How to Couple Landau Theory to an Equation of State

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We show how to construct a Landau-type free energy based on a primary order parameter coupled to finite strain with an elastic energy derived from an arbitrary equation of state $V = V(P)$. The resulting class of models provides an excellent and efficient framework for the systematic study of phase transformations for a wide range of materials up to ultrahigh pressures.

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While the experimental and computational investigation of high pressure phase transitions (HPPTs) has made considerable progress [1–3], on the theoretical side a simple, fairly general, and ready-to-use theoretical concept such as an adaptation of Landau's theory to the case of HPPTs is still lacking. In fact, the gap in many treatments of HPPTs is pretty obvious: The "stiffness" of any solid is characterized by the isothermal compressibility $\kappa(P) = -d\log V(P)/dP$. At high pressures, as the interatomic forces opposing further compression increase, a crystal's volume and lattice parameters develop a certain nonlinear behavior. Various theoretical concepts can be employed to derive so-called equations of state (EOS) [4–6] which describe the hydrostatic pressure dependence of the crystal's reduced volume $v(P) := V(P)/V_0$. At HPPTs anomalies usually appear, e.g., in $V(P)$ resulting from anomalies in the pressure dependence of the lattice parameters $a_i(P)$, $i = 1,2,3$, near a critical pressure P_c. Frequently the corresponding $P - V$ behavior is merely fitted to a number of differently parametrized EOS for each phase [2,3,7]. Admittedly, this procedure seems to fit experimental volume data in many cases, and, e.g., in the case of reconstructive transitions, often it is the only appropriate description known. However, such an approach attempts neither to describe the pressure behavior of individual strain components nor to gain possible further thermodynamic insight, which in principle should be available for, e.g., HPPTs of the group-subgroup type. A more profound theoretical approach would therefore be of vital interest to a broad audience reaching from physicists studying high pressure behavior of materials (crystals, liquid crystals, complex liquids, biological membranes, etc.) to mineralogists and geologists investigating earth's bulk properties.

Let $\boldsymbol{X}$ denote the coordinates describing a crystal in its undeformed reference state, $\hat{\boldsymbol{X}}$ those of a deformed state, and $\hat{\boldsymbol{x}}$ coordinates of some further deformation. Let $\alpha_{ik}(\boldsymbol{X}) = \frac{\partial \hat{X}_i}{\partial X_k} = \delta_{ik} + u_{ik}(\boldsymbol{X})$ denote the accompanying deformation tensor with corresponding Lagrangian strain $e_{ik} = \frac{1}{2}(\sum_n \alpha_{ni}\alpha_{nk} - \delta_{ik})$. Let $F_0[\hat{\boldsymbol{\eta}}(\hat{\boldsymbol{x}}); \hat{\boldsymbol{X}}] = \int_{V(\hat{\boldsymbol{X}})} \times \mathcal{F}_0[\hat{\boldsymbol{\eta}}(\hat{\boldsymbol{x}}); \hat{\boldsymbol{X}}]\, d\hat{V}$ denote the Helmholtz free energy [8] in the deformed state $\hat{\boldsymbol{X}}$. Internal elastic equilibrium in $\hat{\boldsymbol{X}}$ can be characterized by the stationarity of $F_0[\delta\hat{\boldsymbol{\eta}}(\hat{\boldsymbol{x}}); \hat{\boldsymbol{X}}]$ under infinitesimal displacements $\delta\hat{u}_{ik}(\hat{\boldsymbol{x}})$ subject to the *boundary conditions* $\int_{\hat{S}[\hat{X}]} \delta\hat{u}_j n_i\, d\hat{S} \equiv 0$, $i,j = 1,2,3$, expressing invariance of surface. In fact, the corresponding Lagrange multipliers constitute the Cauchy stress tensor τ_{ij} (cf. [9–11]), implying the equilibrium conditions

$$\tau_{ij} \equiv \frac{1}{V(\hat{\boldsymbol{X}})} \frac{\partial F_0(\hat{\boldsymbol{\eta}}; \hat{\boldsymbol{X}})}{\partial \hat{\eta}_{ij}} \bigg|_{\hat{\boldsymbol{\eta}}\equiv\boldsymbol{0}}, \qquad i,j = 1,2,3. \quad (1)$$

Suppose now that a crystal's Helmholtz free energy $F(\boldsymbol{Q}, \hat{\boldsymbol{\eta}}; \hat{\boldsymbol{Y}})$ in the state $\hat{\boldsymbol{Y}}$ depended on n further variables $\boldsymbol{Q} = (Q_1, \dots, Q_n)$ constituting the *primary order parameter* of the Landau theory to be constructed. A calculation similar to [9] then shows that stationarity of F implies the equilibrium conditions

$$\tau_{ij} \equiv \frac{1}{V(\hat{\boldsymbol{Y}})} \frac{\partial F(\bar{\boldsymbol{Q}}, \hat{\boldsymbol{\eta}}; \hat{\boldsymbol{Y}})}{\partial \hat{\eta}_{ij}} \bigg|_{\hat{\boldsymbol{\eta}}\equiv\boldsymbol{0}}, \qquad i,j = 1,2,3, \quad (2a)$$

$$0 \equiv \frac{1}{V(\hat{\boldsymbol{Y}})} \frac{\partial F(\boldsymbol{Q}, \boldsymbol{0}; \hat{\boldsymbol{Y}})}{\partial Q_N} \bigg|_{\boldsymbol{Q}\equiv\bar{\boldsymbol{Q}}}, \qquad N = 1, \dots, n, \quad (2b)$$

where $\bar{\boldsymbol{Q}}$ denotes the equilibrium value of $\boldsymbol{Q}$. We identify the *background Helmholtz free energy* as $F_0(\hat{\boldsymbol{\eta}}; \hat{\boldsymbol{X}}) := F(\boldsymbol{0}, \hat{\boldsymbol{\eta}}; \hat{\boldsymbol{X}})$, evaluated in the state $\hat{\boldsymbol{X}}$ characterized by condition (1). The deformation tensor $\hat{\alpha}_{ik}(\hat{\boldsymbol{X}}) = \frac{\partial \hat{Y}_i}{\partial \hat{X}_k}$ measured in $\hat{\boldsymbol{X}}$ yields the *tensor of spontaneous strain* $\hat{\epsilon}_{ik} = \hat{\epsilon}_{ik}(\bar{\boldsymbol{Q}}) := \frac{1}{2}(\sum_n \hat{\alpha}_{ni}\hat{\alpha}_{nk} - \delta_{ik})$. We now transform Eqs. (2) from the coordinates $\hat{\boldsymbol{Y}}$ referring to the system's true internal equilibrium state to the background reference system $\hat{\boldsymbol{X}}$. Recall that a strain $\hat{\rho}_{ij}$ measured in $\hat{\boldsymbol{Y}}$ corresponds to the *nonlinear superposition* $\hat{\eta}_{ij} := \hat{\epsilon}_{ij} + \sum_{mn} \hat{\alpha}_{mi}\hat{\rho}_{mn}\hat{\alpha}_{nj}$ measured in coordinates $\hat{\boldsymbol{X}}$. Invariance of the free energy then yields the conditions $(i,j = 1,2,3, N = 1, \dots, n)$

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$$\tau_{ij} \equiv \sum_{mn} \frac{\hat{\alpha}_{mi}\hat{\alpha}_{nj}}{J[\hat{\boldsymbol{\alpha}}]} \frac{1}{V(\hat{\boldsymbol{X}})} \frac{\partial F(\bar{\boldsymbol{Q}}, \hat{\boldsymbol{\epsilon}}; \hat{\boldsymbol{X}})}{\partial \hat{\epsilon}_{mn}}, \tag{3a}$$

$$0 \equiv \frac{1}{V(\hat{\boldsymbol{X}})} \frac{\partial F(\boldsymbol{Q}; \hat{\boldsymbol{\epsilon}}; \hat{\boldsymbol{X}})}{\partial Q_N}\bigg|_{\boldsymbol{Q}=\bar{\boldsymbol{Q}}}, \tag{3b}$$

where $J[\hat{\boldsymbol{\alpha}}] := \det J[\hat{\alpha}_{ij}] = V(\hat{\boldsymbol{Y}})/V(\hat{\boldsymbol{X}})$ denotes the relative volume change. In the present work we content our treatment to the case of *linear-quadratic* coupling of strain and order parameter for the sake of simplicity only and therefore consider the *ansatz*

$$F(\boldsymbol{Q}, \hat{\boldsymbol{\epsilon}}; \hat{\boldsymbol{X}}) \equiv V(\hat{\boldsymbol{X}})\Phi(\boldsymbol{Q}; \hat{\boldsymbol{X}}) + V(\hat{\boldsymbol{X}}) \sum_{IJij} d_{ij}^{IJ}(\hat{\boldsymbol{X}}) Q_I Q_J \hat{\epsilon}_{ij} + F_0(\hat{\boldsymbol{\epsilon}}; \hat{\boldsymbol{X}}). \tag{4}$$

In the undeformed reference system $\boldsymbol{X}$, the spontaneous strain ϵ_{ij} is nonlinearly superimposed on the background strain e_{ij}, yielding the total strain $\eta_{kl} = e_{kl} + \sum_{mn} \alpha_{mk}\hat{\epsilon}_{mn}\alpha_{nl}$. Inverting this relation, inserting the resulting expression for $\hat{\epsilon}_{mn}$ into (4), and rearranging terms, we obtain

$$d_{mn}^{MN}(\hat{\boldsymbol{X}}) = \frac{1}{J(\boldsymbol{\alpha})} \sum_{ij} \alpha_{mi} d_{ij}^{MN}(\boldsymbol{X}) \alpha_{nj}, \tag{5a}$$

$$\Phi(\boldsymbol{Q}; \hat{\boldsymbol{X}}) = \frac{\Phi(\boldsymbol{Q}; \hat{\boldsymbol{X}})}{J(\boldsymbol{\alpha})} + \sum_{IJij} \frac{Q_I Q_J}{J(\boldsymbol{\alpha})} d_{ij}^{IJ}(\boldsymbol{X}) e_{ij}. \tag{5b}$$

The ansatz (4) yields the equilibrium conditions

$$\tau_{ij} \equiv \sum_{kl} \frac{\hat{\alpha}_{ki}\hat{\alpha}_{lj}}{[J\hat{\boldsymbol{\alpha}}]} \left[\sum_{KL} d_{kl}^{KL}(\hat{\boldsymbol{X}}) \bar{Q}_K \bar{Q}_L + \frac{1}{V(\hat{\boldsymbol{X}})} \frac{\partial F_0(\hat{\boldsymbol{\epsilon}}; \hat{\boldsymbol{X}})}{\partial \hat{\epsilon}_{kl}}\bigg|_{\hat{\boldsymbol{\epsilon}}=\hat{\boldsymbol{\epsilon}}(\bar{\boldsymbol{Q}})} \right], \tag{6a}$$

$$0 \equiv \frac{\partial \Phi(\bar{\boldsymbol{Q}}; \hat{\boldsymbol{X}})}{\partial \bar{Q}_K} + 2\sum_{L} d_{ij}^{KL}(\hat{\boldsymbol{X}}) \bar{Q}_L \hat{\epsilon}_{ij}(\bar{\boldsymbol{Q}}), \tag{6b}$$

To overcome the apparent nonlinearities, we treat $\hat{\epsilon}_{kl}$ as infinitesimal [12]. Then $\hat{\alpha}_{mi}\hat{\alpha}_{nj}/J[\hat{\boldsymbol{\alpha}}] \approx \delta_{mi}\delta_{nj}$ and $F_0(\hat{\boldsymbol{\epsilon}}; \hat{\boldsymbol{X}}) \approx V(\hat{\boldsymbol{X}})[\sum_{ij} \tau_{ij}\hat{\epsilon}_{ij} + \frac{1}{2}\sum_{ijkl} C_{ijkl}(\hat{\boldsymbol{X}}) \times \hat{\epsilon}_{ij}\hat{\epsilon}_{kl}]$, where $C_{ijkl}(\hat{\boldsymbol{X}})$ are the crystal's elastic constants in the state $\hat{\boldsymbol{X}}$, so Eq. (6a) simplifies to

$$\sum_{KL} d_{ij}^{KL}(\hat{\boldsymbol{X}}) \bar{Q}_K \bar{Q}_L + \sum_{kl} C_{ijkl}(\hat{\boldsymbol{X}}) \hat{\epsilon}_{kl}(\bar{\boldsymbol{Q}}) \equiv 0. \tag{7a}$$

Solving (7a) for $\epsilon_{ij}(\bar{\boldsymbol{Q}})$ and inserting this into (6b), we recognize [13] the *renormalized order parameter potential*

$$\Phi_R(\boldsymbol{Q}; \hat{\boldsymbol{X}}) := \Phi(\boldsymbol{Q}; \hat{\boldsymbol{X}}) - \frac{1}{4} \sum_{IJKL} Q_I Q_J Q_K Q_L \times \left(2 \sum_{ijkl} d_{ij}^{IJ}(\hat{\boldsymbol{X}}) C_{ijkl}^{-1}(\hat{\boldsymbol{X}}) d_{kl}^{KL}(\hat{\boldsymbol{X}}) \right). \tag{8}$$

Introducing the tensor

$$T_{rost}(\hat{\boldsymbol{X}}) := \frac{1}{J(\boldsymbol{\alpha})} \sum_{ijkl} \alpha_{ir}\alpha_{jo} C_{ijkl}^{-1}(\hat{\boldsymbol{X}}) \alpha_{ks}\alpha_{lt}, \tag{9}$$

$\bar{\boldsymbol{Q}}$ can be calculated as the minimum of

$$\Phi_R(\boldsymbol{Q}; \boldsymbol{X}) = \Phi(\boldsymbol{Q}; \boldsymbol{X}) + \sum_{IJij} d_{ij}^{IJ}(\boldsymbol{X}) e_{ij}(\boldsymbol{\tau}) Q_I Q_J - \sum_{\substack{IJKL \\ ijkl}} d_{ij}^{IJ}(\boldsymbol{X}) \frac{T_{ijkl}(\hat{\boldsymbol{X}})}{2} d_{kl}^{KL}(\boldsymbol{X}) \times Q_I Q_J Q_K Q_L, \tag{10}$$

and the total strain $\boldsymbol{\eta}$ is elegantly expressed as [14]

$$\eta_{ij} = e_{ij} - \sum_{KL} \bar{Q}_K \bar{Q}_L \sum_{kl} d_{kl}^{KL}(\boldsymbol{X}) T_{ijkl}(\hat{\boldsymbol{X}}). \tag{11}$$

In the spirit of Landau theory, we assume $\Phi(Q; X)$ to be a polynomial in the variables Q_L with *constant* coefficients, leading to a purely "geometrical" stress dependence of $\Phi_R(\boldsymbol{Q}; \boldsymbol{X})$. To complete our approach, it remains to compute the order-parameter independent "background," i.e., the stress dependence of the elastic constants $C_{ijkl}(\hat{\boldsymbol{X}})$ and the components α_{ij}. For this purpose, we recall Hooke's law, stated as [15]

$$\tau_{ij}(\hat{\boldsymbol{Y}}) = B_{ijkl}(\hat{\boldsymbol{X}})\hat{\epsilon}_{kl} + O(\hat{\boldsymbol{\epsilon}}^2) \tag{12}$$

with the *Birch coefficients*

$$B_{ijkl}(\hat{\boldsymbol{X}}) = C_{ijkl}(\hat{\boldsymbol{X}}) + \tfrac{1}{2}[\tau_{il}(\hat{\boldsymbol{X}})\delta_{jk} + \tau_{jl}(\hat{\boldsymbol{X}})\delta_{ik} + \tau_{ik}(\hat{\boldsymbol{X}})\delta_{jl} + \tau_{jk}(\hat{\boldsymbol{X}})\delta_{il} - 2\tau_{ij}(\hat{\boldsymbol{X}})\delta_{kl}]. \tag{13}$$

More generally, the *compliances* $S_{ijkl}[\boldsymbol{\tau}] := B_{ijkl}^{-1}(\hat{\boldsymbol{X}})$ appear in the nonlinear partial differential equations

$$\frac{\partial e_{ij}[\boldsymbol{\tau}]}{\partial \tau_{kl}} = \sum_{mn} \alpha_{mi}[\boldsymbol{\tau}]\alpha_{nj}[\boldsymbol{\tau}] S_{mnkl}[\boldsymbol{\tau}] \tag{14}$$

$(i, j, k, l = 1, 2, 3)$ with boundary conditions $e_{ij}[\boldsymbol{0}] = 0$. For crystals of orthorhombic or higher symmetry subject to hydrostatic pressure $\tau_{ij} = -P\delta_{ij}$ and coordinates $\boldsymbol{X}$ chosen along the crystal axes, all off-diagonal deformation tensor components vanish, i.e., $\alpha_{ij}(P) = \alpha_i(P)\delta_{ij}$. Compressibility and compliance are related by (cf. [15]) $\kappa(P) = \sum_{i,k=1,2,3} S_{ik}[P]$, and (14) reduces to the definition of the *axial compressibilities*

$$\frac{d \log \alpha_i[P]}{dP} = -\sum_{k} S_{ik}[P] =: \kappa_i(P) \qquad (i = 1, 2, 3) \tag{15}$$

with $\alpha_i[0] = 1$. This suggests the ansatz

$$S_{ij}[P] \equiv \frac{\kappa(P)}{\kappa_0} \left(S_{ij}^0 + \sum_{n=1}^{\infty} \kappa_{ij}^n P^n \right), \tag{16}$$

where S_{ij}^0 denotes the zero-pressure compliance and the expansion coefficients κ_{ij}^n satisfy the conditions $\sum_{ij} \kappa_{ij}^n = 0 \ \forall\ n \in \mathbb{N}$. Using (16), Eq. (15) is integrated to give

TABLE I. Numbers q_2, q_3, q_4 of relevant (i.e., nonshear) elastic constants of second, third, and fourth order as determined from Refs. [18,19].

	Cubic I	Cubic II	Tetragonal	Orthorhombic
q_2	2	2	4	6
q_3	3	4	6	10
q_4	4	4	8	15

$$\alpha_i(P) = \prod_{n=0}^{\infty}[v(P)e^{-n\int_0^1 dt\,\log v(Pt)t^{n-1}}]^{P^n\kappa_i^n/\kappa_0}, \qquad (17)$$

where $\kappa_i^n := \sum_j \kappa_i^n$, from which the hydrostatic Lagrangian background strain $\boldsymbol{e}(P)$ is instantly computed. From this, one can in principle calculate all relevant observables such as the total (principal) strains $\eta_{ii}(P)$, $V(P)$, $\kappa_i(P)$, $C_{ij}(P)$, P-hysteresis intervals, etc. Indeed, the axial compressibilities κ_i^0 $(i = 1, 2, 3)$ and the parameters entering the chosen EOS can easily be determined from e.g., the lattice parameter slopes at $P = 0$ and an EOS fit of volume data. In addition, cubic symmetry implies that $\kappa_i^n = 0, n > 0$, while for tetragonal and orthorhombic symmetry the constants κ_i^n can usually be put to zero for $n \geq 2$ in practical applications, since the dominant P dependence of $S_{ij}(P)$ is already covered by the prefactors in Eq. (16). The suggested approximation therefore just calls for $q_2 - 1$ additional elastic parameters κ_{ij}^1, where q_2 is the number of independent second order elastic constants. Compared to the $q_3 + q_4$ fit parameters taken from Table I required for a free energy expansion using, say, only third and fourth order elastic constants, the use of the above approximation scheme is thus rewarded by a dramatic *reduction of the number of relevant fit parameters*.

An analysis of measurements of tetragonal BaCu(Si_4O_{10}) and BaCr(Si_4O_{10}) single crystals illustrates the simplicity of using the above concepts. By x-ray diffraction the P dependence of lattice parameters $a_1(P) = a_2(P)$, $a_3(P)$, and the unit cell volumes $V(P)$ of both crystals were measured very detailed at room temperature in a diamond anvil cell. One observes first order HPPTs at approximately 2.67 and 2.26 GPa, respectively, characterized by discontinuities in $a_1(P)$, $a_3(P)$, and $V(P)$, the first order character being more pronounced for BaCu(Si_4O_{10}). $\Phi(\boldsymbol{Q}; \boldsymbol{X})$ is constructed in the following standard way [16]: The symmetry reduction $P4/ncc$ to $P42_12$ is driven by the one-dimensional irreducible representation τ_2 at the wave vector $\boldsymbol{k} = \boldsymbol{0}$, yielding a one component order parameter Q, which is zero in the paraphase $(P < P_c)$ and nonzero in the distorted phase $(P > P_c)$, suggesting the standard form $\Phi(Q; X) = \frac{A(X)}{2}Q^2 + \frac{B(X)}{4}Q^4 + \frac{C(X)}{6}Q^6$, where $A(\boldsymbol{X}), C(\boldsymbol{X}) > 0$. The tetragonal symmetry also dictates $d_1(\boldsymbol{X}) = d_2(\boldsymbol{X})$. Let $K_0 := 1/\kappa_0$ denote the isothermal bulk modulus at $P = 0$. The Murnaghan equation of state (MEOS) [17]

$$v(P) \equiv (1 + K_0'P/K_0)^{-1/K_0'}, \qquad (18)$$

which is based on the simple ansatz $1/\kappa(P) =: K(P) = K_0 + K_0'P$, is frequently used to describe (P, V) data and reproduces the values of $K(P)$ correctly up to about 10% $[v(P) > 0.9]$ while being algebraically much simpler than other approaches such as the "Vinet" or the "Birch-Murnaghan" EOS used in the compression range $v(P) < 0.9$ (cf., e.g., Refs. [4–7]). Figures 1 and 2 show corresponding fits of unit cell volume and axes of both BaCu(Si_4O_{10}) and BaCr(Si_4O_{10}) using the parameter values of Table II. With these values, Eq. (10) yields possible pressure ranges for hysteresis effects of 2.6–2.9 GPa and 2.2–2.4 GPa for BaCu(Si_4O_{10}) and BaCr(Si_4O_{10}), respectively. From the parameters one also calculates that the geometrical error introduced in assuming the spontaneous strain $\hat{\boldsymbol{\epsilon}}$ to be infinitesimal

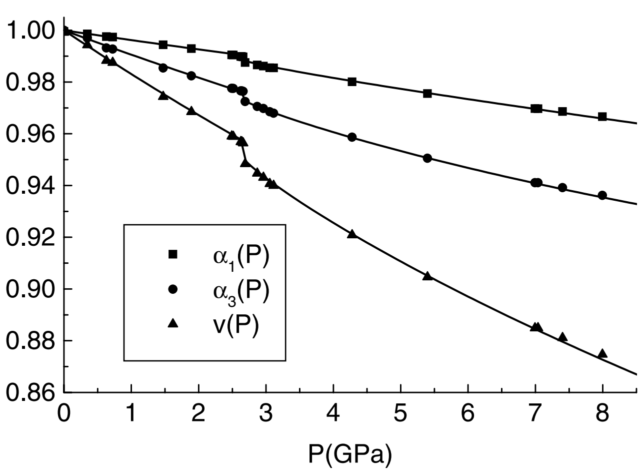

FIG. 1. Fit of the P dependence of reduced lattice parameters $\alpha_1(P), \alpha_3(P)$, and reduced unit cell volume $v(P)$ for BaCu(Si_4O_{10}) using parameters from Table II.

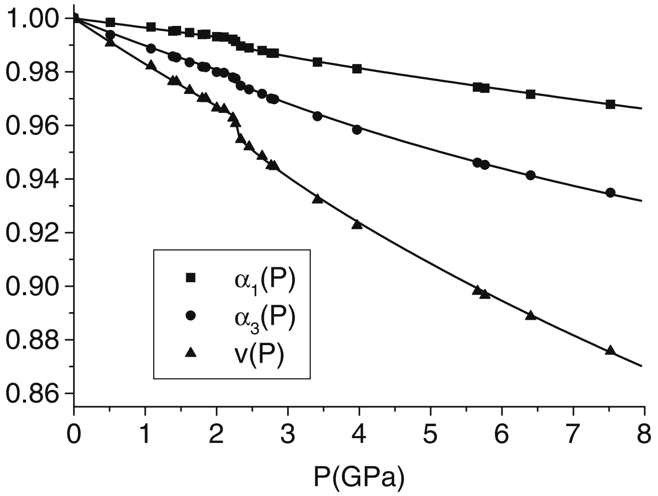

FIG. 2. Fit of the P dependence of reduced lattice parameters $\alpha_1(P), \alpha_3(P)$, and reduced unit cell volume $v(P)$ for BaCr(Si_4O_{10}) using parameters from Table II.

TABLE II. Parameters used in Figs. 1 and 2.

	BuCu(Si_4O_{10})	BaCr(Si_4O_{10})
$A(\boldsymbol{X})$	0.491 GPa	0.449 GPa
$B(\boldsymbol{X})$	-1.0 GPa	-0.2 GPa
$C(\boldsymbol{X})$	15.0 GPa	20 GPa
$d_1(\boldsymbol{X})$	2.82 GPa^{-1}	13.75 GPa^{-1}
$d_3(\boldsymbol{X})$	7.23 GPa^{-1}	-0.33 GPa^{-1}
$a_1(0)$	7.447 Å	7.535 Å
$a_3(0)$	16.14 Å	16.09 Å
κ_1^0	0.0039 GPa^{-1}	0.0035 GPa^{-1}
κ_3^0	0.0097 GPa^{-1}	0.001 085 GPa^{-1}
S_{11}^0	0.0028 GPa^{-1}	0.003 48 GPa^{-1}
S_{33}^0	0.0066 GPa^{-1}	0.0089 GPa^{-1}
K_0'	2.9	4.1
κ_{11}^1	-8.0×10^{-4} GPa^{-2}	1.9×10^{-4} GPa^{-2}
κ_{12}^1	0.53×10^{-4} GPa^{-2}	-1.7×10^{-4} GPa^{-2}
κ_{13}^1	7.4×10^{-4} GPa^{-2}	2.3×10^{-4} GPa^{-2}

is smaller than 0.35% and 0.9%, respectively, yielding an error <0.1% in the total strain $\boldsymbol{\eta}$. In addition, we point out that instead of introducing $q_3 + q_4 = 14$ third and fourth order elastic constants necessary for a fourth order expansion the elastic free energy (cf. Table I), the present approach gets away with just $q_2 - 1 = 3$ additional constants κ_{11}^1, κ_{12}^1, κ_{13}^1 and the parameter K_0' entering the MEOS. Finally, notice that in principle also quantities like soft mode frequencies and even the (P, T) phase diagram at high pressures can be investigated using the present type of approach once we allow for the parameter $A(\boldsymbol{X})$ to be, say, linearly T-dependent and consider a temperature-dependent EOS.

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[12] This does, of course, *not* imply that the *total* strain $\boldsymbol{\eta}$ is assumed to be infinitesimal.

[13] The inverse of a fourth order tensor A_{ijkl} is defined by $\sum_{mn} A_{ijmn} A_{mnkl}^{-1} \equiv \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ (cf. [15]).

[14] Equations (7a), (10), and (11) are reminiscent of the corresponding results of the infinitesimal theory, the tensor $T_{ijkl}(\hat{\boldsymbol{X}})$ being replaced by the inverse zero-stress elastic constants $C_{ijkl}^{-1}(\boldsymbol{X})$.

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