[\frac{n(\vec{x})}{n_l} \right] - \Delta n(\vec{x}) - n(\vec{x}) \Delta\mu \right] d^3x \\ &\quad - \frac{1}{2} \int_V \int_V C_l^{(2)}(n_l; \vec{x}_1 - \vec{x}_2) \Delta n(\vec{x}_1) \\ &\quad \times \Delta n(\vec{x}_2) d^3x_1 d^3x_2 + \dots \end{aligned} \quad (2.2)$$

is varied over the single particle density $n(\vec{x})$. The chemical potential and the variational potential in this expression are in units of $k_B T$, while $\Delta n(\vec{x}) = n(\vec{x}) - n_l$ and $\Delta\mu = \mu_s - \mu_l$. For a given μ_l the equilibrium density of the liquid is n_l . The equilibrium density of the solid is the density $n(\vec{x})$ for which the minimum in Eq. (2.1) is achieved.

The expansion in Eq. (2.2) is usually truncated at the second order so that the information about the interparticle interactions is limited to the direct two-point correlations $C_l^{(2)}(n_l; \vec{x})$ (the reference “liquid” state is assumed translationally invariant). This correlation is easily experimentally available from the structure factor $S_l(n_l; \vec{q})$,

$$C_l(n_l; \vec{q}) = 1 - \frac{1}{S_l(n_l; \vec{q})}, \quad (2.3)$$

where $C_l(n_l; \vec{q})$ is n_l times the Fourier transform of $C_l^{(2)}(n_l; \vec{x})$. Alternatively, C_l can be extracted from computer simulations or evaluated in approximate theories of the liquid state.

It was shown recently²² that in the case in which C_l is known as a function of n_l , rather than only for a single density n_l , an infinite subset of higher than second-order terms can be summed in Eq. (2.2) resulting in an effective second-order term where $C_l(n_l; \vec{q})$ is “renormalized” as

$$C_l(n_l; \vec{q}) \rightarrow \frac{n_l}{n_s} C_l(n_s; \vec{q}), \quad (2.4)$$

n_s being the average solid density. The terms which are summed are of the form

$$|\Delta n(\vec{q})|^2 (n_s - n_l)^m, \quad m = 0, 1, \dots, \infty. \quad (2.5)$$

Unfortunately, for the DRP model considered here the

direct pair correlation function is available only at a single density. On the other hand, for hard-sphere or Lennard-Jones systems, for which approximate functional dependence on n_l is available, we do not find icosahedral quasicrystalline equilibria.

It is important to emphasize that the RY theory described above can be generalized to the cases where the reference state or the chemical potentials are not uniform. Similarly, it is not necessary to assume that the equilibrium state is periodic. In principle, the functional minimization should give the functional form for the equilibrium solid density irrespective of whether or not it is periodic. In practice, however, one must assume a particular, sufficiently simple form and parametrization of $n(\vec{x})$. For example, one can assume that $n(\vec{x})$ is either periodic or incommensurate. If for both assumptions a minimum is found, then the structure with the lower minimum is the more stable one.

It is well known⁵ that an incommensurate structure given by a density $n(\vec{x})$ can always be represented as a cut through a higher, D -dimensional ($D > 3$) periodic density $\bar{n}(\bar{\mathbf{x}})$,

$$n(\vec{x}) = \bar{n}(\bar{\mathbf{x}}) \Big|_{\mathbf{x}^\perp = \mathbf{0}^\perp} \equiv \bar{n}(\vec{x} = \mathbf{x}^\parallel, \mathbf{x}^\perp = \mathbf{0}^\perp), \quad (2.6)$$

where we use either no superscripts or we use the superscript \parallel to indicate functions or variables in the three-dimensional physical space, while we use the superscript \perp in its complement. Functions or variables in the hyperspace are denoted by an overbar and carry no superscripts. Vectors in the physical space will also be denoted using the usual \vec{x} notation. Thus $\vec{x} \equiv \mathbf{x}^\parallel$. This one-to-one correspondence between the periodic densities in the hyperspace and the incommensurate densities in the physical space is proved in Appendix A.

Minimization in Eq. (2.1) can be carried out assuming a quasiperiodic density defined by Eq. (2.6), or by its Fourier-series expansion [cf. Appendix A, Eqs. (A1) and (A5)]

$$n(\mathbf{x}^\parallel) = \sum_{\bar{\mathbf{Q}}} \bar{n}(\bar{\mathbf{Q}}) e^{i\bar{\mathbf{Q}} \cdot \mathbf{x}^\parallel}, \quad (2.7)$$

where the sum is over the reciprocal hyperlattice, and $\bar{n}(\bar{\mathbf{Q}})$ is the $\bar{\mathbf{Q}}$ th Fourier coefficient of $\bar{n}(\bar{\mathbf{x}})$. However, in this case it is more convenient to rewrite Eq. (2.1) in terms of an equivalent minimization in the hyperspace,

$$\frac{\Delta G}{V} = \frac{\Delta \bar{G}}{\bar{V}} = \min_{\bar{n}(\bar{\mathbf{x}})} \left\{ \frac{\Delta \bar{W}[\bar{n}(\bar{\mathbf{x}}); \Delta \mu, \bar{V}, T]}{\bar{V}} \right\}, \quad (2.8)$$

where

$$\begin{aligned} \frac{\Delta \bar{W}}{\bar{V}} &= \frac{1}{\bar{V}} \int_{\bar{V}} \bar{n}(\bar{\mathbf{x}}) \ln \left[\frac{\bar{n}(\bar{\mathbf{x}})}{n_l} \right] - \Delta \bar{n}(\bar{\mathbf{x}}) - \bar{n}(\bar{\mathbf{x}}) \Delta \mu \Big| d^D \bar{\mathbf{x}} \\ &\quad - \frac{1}{2\bar{V}} \int_{\bar{V}} \int_{\bar{V}} \bar{C}_l^{(2)}(n_l; \bar{\mathbf{x}}_1 - \bar{\mathbf{x}}_2) \Delta \bar{n}(\bar{\mathbf{x}}_1) \\ &\quad \times \Delta \bar{n}(\bar{\mathbf{x}}_2) d^D \bar{\mathbf{x}}_1 d^D \bar{\mathbf{x}}_2 + \dots \end{aligned} \quad (2.9)$$

or, using the Fourier-series decomposition of $\bar{n}(\bar{\mathbf{x}})$,

$$\begin{aligned} \frac{\Delta \bar{W}}{\bar{V}} &= \frac{1}{\bar{v}_c} \int_{\bar{v}_c} \left[\bar{n}(\bar{\mathbf{x}}) \ln \left[\frac{\bar{n}(\bar{\mathbf{x}})}{n_l} \right] - \Delta \bar{n}(\bar{\mathbf{x}}) - \bar{n}(\bar{\mathbf{x}}) \Delta \mu \right] d^D \bar{\mathbf{x}} \\ &\quad - \frac{1}{2n_l} \sum_{\bar{\mathbf{Q}}} \bar{C}_l(n_l; \bar{\mathbf{Q}}) |\Delta \bar{n}(\bar{\mathbf{Q}})|^2 + \dots, \end{aligned} \quad (2.10)$$

where \bar{v}_c denotes the unit hypercell. As we prove in Appendix B, the first equality in Eq. (2.8) (in the limit $V \rightarrow \infty$ and $V^\perp \rightarrow \infty$) requires that the chemical potentials and the liquid density in the hyperspace are equal to the corresponding quantities in the physical space. Also, the pair correlation function $\bar{C}_l(\bar{\mathbf{Q}})$ must be defined in terms of the physical $C_l(\bar{\mathbf{Q}})$ as

$$\bar{C}_l(n_l; \bar{\mathbf{Q}}) \equiv C_l(n_l; \mathbf{Q}^\parallel), \quad (2.11)$$

or, equivalently,

$$\bar{C}_l^{(2)}(n_l; \bar{\mathbf{x}}) \equiv C_l^{(2)}(n_l; \mathbf{x}^\parallel) \delta^\perp(\mathbf{x}^\perp). \quad (2.12)$$

The expression Eq. (2.10) has the same analogous form as the expression Eq. (2.2) would have for a periodic density in the physical space,

$$\begin{aligned} \frac{\Delta W}{V} &= \frac{1}{v_c} \int_{v_c} \left[n(\vec{x}) \ln \left[\frac{n(\vec{x})}{n_l} \right] - \Delta n(\vec{x}) - n(\vec{x}) \Delta \mu \right] d^3 \mathbf{x} \\ &\quad - \frac{1}{2n_l} \sum_{\bar{\mathbf{Q}}} C_l(n_l; \bar{\mathbf{Q}}) |\Delta n(\bar{\mathbf{Q}})|^2 + \dots \end{aligned} \quad (2.13)$$

Either in the case of a periodic or an incommensurate structure, one may truncate the Fourier series at a finite number of terms and consider the corresponding Fourier coefficients as variational parameters. However, even near the melting temperature, atoms in a solid are fairly well localized, to within several percent of the typical interatomic separation, so that a large number ($\sim 10^3 - 10^4$) of the Fourier coefficients must be kept. Therefore, in periodic structures it is often more accurate and much simpler to expand the density into a sum of Gaussians centered at the ideal atomic positions. For example, an arbitrary monatomic crystal structure, with N_c sites per primitive lattice cell and with an equilibrium density of vacancies, can be parametrized by $3N_c + 10$ parameters.²²

In the case of incommensurate quasicrystals, a finite-temperature Gaussian smearing of the ideal atomic positions may not be sufficient. Following Eq. (2.6), the ideal atomic sites of a three-dimensional incommensurate crystal can be represented by $(D - 3)$ -dimensional nonintersecting atomic surfaces periodically decorating the hypercrystal. Smearing of these surfaces in the physical, parallel directions reflects, as in the case of ordinary crystals, low-energy, long-wavelength phonon excitations. However, incommensurate crystals may also have low-energy, long-wavelength phason excitations which would, at sufficiently high temperature, smear the surfaces also in the complementary, perpendicular directions. Such smearing corresponds to introducing partial occupancy of atomic sites in the physical space.

For example, the surfaces for the usual, modulated incommensurate crystals are continuous and unbounded in the complementary space, and gapless phason excitations exist at $\vec{q} = 0$. Note that although the displacement field

associated with the phasons can be obviously described as a linear combination of the phonons, such a representation would require phonons with arbitrarily large \bar{q} . Consequently, phonons and phasons can be considered as independent excitations at small \bar{q} .

On the other hand, if the atomic surfaces are bounded, as in the case of quasicrystals, then small fluctuations in the \mathbf{x}^\perp directions do not correspond to small atomic displacements in the real space. Instead, atomic rearrangements, possibly at arbitrarily large scale, are necessary.⁴⁵ Such rearrangements may involve large energy barriers. In this case, phasons are pinned at low temperatures and the smearing is not dynamic but due to quenched random distribution of phasons. Quasicrystals, such as icosahedral quasicrystals, are a special class of pinned incommensurate crystals in which the atomic surfaces are bounded (discontinuous) due to the symmetry-imposed constraints.⁴⁶ Nevertheless, one expects that phasons are unpinned at sufficiently high temperatures. Therefore, the atomic surfaces are smeared in the \mathbf{x}^\perp directions at any temperature due to either quenched or thermalized phasons. As we shall see in the following section, this conclusion is at least self consistent in the sense that the elastic energy calculated using it is indeed quadratic in the phonon and phason strains.

In analogy to the Gaussian expansion of crystal density, a hypercrystal density can be expanded in terms of Gaussian-smearred surfaces. In the simplest case, with a single Gaussian, analogous to the case of a primitive monatomic crystal, this amounts to the ansatz

$$\bar{n}(\bar{Q}) = n_s \frac{\bar{n}_0(\bar{Q})}{n_{0s}} e^{-(1/4)\bar{Q}\cdot(\bar{A}\cdot\bar{\Gamma}_0\cdot\bar{A})\cdot\bar{Q}} \quad (2.14)$$

where $\bar{\Gamma}_0$ is the Gaussian width matrix and $\bar{n}_0(\bar{Q})$ is the Fourier amplitude for the ideal, unsmeared hypercrystal with the average density $n_0 \equiv \bar{n}_0(\bar{0})$ and the hyperlattice-constant matrix \bar{A} (see Appendix B). The prefactor must also be determined by the minimization because an equilibrium density of vacancies (or interstitials) is possible.⁴⁷ Note that in the case of a primitive, monatomic crystal, $n_0(\bar{Q})$ is independent of \bar{Q} so that $n_0(\bar{Q})/n_{0s} = 1$. This is not true for quasicrystals. However, following Ref. 8, we shall absorb the prefactor $\bar{n}_0(\bar{Q})/n_{0s}$ into an effective Gaussian factor $\bar{\Gamma}$,

$$\bar{n}(\bar{Q}) = n_s e^{-(1/4)\bar{Q}\cdot(\bar{A}\cdot\bar{\Gamma}\cdot\bar{A})\cdot\bar{Q}} \quad (2.15)$$

or

$$\bar{n}(\bar{x}) = \frac{n_s}{(\pi^D \det \bar{\Gamma})^{1/2}} \sum_{\bar{R}} e^{-(\bar{x}-\bar{R})\cdot(\bar{A}\cdot\bar{\Gamma}\cdot\bar{A})^{-1}\cdot(\bar{x}-\bar{R})}. \quad (2.16)$$

A more general case is straightforward but tedious.

Assuming narrow Gaussians, the entropy term in Eqs. (2.9) and (2.10) can now be easily evaluated by neglecting the overlaps between different Gaussians (see Appendix B). Therefore, the variational potential Eq. (2.10) becomes

$$\begin{aligned} \frac{\Delta \bar{W}}{n_i \bar{V}} &= \Delta \bar{w}_G(\xi, \bar{B}, \bar{\Gamma}; \Delta\mu) \\ &\equiv \xi \left[\ln \left[\frac{\xi}{\pi^{D/2} \det \bar{\Gamma}^{1/2}} \right] - \frac{D}{2} \right] - (\xi - 1) - \xi \Delta\mu \\ &\quad - \frac{1}{2} C_l(\bar{0})(1 - 2\xi) - \frac{1}{2} \xi^2 \sum_{\bar{M} \in \mathbb{Z}^D} C_l(|\mathbf{B}\cdot\bar{M}|) h_M(\bar{\Gamma}), \end{aligned} \quad (2.17)$$

where $\xi = n_s/n_l$,

$$h_M(\bar{\Gamma}) = e^{-2\pi^2 \bar{M}\cdot\bar{\Gamma}\cdot\bar{M}} = e^{-(1/2)\bar{Q}\cdot(\bar{A}\cdot\bar{\Gamma}\cdot\bar{A})\cdot\bar{Q}}, \quad (2.18)$$

the reciprocal quasilattice matrix \mathbf{B} , which generates the reciprocal quasilattice vectors $\bar{Q} = \mathbf{B}\cdot\bar{M}$, and the reciprocal hyperlattice matrix \bar{B} , which generates the reciprocal hyperlattice vectors $\bar{Q} = \bar{B}\cdot\bar{M}$, $\bar{M} \in \mathbb{Z}^D$, are defined in Appendix A, Eqs. (A2) and (A4).

The physical and the complementary space will generally transform under different symmetry groups so that the Gaussian width matrix will split into two independent components. For icosahedral symmetry each of these components as well as \bar{A} and \bar{B} are proportional to the identity. That is, $\det \bar{\Gamma} = (\Gamma^\parallel \Gamma^\perp)^3$ and

$$h_M(\bar{\Gamma}) = e^{-2\pi^2(\Gamma^\parallel \mathbf{M}^{\parallel 2} + \Gamma^\perp \mathbf{M}^{\perp 2})} = e^{-(a^2/2)(\Gamma^\parallel \mathbf{Q}^{\parallel 2} + \Gamma^\perp \mathbf{Q}^{\perp 2})}, \quad (2.19)$$

where $\Gamma^\parallel, \Gamma^\perp$ are scalars. Results obtained using this equation and the DRP direct pair correlation function will be described in Sec. IV.

III. ELASTIC MODULI

We derived in I a theory for calculating elastic moduli of periodic crystals. This theory took into account a relaxation on the unit-cell scale of the strained density as well as a change in the equilibrium density of vacancies. In this paper we shall assume to be in the regime where the microscopic strain is equal to the macroscopic strain, and we shall assume that no relaxation occurs. The latter assumption is justified by our observation that the corrections due to relaxation are negligible in fcc and bcc crystal structures obtained from the DRP correlation function.

In principle, density of the strained quasicrystal should be obtained by the functional minimization over the densities whose average is restricted to the average density

$$n_{es} = \frac{n_s}{\det(\bar{\Gamma} + \bar{\epsilon})} \quad (3.1)$$

and with the reciprocal lattice

$$\bar{B}_\epsilon = (\bar{\Gamma} + \bar{\epsilon})^{-1} \cdot \bar{B} \iff \bar{Q}_\epsilon = (\bar{\Gamma} + \bar{\epsilon})^{-1} \cdot \bar{Q}, \quad (3.2)$$

where the strain matrix $\bar{\epsilon}$ has the following form:

$$\bar{\epsilon} = \begin{bmatrix} \epsilon^{\parallel\parallel} & 0 \\ \epsilon^{\perp\parallel} & 0 \end{bmatrix}. \quad (3.3)$$

As we show in Appendix C, a quasicrystal structure is in-

dependent of the $\varepsilon^{\perp\perp}$ and $\varepsilon^{\parallel\parallel}$ components so that they have been taken equal to zero.

In practice, however, we shall assume that the strained quasicrystal density has a more restricted form,

$$n_\varepsilon(\mathbf{x}^{\parallel}) = \frac{1}{\det(\bar{\mathbf{I}} + \bar{\varepsilon})} \sum_{\bar{\mathbf{Q}}} \bar{n}(\bar{\mathbf{Q}}) e^{i\bar{\mathbf{Q}} \cdot \mathbf{x}^{\parallel}}. \quad (3.4)$$

Therefore, a strained quasicrystal can also be viewed as the original $\mathbf{x}^{\perp} = \mathbf{0}$ cut, now taken through the strained hypercrystal. Consequently, by using a formulation of the theory in the hyperspace, cf. Eq. (2.17), we can apply the formulas derived in I for the elastic moduli of crystals. For example, the elastic energy is quadratic in the strain ε ,

$$\frac{E_\varepsilon}{V} = \frac{\bar{E}_\varepsilon}{\bar{V}} = \frac{1}{2} \bar{\varepsilon} : \bar{\mathbf{C}} : \bar{\varepsilon} + O(\bar{\varepsilon}^3), \quad (3.5)$$

where the elastic modulus tensor is

$$\begin{aligned} \frac{\bar{C}_{ijkl}}{k_B T n_l} &= \delta_{ij} \delta_{kl} \left[\frac{\kappa_T^{-1} + 2 \Delta P}{k_B T n_l} + 1 - \xi \right] \\ &\quad - \frac{\xi^2}{2} \sum_{\bar{\mathbf{Q}} (\neq \bar{\mathbf{0}})} \left[\frac{\ddot{C}_l}{|\bar{\mathbf{Q}}^{\parallel}|^2} - \frac{\dot{C}_l}{|\bar{\mathbf{Q}}^{\parallel}|^3} \right] \\ &\quad \times h_M(\Gamma) \bar{Q}_i \bar{Q}_j \bar{Q}_k \bar{Q}_l. \end{aligned} \quad (3.6)$$

This expression follows from Eq. (I.4.29) where $\Delta\alpha = \Delta\Gamma$ is set equal to zero, and Eqs. (I.4.30) and (I.C18)–(I.C21). Clearly, only $C^{\parallel, \parallel; \parallel, \parallel}$, $C^{\parallel, \parallel; \perp, \perp} = C^{\perp, \perp; \parallel, \parallel}$, and $C^{\perp, \perp; \perp, \parallel}$ components of the tensor $\bar{\mathbf{C}}$ are the physically relevant ones.

In analogy to the dynamical situation investigated in Ref. 42, it is interesting to examine the effect of phase degrees of freedom on the usual elastic response of a quasicrystal measured, for example, by applying a constant stress σ . If the phase degrees of freedom are sluggish, having long-response times, then at short times the quasicrystal distorts like an ordinary solid with the elastic-modulus tensor $\mathbf{C} \equiv C^{\parallel, \parallel; \parallel, \parallel}$. However, at long times, after the phase strain has adjusted to follow the usual strain, the correct strain-stress relationship is

$$\sigma = C_{\text{eff}} : \varepsilon, \quad (3.7)$$

so that the quasicrystal responds as an ordinary solid with the elastic-modulus tensor

$$\begin{aligned} C_{\text{eff}} &\equiv C_{\text{eff}}^{\parallel, \parallel; \parallel, \parallel} \\ &= C^{\parallel, \parallel; \parallel, \parallel} - C^{\parallel, \parallel; \perp, \perp} (C^{\perp, \perp; \perp, \perp})^{-1} C^{\perp, \perp; \parallel, \parallel}. \end{aligned} \quad (3.8)$$

In the following section we shall apply Eqs. (3.6) and (3.8) to icosahedral quasicrystals. Presence of such a high symmetry considerably simplifies the sum in Eq. (3.6). The resulting expressions have been derived elsewhere.⁴⁸ However, for the sake of completeness we summarize them in Appendix D.

IV. ICOSAHEDRAL QUASICRYSTALS

Short-range icosahedral orientational ordering is particularly prominent in metallic glasses. The characteristic split second peak in the structure factor is considered

a signature of this ordering.^{49,39} On the other hand, as we mentioned in the introduction, short-range orientational ordering can become long range and even trigger a long-range positional ordering. Since amorphous cobalt and amorphous iron both show strong signs of short-range icosahedral ordering, they are good candidates for exploring the interplay between local icosahedral clustering and icosahedral quasicrystalline ordering.

Experimental data for the structure factor of amorphous cobalt³⁷ and iron (modeled by a DRP structure)³⁸ were recently used in a RY calculation of fcc and bcc crystal structures and simple icosahedral (si), body-centered icosahedral (bci), and face-centered icosahedral (fci) quasicrystal structures.⁸ The structure factor of the amorphous phase was assumed weakly temperature dependent and equal to the structure factor of the reference supercooled liquid at the glass transition. The supercooled liquid was also assumed incompressible and the structure factors at small q were linearly extrapolated to zero. As a result, it was found that bci and fci quasicrystal structures are never more stable than the reference liquid. The si, bcc, and fcc structures were found progressively more stable than the liquid, with fcc the most stable and si the least stable of the three.

In this paper we focused our attention on the DRP model of the supercooled reference liquid. The DRP structure factor is in very good agreement with experimental results for amorphous iron.³⁸ The agreement is somewhat worse but still very good for amorphous nickel.³⁸ We obtained $S_l(q)$ by digitizing the curve given in Fig. 8(a) of Ref. 38. Since in this paper we are interested in calculating the elastic moduli, we cannot use the assumption of incompressible liquid, that is $S_l(0) = 0$, which would lead to an incompressible solid. As a consequence, we used the linear extrapolation of the DRP structure factor to the value

$$S_l(0) = \kappa_T n_l k_B T \approx 0.004. \quad (4.1)$$

Compressibility κ_T of the supercooled liquid at the glass transition ($T_g \approx 600$ K) (Ref. 50) was estimated using the bulk modulus of crystalline Fe at this temperature, $B \approx 1.5 \times 10^{12}$ ergs/cm³, while the density was similarly estimated to $n_l \approx 8.0 \times 10^{22}$ cm⁻³.

As in Ref. 8, our calculations were done for $\mu_s = \mu_l$. We first repeated the calculations of Ref. 8 using our digitized $S(q)$ and Eq. (4.1). We found the same relative stability of phases as in Ref. 8, although a small difference in the equilibrium values of the parameters, and a somewhat larger difference in the equilibrium values of the grand potential was present. For example, for fcc iron (DRP) we find $\Delta G \approx -4.74 n_s k_B T$, $a \approx 3.46$ Å, and $\Gamma = 2.96 \times 10^{-4}$, while $\Delta G \approx -4.84 n_s k_B T$, $a \approx 3.48$ Å, and $\Gamma \approx 2.82 \times 10^{-4}$ are quoted in Ref. 8. This difference is unrelated to a change in $S_l(0)$ since ξ is assumed equal to one. We ascribe them to small differences in digitizing the $S_l(q)$ curve and in different interpolation schemes for the values between the digitized points. We used quadratic, symmetrized interpolation.

We also performed calculations where we relaxed the condition $\xi \equiv n_s/n_l = 1$. We found the equilibrium fcc and bcc structures with $\Delta G \approx -5.8 n_s k_B T$, $\xi = 1.09$,

TABLE I. All entries are in units $10^3 k_B T n_s$. Rows one and two give the elastic moduli for fcc and bcc structures (Voigt notation). The numbers in parentheses give the *reduction* caused by the relaxation of the Gaussian widths under the strain. Third row gives the eigen moduli for the icosahedral quasicrystal. The first three moduli give the short-time response of the phonon strain to the usual stress. The numbers in parentheses give the reduction due to the long-time phason relaxation. The last row gives the eigenmodulus of the reference supercooled liquid molded by the dense random packing. The eigenmoduli of cubic iron and nickel near 600 K are given in parentheses as a reference since DRP is an excellent model for the structure of amorphous iron and nickel. The second column gives the pressure relative to the pressure of the reference liquid.

	ΔP	$C_{11} + 2C_{12} = \Lambda_1$	$C_{11} - C_{12}$	C_{44}	Λ_4	Λ_{5+}	Λ_{5-}
fcc	5.8×10^{-3}	8.27(0.02)	1.53(0.01)	2.00			
bcc	1.6×10^{-3}	3.33(0.03)	1.35(0.08)	0.42			
si	9.5×10^{-4}	1.30	0.52(0.04)	0.26(0.02)	0.14	0.54	0.16
DRP(Fe,Ni)	$0.0 \times 10^{-\infty}$	0.75(0.75)	(0.13)	(0.17)			

$a \approx 3.47 \text{ \AA}$, $\Gamma \approx 1.62 \times 10^{-4}$, and $\Delta G \approx -1.7 n_s k_B T$, $\xi \approx 1.05$, $a \approx 2.92 \text{ \AA}$, $\Gamma \approx 2.60 \times 10^{-4}$, respectively. Results quoted in Table I correspond to these values.

Among the icosahedral structures, we only examined the vertex, simple icosahedral (si) quasicrystalline structure. We found an equilibrium $\Delta G \approx -0.95 n_s k_B T$ at $\bar{a} = 1.36 \text{ \AA}$, $\Gamma^{\parallel} \approx 2.8 \times 10^{-3}$, and $\Gamma^{\perp} \approx 4.9 \times 10^{-2}$ which are again close to $\xi = 1$ values $\Delta G \approx -1.26 n_s k_B T$, $\bar{a} \approx 1.36 \text{ \AA}$, $\Gamma^{\parallel} \approx 2.3 \times 10^{-3}$, and $\Gamma^{\perp} \approx 4.7 \times 10^{-2}$ reported in Ref. 8.

The results of our calculations of the elastic moduli for structures described above are summarized in Table I. Using the Voigt notation, we list for cubic crystals the three eigen moduli: $3B = C_{11} + 2C_{12}$, $C_{11} - C_{12}$, and C_{44} , where B is the bulk modulus. These moduli can be also defined for short-time and long-time response of the icosahedral quasicrystal to externally applied stress. However, of these three moduli, only $3B$ remains an eigenmodulus also in the quasicrystal. The four eigenmoduli of the quasicrystal are defined in Appendix D and listed in Table I.

For crystalline structures, we calculated the elastic moduli with and without relaxation of the Gaussians. It is evident from Table I that the relaxation affects the moduli by only a few percent. This justifies neglecting such relaxation in more complicated calculations for icosahedral quasicrystals.

V. CONCLUSION

Besides developing a theory for calculating elastic moduli of incommensurate crystals, an important objective of this paper was to gain insights into differences and similarities between related crystalline and quasicrystalline structures. Therefore, a quick glance at Table I reveals that the usual elastic moduli of quasicrystals are significantly lower than the same moduli in crystals for either short- or long-time response. However, this result might be largely a manifestation of the fact that in our example the icosahedral structure was less stable than the crystal structure, so that elastic moduli were evaluated at a lower pressure.

Indeed, crystalline moduli calculated here are almost a factor of 10 greater than the moduli measured in cubic iron and nickel. We believe that this large difference reflects less inaccuracy of the elastic moduli at $\Delta P = 0$

than incorrect prediction of their pressure dependencies, notwithstanding indications that higher-order correlations which are usually neglected in Eq. (2.2) can be important.³⁴ Therefore, it is important that a future work repeats our calculations with nonzero values of $\Delta\mu$, so that the elastic moduli at $\Delta P = 0$ can be determined.

A future work should also examine the role of the relaxation of $\bar{\Gamma}$ on the values of the elastic moduli. Although such relaxation has a small effect in crystal structures, indicating a small contribution from Γ^{\parallel} relaxation, it is possible that a relaxation of Γ^{\perp} would have a larger effect.

Clearly, the four icosahedral eigenmoduli shown in Table I are positive and, consequently, the examined icosahedral structure is locally stable with respect to either small elastic displacements or phase distortions. However, it is important to emphasize that stability of quasicrystalline structures is very sensitive on the small- q extrapolation of the reference structure factor. Unfortunately, a quasicrystalline density necessarily has components at arbitrarily small values of q , precisely where the contribution to the variational potential is the least favorable, and where the experimental data are not available. For example, if we want to mimic flatness of the structure factor at small q , we can extrapolate the structure factor smoothly to the line $S_l(q) = S_l(0)$. Such an extrapolation is particularly unfavorable for a quasicrystal structure and we find that the icosahedral structure becomes elastically unstable for cobalt,³⁴ while it cannot be found at all for iron.

Therefore, for a successful application of the above theory it is essential that the compressibility and the small- q behavior of the structure factor are well known for the reference liquid. Lack of this knowledge is one of the main reasons the theory has not been applied to real quasicrystals. An additional reason is that the convergence of the reciprocal space sum in Eq. (2.17) is usually slow, requiring $S_l(q)$ at rather large q 's, often beyond experimentally introduced truncations. The data for amorphous cobalt also suffer from this drawback and corresponding results are consequently omitted from this paper.

In view of the above cautionary remarks, it is natural to consider formation of the icosahedral structure from the hard-sphere or Lennard-Jones liquids for which the structure factor is known with a high degree of accuracy.

Unfortunately, we could not find any icosahedral quasicrystalline solutions of Eq. (2.8). However, it might be helpful to add a degree of realism by considering a multicomponent system. A judiciously chosen mixture, with a strong tendency for icosahedral clustering, could promote a long-range icosahedral positional ordering. The theory presented here can be easily generalized to deal with such multicomponent systems.

In this paper we were mainly interested in a comparison between the elastic moduli of crystals and quasicrystals. Therefore, it was important that the calculations for both types of structures were done with the identical set of assumptions, while a possible influence of the higher-order terms neglected in Eq. (2.2) was of secondary importance. Clearly, there are no obstacles in extending our theory to include arbitrary higher-order terms.

We have circumvented the question of phason pinning and phason dynamics by defining the elastic energy as the difference between the thermodynamic potentials of the static equilibrium strained and unstrained quasicrystals. To consider the strained structure as resulting from an actual "continuous" distortion of the unstrained structure it is necessary to assume a sufficiently slow rate of distortion and a sufficiently high temperature. Plausibility of this assumption and a more microscopic analysis of the phase degrees of freedom at low temperatures will be presented elsewhere.⁵¹

In summary, we presented a method for calculating elastic moduli of incommensurate crystals. The method can be easily generalized to include higher-order correlations as well as multicomponent structures. A comparison with existing experimental results for crystals indicate that additional calculations with $\Delta\mu \neq 0$, and including some higher-order correlations, will have to be carried out. Elastic moduli of icosahedral quasicrystals are found to be smaller than the related moduli of crystals, but again additional calculations are necessary. Values of the elastic moduli and the elastic stability of the quasicrystals are very sensitive to both the small- q extrapolation and the large- q cutoff of the reference structure factor $C_l(q)$. Generally, a sufficiently strong phonon-phason interaction λ_i will destabilize the quasicrystalline structure. The instability occurs precisely at the same point, $\lambda_i = \lambda_{ij} \lambda_j^{\frac{1}{2}}$, where the effective, long-time shear modulus of the quasicrystal becomes equal to zero. Measurements of the short- and long-time bulk and shear moduli of the quasicrystal are not sufficient to determine all the moduli of the quasicrystal. Debye-Waller factors and diffuse scattering⁴⁴ offer an indirect means of measuring these moduli.

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APPENDIX A: HYPERCRYSTALS AND INCOMMENSURATE CRYSTALS

In this section we shall establish a connection between incommensurate crystals in three dimensions and periodic structures (hypercrystals) in higher-dimensional spaces.

By definition, density of an incommensurate quasicrystal $n(\vec{x})$, although not periodic, has a discrete Fourier transform. In other words,

$$n(\vec{x}) = \sum_{\vec{Q}} n(\vec{Q}) e^{i\vec{Q}\cdot\vec{x}}, \quad (\text{A1})$$

where the summation is over a lattice $\{\vec{Q}\}$ of rank $D > 3$ ($D=3$ corresponds to the case of ordinary crystals). Therefore, there exists the smallest set of basis vectors \vec{b}_α , $\alpha=1, 2, \dots, D$, such that every scattering vector \vec{Q} is an *integral* linear combination of these basis vectors,

$$\vec{Q} = \mathbf{B} \cdot \vec{\mathbf{M}}, \quad \vec{\mathbf{M}} \in \mathbb{Z}^D, \quad (\text{A2})$$

where $\text{Rank} \mathbf{B} = 3$ and $\mathbf{B} \equiv (\vec{b}_1, \dots, \vec{b}_D)$. Clearly, for a *given* basis $\{\vec{b}_\alpha\}_{\alpha=1}^D$, the Fourier coefficient $n(\vec{Q})$ is a function of $\vec{\mathbf{M}}$ only, $n(\vec{Q}) \equiv n_{\vec{\mathbf{M}}}$. Therefore, if a basis \vec{b}_α of a D -dimensional reciprocal hyperlattice is also *fixed*, a one-to-one correspondence between the incommensurate density $n(\vec{x})$ and a hypercrystal density

$$\bar{n}(\vec{\mathbf{x}}) = \sum_{\vec{Q}} \bar{n}(\vec{Q}) e^{i\vec{Q}\cdot\vec{\mathbf{x}}} \quad (\text{A3})$$

can also be established using the one-to-one correspondence

$$\vec{Q} = \mathbf{B} \cdot \vec{\mathbf{M}} \leftrightarrow \vec{\bar{Q}} = \bar{\mathbf{B}} \cdot \vec{\bar{\mathbf{M}}}, \quad \vec{\bar{\mathbf{M}}} \in \mathbb{Z}^D, \quad (\text{A4})$$

where $\bar{\mathbf{B}} \equiv (\vec{b}_1, \dots, \vec{b}_D)$, and the consequent identification

$$\bar{n}(\vec{\bar{Q}}) = n(\vec{Q}) = n_{\vec{\mathbf{M}}}. \quad (\text{A5})$$

Therefore, in the direct space, the two densities are related by

$$n(\vec{x}) = \bar{n}[\vec{\bar{x}}(\vec{x})] \quad (\text{A6})$$

provided the three-dimensional hyperplane $\vec{\bar{x}}(\vec{x})$ is defined by

$$\vec{\bar{x}} = \frac{1}{2\pi} \bar{\mathbf{A}} \cdot \mathbf{B} \cdot \vec{x} + \vec{c}, \quad \bar{\mathbf{B}} \cdot \vec{c} \in \mathbb{Z}^D, \quad (\text{A7})$$

where the basis $\bar{\mathbf{A}} \equiv (\bar{\mathbf{A}}_1, \dots, \bar{\mathbf{A}}_D)$ of the direct-space hyperlattice is defined by

$$\frac{1}{2\pi} \bar{\mathbf{A}} = \bar{\mathbf{B}}^{-1}. \quad (\text{A8})$$

The basis $\bar{\mathbf{B}}$ is completely arbitrary except for the condi-

tion $\det \bar{\mathbf{B}} \neq 0$. However, certain metric considerations can be used to restrict the choice of $\bar{\mathbf{B}}$. For example, since $\text{Rank} \mathbf{B} = 3$, it is always possible to choose

$$\bar{B}_{i\alpha} = B_{i\alpha}, \quad i = 1, 2, 3 \quad (\text{A9})$$

while $\bar{B}_{i\alpha}$, $i = 4, 5, \dots, D$, is still arbitrary, ensuring that the three-plane Eq. (A7), identified by the superscript \parallel , has the identical basis as the physical space. That is, Eq. (A7) can be replaced by

$$\bar{\mathbf{x}} \equiv (\mathbf{x}^{\parallel}, \mathbf{x}^{\perp}) = (\bar{\mathbf{x}}, \mathbf{0}^{\perp}), \quad (\text{A10})$$

where \perp denotes the $(D-3)$ -dimensional subspace orthogonal to the physical space, and the quasicrystal density is explicitly a cut through the hypercrystal density [cf. Eq. (A6)],

$$n(\bar{\mathbf{x}}) = \bar{n}(\bar{\mathbf{x}}, \mathbf{0}^{\perp}). \quad (\text{A11})$$

It is often useful to impose additional metric restrictions on the choice of the reciprocal hyperlattice. For example, one might require that the orthogonal symmetry operations which leave the set $\{\bar{Q}\}$ invariant, be represented by orthogonal symmetry operations in the hyperspace.

In the case of experimentally observed icosahedral quasicrystals, the basis \mathbf{B} is formed by $D=6$ vectors \bar{b}_{α} along the six fivefold axes (vertices) of an icosahedron. We shall choose the orientation

$$\bar{B}_{i\alpha} = B_{i\alpha} = \frac{Q_0}{\sqrt{\tau+2}} \begin{pmatrix} \tau & \tau & 1 & 0 & 0 & 1 \\ 0 & 0 & \tau & 1 & -1 & -\tau \\ 1 & -1 & 0 & \tau & \tau & 0 \end{pmatrix}, \quad i = 1, 2, 3 \quad (\text{A12})$$

where Q_0 is the magnitude of the vectors in the basis, and $\tau = \frac{1}{2}(1 + \sqrt{5})$. Since the rows of \mathbf{B} are mutually orthogonal and of the same magnitude $\bar{Q}_0 \equiv Q_0 \sqrt{2}$, one can immediately choose $\bar{\mathbf{B}}$ to generate a hypercubic lattice. This choice is also dictated by the fact that an action \mathbf{G} of the icosahedral group on the physical, parallel space spanned by $\bar{\mathbf{B}}_i$ [Eq. (A12)] is equivalent to an action $\bar{\mathbf{G}}$ which permutes and changes the signs of the vectors \bar{b}_{α} , that is, to an action of a subgroup of the hypercubic group. Therefore, $\bar{\mathbf{B}}$ is proportional to a rotation which rotates a coordinate system aligned with the hypercubic axes into one aligned with the parallel and perpendicular spaces. The basis of the perpendicular space can be chosen by observing that $\bar{\mathbf{G}}$ is reducible with its perpendicular block although inequivalent to \mathbf{G} , related to it by an outer automorphism of the icosahedral group. Therefore, the perpendicular components of the vectors \bar{b}_{α} can also be aligned with the fivefold axes of an icosahedron. Specifically, we choose⁴⁸

$$\bar{B}_{i\alpha} = \frac{Q_0}{\sqrt{\tau+2}} \begin{pmatrix} 1 & 1 & -\tau & 0 & 0 & -\tau \\ 0 & 0 & 1 & -\tau & \tau & -1 \\ -\tau & \tau & 0 & 1 & 1 & 0 \end{pmatrix}, \quad i = 4, 5, 6 \quad (\text{A13})$$

to span the three-dimensional perpendicular space.

In summary, the hypercubic lattice is generated by

$$\bar{\mathbf{a}}_{\alpha} = \bar{a} \hat{\mathbf{e}}_{\alpha}, \quad \hat{\mathbf{e}}_{\alpha} = \hat{\mathbf{e}}_{\beta} = \delta_{\alpha\beta}, \quad (\text{A14})$$

$$\bar{a} = \frac{2\pi}{\bar{Q}_0}. \quad (\text{A15})$$

Its orientation with respect to the parallel and perpendicular spaces is given by Eqs. (A12) and (A13), which are proportional to the projections of the vectors $\hat{\mathbf{e}}_{\alpha}$ on the two spaces.

APPENDIX B: RY FUNCTIONAL FOR HYPERCRYSTALS

In this appendix we prove that the RY variational energy per unit volume of an incommensurate crystal equals the RY variational energy per unit volume of the associated hypercrystal.

The RY functional is a sum of local and nonlocal terms. The local term has the form [cf. Eq. (2.2)]

$$\frac{\Delta W_{\text{loc}}}{V} = \frac{1}{V} \int_V F(n(\bar{\mathbf{x}})) d^3 \bar{\mathbf{x}}. \quad (\text{B1})$$

For a quasiperiodic density given by Eq. (2.6) this expression can be rewritten

$$\frac{\Delta W_{\text{loc}}}{V} = \frac{1}{V} \int_V F(\bar{n}(\bar{\mathbf{x}})) \delta^{\perp}(\mathbf{x}^{\perp}) d^D \bar{\mathbf{x}}. \quad (\text{B2})$$

By substituting

$$\bar{\mathbf{x}} \rightarrow \bar{\mathbf{x}} - \bar{\mathbf{R}}, \quad (\text{B3})$$

where $\bar{\mathbf{R}}$ is a hyperlattice vector and the new $\bar{\mathbf{x}}$ is restricted to the unit hypercell \bar{v}_c , Eq. (B2) can be replaced by

$$\frac{\Delta W_{\text{loc}}}{V} = \frac{1}{V} \int_{\bar{v}_c} F(\bar{n}(\bar{\mathbf{x}})) \sum_{\bar{\mathbf{R}}} \delta^{\perp}(\mathbf{x}^{\perp} - \bar{\mathbf{R}}^{\perp}) d^D \bar{\mathbf{x}}. \quad (\text{B4})$$

The sum over the δ functions is nothing but the density of $\bar{\mathbf{R}}^{\perp}$ points in the perpendicular space, which, by virtue of incommensurability, is uniform and equal to V/\bar{v}_c ,

$$\lim_{V \rightarrow \infty} \sum_{\bar{\mathbf{R}}} \delta^{\perp}(\mathbf{x}^{\perp} - \bar{\mathbf{R}}^{\perp}) \rightarrow \frac{V}{\bar{v}_c}. \quad (\text{B5})$$

Consequently,

$$\frac{\Delta W_{\text{loc}}}{V} = \frac{1}{V} \int_{\bar{v}_c} F(\bar{n}(\bar{\mathbf{x}})) d^D \bar{\mathbf{x}} = \frac{\Delta \bar{W}_{\text{loc}}}{\bar{V}}, \quad (\text{B6})$$

where $\bar{n}_l \equiv n_l$ and $\Delta \bar{\mu} \equiv \Delta \mu$.

The nonlocal terms in the RY functional are most easily evaluated using the explicit Fourier decomposition of the density [Eq. (A1)]. The typical nonlocal term has the form

$$\frac{\Delta W_{\text{nonl}}^{(m)}}{n_l V} = -\frac{1}{m!} \left[\prod_{i=1}^m \sum_{\bar{\mathbf{M}}_i \in Z^D} \right] \bar{\delta}_{\Sigma \bar{\mathbf{M}}_j} \times C^{(m)}(\bar{Q}_1, \dots, \bar{Q}_{m-1}) \prod_{i=1}^m \frac{\Delta n(\bar{Q}_i)}{n_l}, \quad (\text{B7})$$

where $\bar{\delta}_{\bar{\mathbf{M}}} \equiv \prod_{\alpha=1}^D \delta_{\bar{M}_{\alpha}, 0}$ is a generalized Kronecker delta. Clearly, since \bar{Q}_i is in one-to-one correspondence with the

reciprocal hyperlattice vector \bar{Q}_i , such nonlocal terms can be expressed also as

$$\begin{aligned} \frac{\Delta W_{\text{nonl}}^{(m)}}{n_l V} &= -\frac{1}{m!} \left[\prod_{i=1}^m \sum_{\bar{M}_i \in \mathbb{Z}^D} \right] \bar{\delta}_{\Sigma \bar{M}_j} \\ &\times \bar{C}^{(m)}(\bar{Q}_1, \dots, \bar{Q}_{m-1}) \prod_{i=1}^m \frac{\Delta \bar{n}(\bar{Q}_i)}{\bar{n}_i} \\ &= \frac{\Delta \bar{W}_{\text{nonl}}^{(m)}}{n_l V}, \end{aligned} \quad (\text{B8})$$

where we exploited Eq. (A5) and defined

$$\begin{aligned} \bar{C}^{(m)}(\bar{Q}_1, \dots, \bar{Q}_{m-1}) &\equiv C^{(m)}(\bar{Q}_1, \dots, \bar{Q}_{m-1}), \\ \bar{Q}_i &\equiv \mathbf{Q}_i^{\parallel} \end{aligned} \quad (\text{B9})$$

or, in the direct space, using translational invariance,

$$\begin{aligned} \bar{C}^{(m)}(\bar{\mathbf{x}}_1 - \bar{\mathbf{x}}_m, \dots, \bar{\mathbf{x}}_{m-1} - \bar{\mathbf{x}}_m) \\ &\equiv C^{(m)}(\bar{\mathbf{x}}_1 - \bar{\mathbf{x}}_m, \dots, \bar{\mathbf{x}}_{m-1} - \bar{\mathbf{x}}_m) \\ &\times \prod_{i=1}^{m-1} \delta^{\perp}(\bar{\mathbf{x}}_i^{\perp} - \bar{\mathbf{x}}_m^{\perp}). \end{aligned} \quad (\text{B10})$$

Therefore, term-by-term, we see that minimization of the RY density functional in the subspace of quasiperiodic densities defined by Eq. (A1) is equivalent to minimization of the D -dimensional RY density functional in the subspace of periodic densities defined by Eq. (A3), provided that the direct m -point correlations of a reference liquid in the hyperspace are defined by Eqs. (B9) and (B10).

As described in Sec. II and in Ref. 44, a thermal disordering of an incommensurate crystal can be described in terms of a Gaussian smearing of the atomic hypersurfaces decorating the hypercell. That is, the atomic surfaces are convoluted with a Gaussian and Eqs. (2.14)–(2.16) follow. Normalization of the Gaussian is n_s/n_{0s} , where n_s and n_{0s} are the densities of the disordered and undisordered quasicrystals, respectively. The cases $n_s/n_{0s} > 1$ and $n_s/n_{0s} < 1$ correspond to the presence of interstitials or vacancies, respectively.

If the overlap between the Gaussians in different unit hypercells is small, the first term in the nonlocal part of the grand-canonical thermodynamic potential, Eqs. (2.10) and (B6) can be approximated by

$$\begin{aligned} \frac{1}{\bar{v}_c} \int \frac{n_s e^{-\bar{\mathbf{x}} \cdot \bar{\Gamma}_A^{-1} \cdot \bar{\mathbf{x}}}}{\bar{v}_c (\pi^D \det \bar{\Gamma})^{1/2}} \left[\ln \left(\frac{\xi}{(\pi^D \det \bar{\Gamma})^{1/2}} \right) \right. \\ \left. - \bar{\mathbf{x}} \cdot \bar{\Gamma}_A^{-1} \cdot \bar{\mathbf{x}} \right] d^D \bar{\mathbf{x}}, \end{aligned} \quad (\text{B11})$$

where $\bar{\Gamma}_A \equiv \bar{\mathbf{A}} \cdot \bar{\Gamma} \cdot {}^t \bar{\mathbf{A}}$, $\xi = n_s/n_l$, and $\bar{v}_c = \det \bar{\mathbf{A}}$. Since the Gaussians are assumed well localized, the integral in Eq. (B11) can be extended over the entire hyperspace and thus reduced to trivial Gaussian integrations. Therefore, we find

$$\begin{aligned} \frac{\Delta \bar{W}_{\text{loc}}}{n_l \bar{V}} &= \xi \left[\ln \left(\frac{\xi}{(\pi^D \det \bar{\Gamma})^{1/2}} \right) - \frac{D}{2} \right] \\ &- (\xi - 1) - \xi \Delta \mu, \end{aligned} \quad (\text{B12})$$

which is precisely the term given in Eq. (2.17).

In the opposite limiting case where the Gaussians are broad, $\bar{n}(\bar{\mathbf{x}}) \approx n_s$ and the local term of Eq. (2.9) can be expanded in the small quantity $\bar{n}(\bar{\mathbf{x}}) - n_s$. Then, Eq. (B12) is replaced by

$$\begin{aligned} \frac{\Delta \bar{W}_{\text{loc}}}{n_l \bar{V}} &= \xi \ln \xi - (\xi - 1) - \xi \Delta \mu \\ &+ \xi \sum_{l=2}^{\infty} \frac{2l-1}{l(l-1)} \left[\prod_{i=1}^l \sum_{\bar{Q}_i (\neq \bar{0})} \right] \\ &\times \bar{\delta}_{\Sigma \bar{M}_i} \left[\prod_i h_{\bar{M}_i}(\bar{\Gamma}) \right]^{1/2}. \end{aligned} \quad (\text{B13})$$

In this paper we consider only the situation where the Gaussians can be assumed narrow so that Eq. (B12) applies. This might not be the case for incommensurate crystals with continuous atomic hypersurfaces.

In conclusion to this section we would like to recall the exact inflation symmetry of icosahedral quasicrystals.^{8,52–56} A detailed analysis of the inflation symmetry and its application to the RY theory and Gaussian ansatz will be presented elsewhere.⁵⁷ Briefly, the inflation symmetry is a discrete symmetry

$$\bar{a} \rightarrow \tau^{3m} \bar{a}, \quad (\text{B14})$$

$$\Gamma^{\parallel} \rightarrow \tau^{-6m} \Gamma^{\parallel}, \quad (\text{B15})$$

$$\Gamma^{\perp} \rightarrow \tau^{6m} \Gamma^{\perp}, \quad (\text{B16})$$

which is related to the invariance of \mathbb{Z}^6 under

$$\mathbf{M}^{\parallel} \rightarrow \tau^{3m} \mathbf{M}^{\parallel}, \quad (\text{B17})$$

$$\mathbf{M}^{\perp} \rightarrow \tau^{-3m} \mathbf{M}^{\perp}, \quad (\text{B18})$$

where $\bar{\mathbf{M}} \in \mathbb{Z}^6$ and m is any integer. Clearly, because of the invariance of \mathbb{Z}^6 under Eqs. (B17) and (B18), the variational potential Eq. (2.17) is invariant under transformation [Eqs. (B14)–(B16)]. Consequently, if a minimum is found at \bar{a} , Γ^{\parallel} , and Γ^{\perp} , then an infinite family of minima related by Eqs. (B14)–(B16) must be also found. We used this as a check to our numerical minimization.

APPENDIX C: DISTORTIONS OF INCOMMENSURATE CRYSTALS

A uniform translation \bar{u} changes the density $n(\bar{\mathbf{x}})$ into $n(\bar{\mathbf{x}} + \bar{u})$, or equivalently, $n(\bar{Q})$ into $e^{i\bar{Q} \cdot \bar{u}} n(\bar{Q})$. It is clear from Eqs. (B1) and (B7) that, just as for crystals, this transformation does not change the RY variational potential. However, the density $n(\bar{\mathbf{x}})$ can also be changed by translating the hypercrystal density $\bar{n}(\bar{\mathbf{x}})$ into $\bar{n}(\bar{\mathbf{x}} + \bar{u}^{\perp})$, or equivalently, $\bar{n}(\bar{Q})$ into $e^{i\mathbf{Q}^{\perp} \cdot \bar{u}^{\perp}} \bar{n}(\bar{Q})$. It is clear from Eqs. (B6) and (B8) that this transformation does not change the RY functional since the corresponding D -dimensional RY functional is invariant under any D -dimensional uniform displacement, including the pure

phonon displacement $\mathbf{u}^{\parallel} = \vec{u}$ and the pure phason displacement \mathbf{u}^{\perp} .

Since the RY functional of the incommensurate crystal is invariant under the uniform displacement \vec{u} , it is natural to define a uniform elastic distortion of the incommensurate crystal by

$$\vec{u}(\vec{x}) = \varepsilon^{\parallel\parallel} \cdot \vec{x}, \quad (\text{C1})$$

and

$$\mathbf{u}^{\perp}(\vec{x}) = \varepsilon^{\perp\perp} \cdot \vec{x}. \quad (\text{C2})$$

More generally, the density of the distorted incommensurate crystal is characterized by the distorted reciprocal hyperlattice

$$\bar{\mathbf{Q}} \rightarrow \bar{\mathbf{Q}}_{\varepsilon} = \begin{pmatrix} \mathbf{Q}^{\parallel} - \varepsilon^{\parallel\parallel} \cdot \mathbf{Q}^{\parallel} - \varepsilon^{\perp\perp} \cdot \mathbf{Q}^{\perp} \\ \mathbf{Q}^{\perp} \end{pmatrix}. \quad (\text{C3})$$

In other words, phonon and phason strained incommensurate crystal is equivalent to the strained hypercrystal with the strain matrix

$$\varepsilon \equiv \begin{pmatrix} \varepsilon^{\parallel\parallel} & 0 \\ \varepsilon^{\perp\perp} & 0 \end{pmatrix}. \quad (\text{C4})$$

Note, although $\varepsilon^{\parallel\parallel}$ must be symmetric by virtue of the rotational invariance of the variational functional, ε is not symmetric. Indeed, $\varepsilon^{\perp\perp}$ and $\varepsilon^{\parallel\perp}$ components of ε are not present since the RY functional is invariant under these distortions. They correspond to

$$\begin{pmatrix} \mathbf{Q}^{\parallel} \\ \mathbf{Q}^{\perp} \end{pmatrix} \rightarrow \begin{pmatrix} \mathbf{Q}^{\parallel} \\ \mathbf{Q}^{\perp} - \varepsilon^{\parallel\perp} \cdot \mathbf{Q}^{\parallel} - \varepsilon^{\perp\perp} \cdot \mathbf{Q}^{\perp} \end{pmatrix}. \quad (\text{C5})$$

However, since according to Eq. (B9) $\bar{C}^{(m)}$ depends only on the $\mathbf{Q}^{\parallel} = \bar{\mathbf{Q}}$ component of $\bar{\mathbf{Q}}$, which is unchanged under this strain, the equilibrium strained density $n_{\varepsilon}(\vec{x})$ is exactly equal to the strained density $n(\vec{x})$. In fact, this is precisely a manifestation of the arbitrariness in the choice of the perpendicular components of the reciprocal hyperlattice:

$$\bar{\mathbf{b}}_{\alpha} \rightarrow \begin{pmatrix} \mathbf{b}_{\alpha}^{\parallel} \\ \mathbf{b}_{\alpha}^{\perp} - \varepsilon^{\parallel\perp} \cdot \mathbf{b}_{\alpha}^{\parallel} - \varepsilon^{\perp\perp} \cdot \mathbf{b}_{\alpha}^{\perp} \end{pmatrix}. \quad (\text{C6})$$

In the main text we shall assume that the equilibrium-strained density corresponds to the uniformly strained density at *all* scales including the unit hypercell scale. That is,

$$\bar{n}_{\varepsilon}(\vec{x}) = \sum_{\bar{\mathbf{Q}}} \frac{\bar{n}(\bar{\mathbf{Q}})}{\det(\bar{\mathbf{I}} + \bar{\varepsilon})} e^{i\bar{\mathbf{Q}} \cdot \vec{x}}, \quad (\text{C7})$$

or

$$\bar{n}_{\varepsilon}(\bar{\mathbf{Q}}_{\varepsilon}) = \frac{\bar{n}(\bar{\mathbf{Q}})}{\det(\bar{\mathbf{I}} + \bar{\varepsilon})}. \quad (\text{C8})$$

APPENDIX D: ICOSAHEDRAL ELASTIC MODULI

In a previous publication⁴⁸ we derived a group-theoretical result which can be used to evaluate the sum in Eq. (3.6),

$$\frac{C_{\alpha\beta\gamma\delta}}{k_B T n_l} = m_{\mu} \hat{f}_{\alpha\beta\gamma\delta}^{\mu}, \quad (\text{D1})$$

where

$$m_0 = \frac{\kappa_T + 2 \Delta P}{k_B T n_l} + 1 - \xi, \quad (\text{D2})$$

$$\hat{f}_{\alpha\beta\gamma\delta}^0 = \delta_{\alpha\beta} \delta_{\gamma\delta}, \quad (\text{D3})$$

and for $\mu \neq 0$

$$m_{\mu} = -\frac{\xi^2}{2} \sum_{\bar{\mathbf{Q}} (\neq \bar{\mathbf{0}})} \left[\frac{\dot{C}_l}{|\bar{\mathbf{Q}}|^2} - \frac{\dot{C}_l}{|\bar{\mathbf{Q}}|^3} \right] h_{\bar{\mathbf{M}}}(\bar{\Gamma}) \hat{f}^{\mu}(\bar{\mathbf{Q}}), \quad (\text{D4})$$

while

$$\hat{f}^{\mu}(\bar{\mathbf{Q}}) \equiv \hat{f}_{\alpha\beta\gamma\delta}^{\mu} \bar{\mathbf{Q}}_{\alpha} \bar{\mathbf{Q}}_{\beta} \bar{\mathbf{Q}}_{\gamma} \bar{\mathbf{Q}}_{\delta}, \quad (\text{D5})$$

are the basic quartic polynomials invariant under the symmetry of the incommensurate crystal. They are orthonormal in the sense that

$$\hat{f}_{\alpha\beta\gamma\delta}^{\mu} \hat{f}_{\alpha\beta\gamma\delta}^{\nu} = \delta_{\mu\nu}. \quad (\text{D6})$$

In the case of icosahedral quasicrystals $D=6$, the perpendicular space is three-dimensional, $\mu=0,1,\dots,4$, and using the orientation established in Eqs. (A11) and (A13) we determine

$$\hat{f}^1(\bar{\mathbf{Q}}) = (1/\sqrt{5}) |\mathbf{Q}^{\parallel}|^4, \quad (\text{D7})$$

$$\hat{f}^2(\bar{\mathbf{Q}}) = \sqrt{2/3} |\mathbf{Q}^{\parallel}|^2 |\mathbf{Q}^{\perp}|^2, \quad (\text{D8})$$

$$\begin{aligned} \hat{f}^3(\bar{\mathbf{Q}}) = & \sqrt{3/40} (\{ |\mathbf{Q}^{\parallel}|^2 [Q_1^{\perp 2} - \sqrt{5}(Q_2^{\perp 2} - Q_3^{\perp 2})] \\ & - 8Q_1^{\parallel} Q_2^{\parallel} Q_1^{\perp} Q_2^{\perp} \} + \text{c.p.}) \\ & - \frac{|\mathbf{Q}^{\parallel}|^2 |\mathbf{Q}^{\perp}|^2}{\sqrt{120}}, \end{aligned} \quad (\text{D9})$$

and

$$\hat{f}^4(\bar{\mathbf{Q}}) = \frac{2}{\sqrt{30}} \left[Q_1^{\perp} Q_1^{\parallel} \left[|\mathbf{Q}^{\perp}|^2 - 3\tau Q_2^{\perp 2} + \frac{3}{\tau} Q_3^{\perp 2} \right] + \text{c.p.} \right], \quad (\text{D10})$$

where c.p. denotes cyclic permutations. Since $D=6$ for icosahedral symmetry, the generalized strain $\bar{\varepsilon}$ has fifteen independent components: the six components of the usual symmetric strain $\varepsilon^{\parallel\parallel}$ and the nine components of the phason strain $\varepsilon^{\perp\perp}$. We shall denote the 15 independent strain components by $\bar{\varepsilon}_1 \equiv \varepsilon_{1,1}^{\parallel}$, $\bar{\varepsilon}_2 \equiv \varepsilon_{2,2}^{\parallel}$, $\bar{\varepsilon}_3 \equiv \varepsilon_{3,3}^{\parallel}$, $\bar{\varepsilon}_4 \equiv (1/\sqrt{2})(\varepsilon_{1,2}^{\parallel} + \varepsilon_{2,1}^{\parallel})$, $\bar{\varepsilon}_5 \equiv (1/\sqrt{2})(\varepsilon_{2,3}^{\parallel} + \varepsilon_{3,2}^{\parallel})$, $\bar{\varepsilon}_6 \equiv (1/\sqrt{2})(\varepsilon_{3,1}^{\parallel} + \varepsilon_{1,3}^{\parallel})$, $\bar{\varepsilon}_7 \equiv \varepsilon_{1,1}^{\perp}$, $\bar{\varepsilon}_8 \equiv \varepsilon_{2,2}^{\perp}$, $\bar{\varepsilon}_9 \equiv \varepsilon_{3,3}^{\perp}$, $\bar{\varepsilon}_{10} \equiv \varepsilon_{1,2}^{\perp}$, $\bar{\varepsilon}_{11} \equiv \varepsilon_{2,3}^{\perp}$, $\bar{\varepsilon}_{12} \equiv \varepsilon_{2,3}^{\perp}$, $\bar{\varepsilon}_{13} \equiv \varepsilon_{3,1}^{\perp}$, $\bar{\varepsilon}_{14} \equiv \varepsilon_{3,1}^{\perp}$, $\bar{\varepsilon}_{15} \equiv \varepsilon_{1,3}^{\perp}$. Using this notation the elastic energy Eq. (3.5) can be written as

$$\frac{E_\epsilon}{k_B T n_l V} = \frac{1}{2} \bar{\epsilon}_\alpha \bar{C}_{\alpha,\beta} \bar{\epsilon}_\beta, \quad \alpha, \beta = 1, 2, \dots, 15. \quad (\text{D11})$$

With this notation we have, for example, $\bar{C}_{1,1} = C_{1,1;1,1}^{\parallel,\parallel,\parallel,\parallel}$ and $\bar{C}_{4,4} = 2C_{1,2;1,2}^{\parallel,\parallel,\parallel,\parallel}$, which is different from the Voigt notation.

The 15×15 elastic-modulus matrix $\bar{C}_{\alpha,\beta}$ has, by symmetry, five independent parameters. For example,

$$\bar{C} = \sum_{i=1}^5 c_i \hat{C}_i, \quad (\text{D12})$$

where the five basic matrices \hat{C}_i are orthonormalized,

$$\hat{C}_i \cdot \hat{C}_j = \delta_{ij}, \quad (\text{D13})$$

and given by

$$\hat{C}_1^{\parallel,\parallel,\parallel,\parallel} = \frac{1}{\sqrt{6}} I^{\parallel,\parallel,\parallel,\parallel}, \quad (\text{D14})$$

$$\hat{C}_2^{\parallel,\parallel,\parallel,\parallel} = \frac{1}{\sqrt{30}} \begin{pmatrix} \begin{pmatrix} 1 & 2 & 2 \\ 2 & 1 & 2 \\ 2 & 2 & 1 \end{pmatrix} & 0_{3 \times 3} \\ 0_{3 \times 3} & \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix} \end{pmatrix}, \quad (\text{D15})$$

$$\hat{C}_3^{\perp,\perp,\perp,\perp} = \frac{1}{3} I^{\perp,\perp,\perp,\perp}, \quad (\text{D16})$$

$$\hat{C}_4^{\perp,\perp,\perp,\perp} = \frac{1}{4\sqrt{5}} \begin{pmatrix} \begin{pmatrix} -\frac{2}{3} & 2 & 2 \\ 2 & -\frac{2}{3} & 2 \\ 2 & 2 & -\frac{2}{3} \end{pmatrix} & 0_{3 \times 6} \\ 0_{6 \times 3} & \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \otimes \begin{pmatrix} \frac{1}{3} - \sqrt{5} & 2 \\ 2 & \frac{1}{3} + \sqrt{5} \end{pmatrix} \end{pmatrix}, \quad (\text{D17})$$

$$\hat{C}_5^{\perp,\perp,\perp,\perp} = i \hat{C}_5^{\parallel,\parallel,\perp,\perp} = \frac{1}{\sqrt{60}} \begin{pmatrix} \begin{pmatrix} 1 & -\tau & \tau^{-1} \\ \tau^{-1} & 1 & -\tau \\ -\tau & \tau^{-1} & 1 \end{pmatrix} & 0_{3 \times 3} \\ 0_{6 \times 3} & \sqrt{2} \begin{pmatrix} -\tau & 0 & 0 \\ \tau^{-1} & 0 & 0 \\ 0 & -\tau & 0 \\ 0 & \tau^{-1} & 0 \\ 0 & 0 & -\tau \\ 0 & 0 & \tau^{-1} \end{pmatrix} \end{pmatrix}. \quad (\text{D18})$$

Only the nonzero blocks are shown and I denotes the appropriate identity. The coefficients c_i can be related to the coefficients m_μ by

$$\begin{aligned} c_1 &= (\sqrt{5}m_1 + 3m_0)/\sqrt{6}, \\ c_2 &= (m_1 + 3\sqrt{5}m_0)/\sqrt{6}, \\ c_3 &= 3m_2/\sqrt{6}, \\ c_4 &= -m_3/\sqrt{6}, \\ c_5 &= m_4/2. \end{aligned} \quad (\text{D19})$$

Although the icosahedral symmetry allows for five arbitrary parameters, it restricts the number of distinct eigenvalues of $\bar{C}_{\alpha,\beta}$ to four. Moreover, by utilizing this

symmetry it is possible to evaluate these eigenvalues analytically. They are the nondegenerate eigenvalue

$$\Lambda_1 = \frac{1}{\sqrt{6}} (c_1 + \sqrt{5}c_2), \quad (\text{D20})$$

the fourfold eigenvalue

$$\Lambda_4 = \frac{1}{3} \left[c_3 + \frac{\sqrt{5}}{2} c_4 \right], \quad (\text{D21})$$

and the two fivefold eigenvalues

$$\Lambda_{5\pm} = \frac{\lambda_3^{\parallel} + \lambda_5^{\perp}}{2} \pm \frac{[(\lambda_3^{\parallel} - \lambda_5^{\perp})^2 + 4\lambda_5^{\perp}]^{1/2}}{2}, \quad (\text{D22})$$

where

$$\lambda_5^{\parallel} = \frac{1}{\sqrt{6}} \left[c_1 - \frac{1}{\sqrt{5}} c_2 \right], \quad (\text{D23})$$

$$\lambda_5^{\perp} = \frac{1}{3} \left[c_3 - \frac{2}{\sqrt{5}} c_4 \right], \quad (\text{D24})$$

and

$$\lambda_5^i = \frac{c_5^2}{10}. \quad (\text{D25})$$

Note that Λ_1 and λ_5^{\parallel} are the nondegenerate and fivefold-degenerate eigenvalues of the phonon block $C^{\parallel, \parallel; \parallel, \parallel}$ of the tensor \bar{C} , while Λ_4 and λ_5^{\perp} are the fourfold-degenerate and fivefold-degenerate eigenvalues of the phason block $C^{\perp, \parallel; \perp, \parallel}$. Similarly, λ_5^i is the fivefold-degenerate (and the only nonzero) eigenvalue of the product, in either order of the two off-diagonal, interaction blocks of \bar{C} (that is, of $C^{\parallel, \parallel; \perp, \parallel}$, $C^{\perp, \parallel; \parallel, \parallel}$ and $C^{\perp, \parallel; \parallel, \parallel}$, $C^{\parallel, \parallel; \perp, \parallel}$).

Since the short-time response to the usual stress is given by $C^{\parallel, \parallel; \parallel, \parallel}$, the short-time bulk (B) and shear (G) moduli are simply related to Λ_1 and λ_5^{\parallel} ,

$$3B = C_{11} + 2C_{12} = 2\lambda + 3\mu = \Lambda_1 \quad (\text{D26})$$

and

$$2G = C_{11} - C_{12} = 2\mu = \lambda_5^{\parallel}, \quad (\text{D27})$$

where C_{ij} are the elastic moduli in the Voigt notation and

λ and μ are the Lamé coefficients. In order to facilitate comparison with cubic crystals we recall that

$$G = C_{44} = \frac{1}{2}(C_{11} - C_{22}) = \mu \quad (\text{D28})$$

for an isotropic solid.

The long-time response to the stress is given by Eq. (3.8) which can be diagonalized to give

$$\Lambda_{1\text{eff}} = \Lambda_1 \quad (\text{D29})$$

and

$$\lambda_{5\text{eff}}^{\parallel} = \lambda_5^{\parallel} - \frac{\lambda_5^i}{\lambda_5^{\perp}}. \quad (\text{D30})$$

The corresponding long-time bulk and shear moduli can be determined from Eqs. (D26) and (D27) with the appropriate substitutions of Λ_1 and λ_5^{\parallel} by $\Lambda_{1\text{eff}}$ and $\lambda_{5\text{eff}}^{\parallel}$, respectively.

We conclude this section by observing that a sufficiently strong, either positive or negative, phonon-phason coupling c_5 can make Λ_{5-} negative and cause an elastic instability. This happens precisely at the same point

$$\lambda_5^i = \lambda_5^{\parallel} \lambda_5^{\perp} \quad (\text{D31})$$

at which the long-time, effective shear modulus $\lambda_{5\text{eff}}^{\parallel}$ equals zero.

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