Mechanically stable effective isotropic media for all crystal classes: Construction of the third-order elastic constants

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The simplest kind of mutual interactions of long-wavelength acoustic phonons in crystalline elastic media is characterized by sets of components of tensors of the third-order elastic constants. Averaging these components over all directions of axes of a laboratory frame we obtained elastic constants for effective isotropic media corresponding to anisotropic media that belong to all crystal classes. In regions of stability of anisotropic media the corresponding tensors of effective second-order elastic constants are positive, i.e., effective isotropic media are mechanically stable. The obtained sets of elastic constants for effective isotropic media can be used in computer experiments on nonequilibrium gases of long-wavelength acoustic phonons.

I. INTRODUCTION

With the advent of the experimental technique of heat pulses the evolution of nonequilibrium states of phonon gases becomes intensively studied both experimentally and theoretically. These pulses are used as a useful tool for the investigation of many other properties of crystalline solids.[1]

In most experiments beams of long-wavelength acoustic phonons are used. They propagate in crystalline media of various symmetries. Characteristics of such phonons, namely frequencies, phase and group velocities, polarization vectors as well as shapes of slowness surfaces, are defined by a set of independent components $C_{ij,kl}$ $(i,j,k,l=1,2,3)$ of the tensor of second-order elastic constants $\mathcal{C}_2$. This tensor belongs to the class $[V^2]^2$. The number of its independent components n_2 varies from 18 for triclinic to 2 for isotropic media.[2,3]

At ambient temperatures much lower than the Debye temperature, the density of thermal phonons is so low that one can neglect the phonon merging processes and consequently only processes of the spontaneous decay should be accounted for. Characterization of even the simplest kinds of such phonon-phonon processes, that is three-phonon anharmonic spontaneous decay processes, needs familiarity with second-order elastic constants (via frequencies) and also with the whole set of third-order elastic constants $C_{ij,kl,mn}$, which are components of the sixth-rank tensor $\mathcal{C}_3$. Tensor $\mathcal{C}_3$ belongs to the class $[V^2]^3$. The number n_3 of independent elastic constants varies from 56 to 3.[3–5] Together with scattering by lattice imperfections, mutual phonon interactions determine the evolution of the spectral composition of nonequilibrium phonon systems. Such evolution is studied analytically[6] as well as in computer simulations (cf. Refs. 7–10).

Due to their complexity, even spontaneous decay processes are studied, usually in equivalent effective isotropic media (EIM's).[7] For such studies it is indispensable to know elastic constants of the second and third order for the related EIM's, i.e., the components of $\mathcal{C}_2^{(ef)}$ and $\mathcal{C}_3^{(ef)}$, respectively. For cubic elastic media these components were calculated by Tamura,[7] who used the method invented by Fedorov for the second-order elastic constants.[2] Using the method of averaging of components of tensors of elastic constants over all orientations of axes of the laboratory frame we calculated elements of $\mathcal{C}_2^{(ef)}$ for all crystal classes.[11] Hence, we shall not reproduce here these results and confine ourselves only to the discussion of stability of these linear EIM's (cf. Sec. III). The same method is used here for derivation of expressions for the third-order elastic constants of EIM's for all crystal classes (Sec. II).

Additionally, for cubic and transversely isotropic media we already proved that our method of obtaining EIM's reduces complicated spectra of relaxation rates related to the elastic scattering in anisotropic media to simple, purely discrete spectrum, which is characteristic for all isotropic media.[12]

II. TENSORS OF THIRD-ORDER ELASTIC CONSTANTS

For isotropic media the tensor $\mathcal{C}_3$ depends on three parameters α, β, and γ.[13] It can be expressed in terms of the unit second-rank tensor I_2 with the components $\delta_{\alpha,\beta}$,

$$\begin{aligned}C_{\alpha_1\mu_1,\alpha_2\mu_2,\alpha_3\mu_3}=&\alpha\delta_{\alpha_1,\mu_1}\delta_{\alpha_2,\mu_2}\delta_{\alpha_3,\mu_3}\\&+\beta[\delta_{\alpha_1,\mu_1}(\delta_{\alpha_2,\alpha_3}\delta_{\mu_2,\mu_3}+\delta_{\alpha_2,\mu_3}\delta_{\mu_2,\alpha_3})\\&+\delta_{\alpha_2,\mu_2}(\delta_{\alpha_1,\alpha_3}\delta_{\mu_1,\mu_3}+\delta_{\alpha_1,\mu_3}\delta_{\mu_1,\alpha_3})\\&+\delta_{\alpha_3,\mu_3}(\delta_{\alpha_1,\alpha_2}\delta_{\mu_1,\mu_2}+\delta_{\alpha_1,\mu_2}\delta_{\mu_1,\alpha_2})]\\&+\gamma[\delta_{\mu_1,\mu_3}(\delta_{\alpha_1,\alpha_2}\delta_{\mu_2,\alpha_3}+\delta_{\alpha_1,\mu_2}\delta_{\alpha_2,\alpha_3})\\&+\delta_{\alpha_1,\mu_3}(\delta_{\mu_1,\alpha_2}\delta_{\mu_2,\alpha_3}+\delta_{\mu_1,\mu_2}\delta_{\alpha_2,\alpha_3})\\&+\delta_{\mu_1,\alpha_3}(\delta_{\alpha_1,\alpha_2}\delta_{\mu_2,\mu_3}+\delta_{\alpha_1,\mu_2}\delta_{\alpha_2,\mu_3})\\&+\delta_{\alpha_1,\alpha_3}(\delta_{\mu_1,\alpha_2}\delta_{\mu_2,\mu_3}+\delta_{\mu_1,\mu_2}\delta_{\alpha_2,\mu_3})].\end{aligned}$$

Description of an elastically anisotropic medium is more complicated. It is characterized by orientation of crystallographical axes $\mathbf{e}_1',\mathbf{e}_2',\mathbf{e}_3'$ with respect of axes of a laboratory coordinate system. In the case of lowest symmetry classes these axes may not be orthogonal, hence one introduces orthogonal ''crystallographic'' systems of axes $\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3$ related to $\mathbf{e}_1',\mathbf{e}_2',\mathbf{e}_3'$ imposing some conditions on elastic constants (cf. for example, textbook by Sirotin and Shaskolskaya, Ref. 3 Sec. 52). We shall consider tensors $\mathcal{C}_3$ in such orthogonal crystallographic coordinate systems. Consider a rotation Ω of crystallographic axes with respect to laboratory axes through three Euler angles ϕ, θ, and ψ ($0\leqslant\phi<2\pi$, $0\leqslant\theta\leqslant\pi$, $0\leqslant\psi<2\pi$).[14] Denote suitable rotation matrix with matrix elements $\Omega_{\alpha\beta}$ by $\tilde{\Omega}(\phi,\theta,\psi)$.

Suppose that we deal with a rotated elastic anisotropic medium the nonlinear properties of which are characterized by the tensor $\mathcal{C}_3(\Omega)$ with components $C_{\alpha\beta,\gamma\delta,\mu\nu}(\Omega)$,

$$C_{\alpha\beta,\gamma\delta,\lambda\rho}(\Omega)=\sum_{\alpha',\beta',\gamma',\delta',\lambda',\rho'=1}^{3}\Omega_{\alpha\alpha'}\Omega_{\beta\beta'}\times\Omega_{\gamma\gamma'}\Omega_{\delta\delta'}\Omega_{\lambda\lambda'}\Omega_{\rho\rho'}C_{\alpha'\beta',\gamma'\delta',\lambda'\rho'}. \tag{1}$$

To find the tensor elastic constant $\mathcal{C}_{3ef}^{(is)}$ for an effective isotropic elastic medium corresponding to the chosen anisotropic medium, we allow for all possible orientations of crystalline axes with respect to the laboratory coordinate system, i.e., we shall integrate all elements $C_{\alpha\beta,\gamma\delta,\lambda\rho}(\Omega)$ of $\mathcal{C}_3(\Omega)$ over all allowed values of Euler angles,

$$\begin{aligned}C^{(ef)}_{\alpha\beta,\gamma\delta,\lambda\rho}&=\langle C_{\alpha\beta,\gamma\delta,\lambda\rho}(\Omega)\rangle\\&=\sum_{\alpha',\beta',\gamma',\delta',\lambda'\rho'=1}^{3}C_{\alpha'\beta',\gamma'\delta',\lambda'\rho'}\\&\quad\times R_{\alpha\alpha'\beta\beta'\gamma\gamma'\delta\delta',\lambda\lambda'\rho\rho'},\end{aligned} \tag{2}$$

where $C^{(ef)}_{\alpha\beta,\gamma\delta,\lambda\rho}$ is a component of $\mathcal{C}_{3ef}^{(is)}(\Omega)$ and

$$\begin{aligned}R_{\alpha\alpha'\beta\beta'\gamma\gamma'\delta\delta'\lambda\lambda'\rho\rho'}&\equiv(\mathcal{R}_6)_{\alpha\alpha'\beta\beta'\gamma\gamma'\delta\delta'\lambda\lambda'\rho\rho'}\\&=\langle\Omega_{\alpha\alpha'}\Omega_{\beta\beta'}\Omega_{\gamma\gamma'}\Omega_{\delta\delta'}\Omega_{\lambda\lambda'}\Omega_{\rho\rho'}\rangle.\end{aligned}$$

For any function A of Euler angles ϕ, θ, ψ,

$$\langle A(\phi,\theta,\psi)\rangle=\frac{1}{8\pi^2}\int_0^{2\pi}d\phi\int_0^{\pi}d\theta\sin\theta\int_0^{2\pi}d\psi A(\phi,\theta,\psi). \tag{3}$$

The unweighted mean value of the product $(\Omega_{\alpha\alpha'}\Omega_{\beta\beta'}\Omega_{\gamma\gamma'}\Omega_{\delta\delta'}\Omega_{\lambda\lambda'}\Omega_{\rho\rho'})$ depends only on elements of the unit tensor of the second rank I_2. The tensor $\mathcal{R}_6$ has the symmetry $[V^2]^6$, e.g.,

$$\begin{aligned}R_{\alpha\alpha'\beta\beta'\gamma\gamma'\delta\delta'\lambda\lambda'\rho\rho'}&=R_{\alpha'\alpha\beta\beta'\gamma\gamma'\delta\delta'\lambda\lambda'\rho\rho'}\\&=R_{\beta\beta'\alpha\alpha'\gamma\gamma'\delta\delta'\lambda\lambda'\rho\rho'}.\end{aligned} \tag{4}$$

The results of calculation of $C^{(ef)}_{\alpha\beta,\gamma\delta,\rho\lambda}$ (2) for media belonging to all crystal classes are presented in Tables I–III. Using Maple we checked that the obtained expressions agree with the Fumi reduction relations.[4,5] For cubic media our results agree with expressions obtained by Tamura.[7]

TABLE I. Parameter of elasticity alpha for effective isotropic media for all crystal systems.

Crystal class	α_{ef}
C_1,S_2,C_{1h},C_2 C_{2h},C_{2v},D_2,D_{2h}	$\frac{1}{105}(C_{111}+9C_{112}+9C_{113}+9C_{122}+48C_{123}+9C_{133}-30C_{144}-6C_{155}-6C_{166}+C_{222}+9C_{223}+9C_{233}-6C_{244}-30C_{255}-6C_{266}+C_{333}-6C_{344}-6C_{355}-30C_{366}+48C_{456})$
C_3,S_6,C_{3v} D_3,D_{3d}	$\frac{1}{105}(10C_{111}+21C_{112}+3C_{113}+63C_{123}+18C_{133}-84C_{144}+12C_{155}-11C_{222}+C_{333}-12C_{344})$
$C_{3h},C_6,C_{6h},D_6,$ D_{6h},D_{3h},C_{6v}	$\frac{1}{105}(10C_{111}+21C_{112}+3C_{113}+63C_{123}+18C_{133}-84C_{144}+12C_{155}-11C_{222}+C_{333}-12C_{344})$
C_4,S_4,C_{4h},C_{4v} D_{2d},D_4,D_{4h}	$\frac{1}{105}(2C_{111}+18C_{112}+18C_{113}+48C_{123}+18C_{133}-60C_{144}-12C_{155}-12C_{166}+C_{333}-12C_{344}+30C_{366}+48C_{456})$
T,T_h	$\frac{1}{35}(C_{111}+9C_{112}+9C_{113}+16C_{123}-30C_{144}-6C_{155}-6C_{166}+16C_{456})$
O,T_d,O_h	$(\frac{1}{35}C_{111}+18C_{112}+16C_{123}-30C_{144}-12C_{166}+16C_{456})$

III. STABILITY OF EFFECTIVE ISOTROPIC ELASTIC MEDIA

As we mentioned above in computer experiments on propagation of down-converting phonons we are enforced to consider three-phonon anharmonic processes in isotropic EIM's. Anisotropic and the related effective isotropic media have to be mechanically stable, because it guarantees that phonon frequencies are real.

Consider first isotropic media. For an isotropic elastic medium the tensor of second-order elastic constants $\mathcal{C}$ is composed as[15] $\mathcal{C}_2^{(is)}=C_J\mathcal{J}+C_K\mathcal{K}$, where $C_J=(C_{11}+2C_{12})$ and $C_K=(C_{11}-C_{12})$. The tensor $\mathcal{J}$ and the fourth-rank unit tensor $\mathcal{I}$ have, respectively, the following components:

$$\mathcal{J}_{\alpha\mu,\beta\nu}=\tfrac{1}{3}\delta_{\alpha,\mu}\delta_{\beta,\nu},\quad\mathcal{I}_{\alpha\mu,\beta\nu}=\tfrac{1}{2}(\delta_{\alpha,\beta}\delta_{\mu,\nu}+\delta_{\alpha,\nu}\delta_{\beta,\nu}),$$
$$\mathcal{K}=(\mathcal{I}-\mathcal{J}). \tag{5}$$

An elastic isotropic medium is mechanically stable when $C_J>0$, $C_K>0$. According to Table I of Ref. 11, $C_J^{(ef)}$ and $C_K^{(ef)}$ depend only on the matrix elements C_{UU} $(U=4,5,6)$ and C_{UV} $(U,V=1,2,3)$. We shall use this property in the proof of stability of effective linear isotropic media.

Consider an anisotropic medium characterized by the matrix $\tilde{C}_2$ with the elements C_{UV} $(U,V=1,\ldots,6)$. We shall prove that the stability of anisotropic medium yields the stability of the corresponding effective isotropic medium (EIM). An elastic medium is mechanically stable if and only if the matrix $\tilde{C}_2$ is positive.[2] A square matrix is positive if all its main minors are positive. In particular, for $\tilde{C}_2$ the inequalities $C_{UU}>0$ $(U=1,\ldots,6)$ hold and $M_i>0$ $(i=1,2,3)$, where M_i $(i=1,2,3)$ are the following main minors of $\tilde{C}_2$:

TABLE II. Parameter of elasticity beta for effective isotropic media for all crystal systems.

Crystal class	β_{ef}
C_1, S_2, C_{1h}, C_2 $C_{2h}, C_{2v}, D_2, D_{2h}$	$\frac{1}{105}(C_{111}+2C_{112}+2C_{113}+2C_{122}-15C_{123}+2C_{133}+19C_{144}+C_{155}+C_{166}+C_{222}+2C_{223}+2C_{233}+C_{244}+19C_{255}+C_{266}+C_{333}+C_{344}+C_{355}+19C_{366}-36C_{456})$
C_3, S_6, C_{3v} D_3, D_{3d}	$\frac{1}{210}(6C_{111}+7C_{112}+27C_{113}-49C_{123}+8C_{133}+112C_{144}-32C_{155}-C_{222}+2C_{233}+4C_{344})$
$C_{3h}, C_6, C_{6h}, D_6,$ D_{6h}, D_{3h}, C_{6v}	$\frac{1}{210}(6C_{111}+7C_{112}+27C_{113}-49C_{123}+8C_{133}+112C_{144}-32C_{155}-C_{222}+2C_{233}+4C_{344})$
C_4, S_4, C_{4h}, C_{4v} D_{2d}, D_4, D_{4h}	$\frac{1}{105}(2C_{111}+4C_{112}+4C_{113}-15C_{123}+4C_{133}+38C_{144}+2C_{155}+2C_{166}+C_{333}+2C_{344}+19C_{366}-36C_{456})$
T, T_h	$\frac{1}{35}(C_{111}+2C_{112}+2C_{113}-5C_{123}+19C_{144}+C_{155}+C_{166}-12C_{456})$
O, T_d, O_h	$\frac{1}{35}(C_{111}+4C_{112}-5C_{123}+19C_{144}+2C_{166}-12C_{456})$

$$M_1=\begin{vmatrix} C_{11} & C_{12} \\ C_{12} & C_{22} \end{vmatrix}, \quad M_2=\begin{vmatrix} C_{11} & C_{13} \\ C_{13} & C_{33} \end{vmatrix}, \quad M_3=\begin{vmatrix} C_{22} & C_{23} \\ C_{23} & C_{33} \end{vmatrix}. \tag{6}$$

Notice that expressions for $C_J^{(ef)}$, $C_K^{(ef)}$ are invariant in respect to changes of indices $1 \leftrightarrow 2 \leftrightarrow 3$. So we can assume that $C_{11} > C_{22} > C_{33}$ and that $C_{11}=1$. This means that

$$C_{11}=1, \quad C_{22} \in (0,1), \quad C_{33} \in (0, C_{22}). \tag{7}$$

The conditions of positivity of M_i impose three conditions on matrix elements C_{UV} $(U \neq V,\ U,V=1,2,3)$. Namely,

$$C_{12} \in (-\sqrt{C_{22}}, \sqrt{C_{22}}), \quad C_{13} \in (-\sqrt{C_{33}}, \sqrt{C_{33}}),$$
$$C_{23} \in (-\sqrt{C_{22}C_{33}}, \sqrt{C_{22}C_{33}}). \tag{8}$$

We shall prove that the instability of the isotropic effective medium yields instability of the corresponding anisotropic medium. An effective elastic medium is unstable, if at least one of Walpole's coefficients is negative, i.e., we have to prove that

$$(C_J^{(ef)} < 0 \vee C_K^{(ef)} < 0)$$
$$\rightarrow \text{at least one of main minors of } \tilde{C}_2 \text{ is negative.}$$

Therefore, the above statement is equivalent to the statement

$$[(C_J^{(ef)} < 0 \rightarrow \text{at least one of main minors of } \tilde{C}_2$$
$$\text{is negative}) \wedge (C_K^{(ef)} < 0 \rightarrow \text{at least}$$
$$\text{one of main minors of } \tilde{C}_2 \text{ is negative})].$$

TABLE III. Parameter of elasticity gamma for effective isotropic media for all crystal systems.

Crystal class	γ_{ef}
C_1, S_2, C_{1h}, C_2 $C_{2h}, C_{2v}, D_2, D_{2h}$	$\frac{1}{210}(2C_{111}-3C_{112}-3C_{113}-3C_{122}+12C_{123}-3C_{133}-18C_{144}+9C_{155}+9C_{166}+2C_{222}-3C_{223}-3C_{233}+9C_{244}+2C_{255}+9C_{266}+2C_{333}+9C_{344}+9C_{355}-18C_{366}+54C_{456})$
C_3, S_6, C_{3v} D_3, D_{3d}	$\frac{1}{210}(-C_{111}-\frac{21}{2}C_{112}-15C_{113}+21C_{123}-6C_{133}-63C_{144}+45C_{155}+\frac{19}{2}C_{222}+2C_{233}+18C_{334})$
$C_{3h}, C_6, C_{6h}, D_6,$ D_{6h}, D_{3h}, C_{6v}	$\frac{1}{210}(-C_{111}-\frac{21}{2}C_{112}-15C_{113}+21C_{123}-6C_{133}-63C_{144}+45C_{155}+\frac{19}{2}C_{222}+2C_{233}+18C_{334})$
C_4, S_4, C_{4h}, C_{4v} D_{2d}, D_4, D_{4h}	$\frac{1}{105}(2C_{111}-3C_{112}-3C_{113}+6C_{123}-3C_{133}-18C_{144}+9C_{155}+9C_{166}+C_{333}+9C_{344}-9C_{366}-27C_{456})$
T, T_h	$\frac{1}{70}(2C_{111}-3C_{112}-3C_{113}+4C_{123}-18C_{144}+9C_{155}+9C_{166}+18C_{456})$
O, T_d, O_h	$\frac{1}{35}(C_{111}-3C_{112}+2C_{123}-9C_{144}+9C_{166}+9C_{456})$

We shall prove that the following statements are fulfilled: (A) If $C_K < 0$ then at least one of main minors of $\tilde{C}_2$ is negative; (B) if $C_J < 0$ then at least one of main minors of $\tilde{C}_2$ is negative. Consider case (A).

(A) Suppose that $C_K^{(ef)}$ is negative. We shall show that then

$$(i) \quad C_{11}<0 \vee C_{22}<0 \vee C_{33}<0 \vee C_{44}<0 \vee C_{55}<0 \vee C_{66}<0,$$

$$(ii) \quad \vee M_1<0 \vee M_2<0 \vee M_3<0.$$

Assume that the alternative (i) is not fulfilled and consider the inequality $C_K^{(ef)} < 0$, then

$$[\tfrac{1}{3}(C_{11}+C_{22}+C_{33})+(C_{44}+C_{55}+C_{66})]$$
$$< \tfrac{1}{3}(C_{12}+C_{13}+C_{23}), \tag{9}$$

hence

$$(C_{11}+C_{22}+C_{33})^2 < (C_{12}+C_{13}+C_{23})^2. \tag{10}$$

With the help of an obvious inequality

$$C_{RS}C_{UV} < \tfrac{1}{2}(C_{RS}^2 + C_{UV}^2), \tag{11}$$

we rewrite inequality (10)

$$\tfrac{1}{2}(C_{11}^2+C_{22}^2)+\tfrac{1}{2}(C_{22}^2+C_{33}^2)+\tfrac{1}{2}(C_{11}^2+C_{33}^2)+2C_{11}C_{22} +2C_{11}C_{33}+2C_{22}C_{33}<3(C_{12}^2+C_{23}^2+C_{12}^2). \tag{12}$$

Using the inequality (11) once more the inequality (12) can be written in the form containing sum of minors M_1, M_2, M_3,

$$\sum_{i=1}^{3} M_i<0.$$

Hence, at least one of minors M_i $(i=1,2,3)$ is negative.

(B) Now assume that $C_J^{(ef)}$ is negative. If the statement $M_1<0\vee M_2<0\vee M_3<0\vee C_{11}<0\vee C_{22}<0\vee C_{33}<0\vee C_{44}<0\vee C_{55}<0\vee C_{66}<0$ is true, then obviously matrix $\tilde{C}$ is negative. Further, assume that $C_{UU}>0$ $(U=1,\ldots,6)$, then the condition $C_J^{(ef)}<0$ is equivalent to

$$\tfrac{2}{3}(1+C_{22}+C_{33})+(C_{12}+C_{13}+C_{23})<0. \tag{13}$$

In the space of five parameters C_{22}, C_{33}, C_{12}, C_{13}, C_{23}, obeying conditions (7) and (8), we introduced dense enough mesh and checked numerically that if inequality (13) is fulfilled then one of main minors of $\tilde{C}_2$ is negative.

Consequently, if the anisotropic medium is mechanically stable (i.e., $\tilde{C}_2$ is positive) the related effective isotropic medium is also mechanically stable (i.e., $C_J>0$, $C_K>0$). Of course, stability of an EIM does not guarantee the stability of the corresponding anisotropic medium, because main minors of it different than C_{UU} $(U=1,\ldots,6)$ and M_i $(i=1,2,3)$ may be negative.

We also calculated the contribution of piezoelectric terms to the second-order elastic constants for all noncentrosymmetric crystal classes. However, even for strongly piezoelectric crystals (e.g., GaN) the corresponding contribution is two orders smaller than terms related to $C_{\alpha\mu,\beta\nu}$, whereas for weak piezoelectrics (e.g., GaAs)—four orders weaker. For this reason we do no list these results.

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