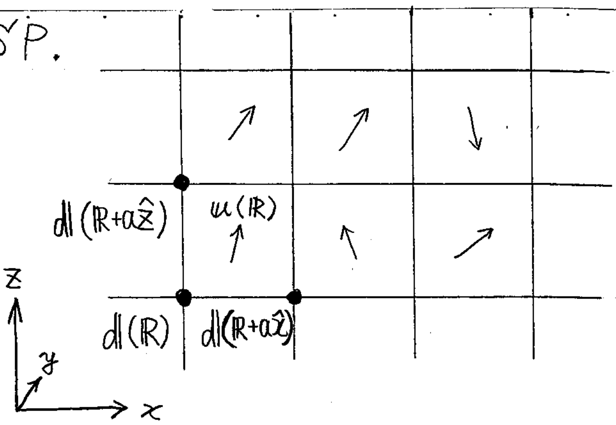
2012-12-11

On acoustic displacements

$\mathbb{d} = \begin{pmatrix} d_x \\ d_y \\ d_z \end{pmatrix}$, Let's use the notations of strain same as chapter 3 of Kittel's ISSP.



$$d_\alpha(\mathbb{R}) = \frac{1}{N}\sum_{\mathbb{k}} \tilde{d}_\alpha(\mathbb{k})\, e^{i\mathbb{k}\cdot\mathbb{R}}$$

$$\tilde{d}_\alpha(\mathbb{k}) = \sum_{\mathbb{R}} d_\alpha(\mathbb{R})\, e^{-i\mathbb{k}\cdot\mathbb{R}}$$

$$\begin{aligned}
\varepsilon_{xx}(\mathbb{R}) &= \frac{1}{a}\left[d_x(\mathbb{R}+a\hat{x}) - d_x(\mathbb{R})\right] \\
&= \frac{1}{\cancel{a}}\frac{\partial d_x(\mathbb{R})}{\partial x}\cancel{a} = \frac{\partial d_x(\mathbb{R})}{\partial x} \\
&= \frac{1}{N}\sum_{\mathbb{k}} \tilde{d}_x(\mathbb{k})\, i k_x\, e^{i\mathbb{k}\cdot\mathbb{R}}
\end{aligned}$$

Fourier coefficients ⟶ Check it!

$$\begin{aligned}
\tilde{\varepsilon}_{xx}(\mathbb{k}) &= \sum_{\mathbb{R}} \varepsilon_{xx}(\mathbb{R})\, e^{-i\mathbb{k}\cdot\mathbb{R}} \\
&= \sum_{\mathbb{R}}\left(\frac{1}{N}\sum_{\mathbb{k}'} \tilde{d}_x(\mathbb{k})\, i k_x\, e^{i\mathbb{k}'\cdot\mathbb{R}}\right) e^{-i\mathbb{k}\cdot\mathbb{R}} \\
&= \frac{1}{N}\sum_{\mathbb{R}}\sum_{\mathbb{k}'} \tilde{d}_x(\mathbb{k})\, i k_x\, e^{i(\mathbb{k}'-\mathbb{k})\cdot\mathbb{R}} \\
&= \frac{1}{N}\sum_{\mathbb{R}} \tilde{d}_x(\mathbb{k})\, i k_x = \tilde{d}_x(\mathbb{k})\, i k_x \qquad \text{—— ☆1}
\end{aligned}$$

⟶ Of course!

Similarly,

$$\varepsilon_{xy}(\mathbb{R}) = \frac{1}{N}\sum_{\mathbb{k}} \tilde{d}_x(\mathbb{k})\, i k_y\, e^{i\mathbb{k}\cdot\mathbb{R}}, \qquad \tilde{\varepsilon}_{xy}(\mathbb{k}) = \tilde{d}_x(\mathbb{k})\, i k_y$$

$$\varepsilon_{yx}(\mathbb{R}) = \frac{1}{N}\sum_{\mathbb{k}} \tilde{d}_y(\mathbb{k})\, i k_x\, e^{i\mathbb{k}\cdot\mathbb{R}}, \qquad \tilde{\varepsilon}_{yx}(\mathbb{k}) = \tilde{d}_y(\mathbb{k})\, i k_y$$

BTW,

$$\begin{aligned}
\eta_6(\mathbb{R}) = e_{xy}(\mathbb{R}) &= \varepsilon_{xy}(\mathbb{R}) + \varepsilon_{yx}(\mathbb{R}) \\
&= \frac{1}{N}\sum_{\mathbb{k}} i\left[d_x(\mathbb{k})k_y + d_y(\mathbb{k})k_x\right] e^{i\mathbb{k}\cdot\mathbb{R}},
\end{aligned}$$

$$\tilde{\eta}_6(\mathbb{k}) = \tilde{e}_{xy}(\mathbb{k}) = i\left[d_x(\mathbb{k})k_y + d_y(\mathbb{k})k_x\right]. \qquad \text{—— ☆2}$$

$$V^{\mathrm{coup,\,inho}}(\mathbb{R})$$

$$= V^{\mathrm{coup,\,inho}}\left(\eta_1(\mathbb{R}), \cdots, \eta_6(\mathbb{R}); y_1(\mathbb{R}), \cdots, y_6(\mathbb{R})\right)$$

$$= \frac{1}{2}\left(\eta_1(\mathbb{R}) \cdots\cdots \eta_6(\mathbb{R})\right) \left(\overleftrightarrow{C} \right) \begin{pmatrix} y_1(\mathbb{R}) \\ y_2(\mathbb{R}) \\ \vdots \\ y_6(\mathbb{R}) \end{pmatrix}$$

$$=$$

Eq. (21) of Nishimatsu 2008

$$\sum_{\mathbb{R}} V^{\mathrm{coup,\,inho}}(\mathbb{R})$$

$$= \frac{1}{2}\sum_{\mathbb{R}} \frac{1}{N}\sum_{\mathbb{k}'} \frac{1}{N}\sum_{\mathbb{k}} {}^t\begin{pmatrix} \tilde{d}_x(\mathbb{k}')\, i k'_x \\ \tilde{d}_y(\mathbb{k}')\, i k'_y \\ \tilde{d}_z(\mathbb{k}')\, i k'_z \\ \tilde{d}_y(\mathbb{k}')\, i k'_z + \tilde{d}_z(\mathbb{k}')\, i k'_y \\ \tilde{d}_z(\mathbb{k}')\, i k'_x + \tilde{d}_x(\mathbb{k}')\, i k'_z \\ \tilde{d}_x(\mathbb{k}')\, i k'_y + \tilde{d}_y(\mathbb{k}')\, i k'_x \end{pmatrix} \Big(C \Big) \begin{pmatrix} \tilde{y}_1(\mathbb{k}) \\ \vdots \\ \tilde{y}_6(\mathbb{k}) \end{pmatrix} \underbrace{e^{i(\mathbb{k}'+\mathbb{k})\cdot\mathbb{R}}}$$

$\mathbb{k}' = -\mathbb{k}$ のときのみ値をもつから、

$$= \frac{1}{2N^2}\sum_{\mathbb{R}}\sum_{\mathbb{k}} {}^t\begin{pmatrix} \tilde{d}_x(-\mathbb{k})(-i k_x) \\ \vdots \\ \tilde{d}_x(-\mathbb{k})(-i k_y) + \tilde{d}_y(-\mathbb{k})(-i k_y) \end{pmatrix} \Big(C \Big) \begin{pmatrix} \tilde{y}_1(\mathbb{k}) \\ \vdots \\ \tilde{y}_6(\mathbb{k}) \end{pmatrix}$$

$$= \frac{1}{2N}\sum_{\mathbb{k}} (-i)\, {}^t\begin{pmatrix} \tilde{d}_x^*(\mathbb{k})\, k_x \\ \vdots \\ \tilde{d}_x^*(\mathbb{k})\, k_y + \tilde{d}_y^*(\mathbb{k})\, k_x \end{pmatrix} \Big(C \Big) \begin{pmatrix} \tilde{y}_1(\mathbb{k}) \\ \vdots \\ \tilde{y}_6(\mathbb{k}) \end{pmatrix} \quad \cdots\cdots (22)$$

$$x^*(\mathbb{k})\, C\, y(\mathbb{k}) + x^*(-\mathbb{k})\, C\, y(-\mathbb{k})$$

$$= x^*(\mathbb{k})\, C\, y(\mathbb{k}) + x(\mathbb{k})\, C\, y^*(\mathbb{k})$$

$$= x^*(\mathbb{k})\, C\, y(\mathbb{k}) + \left[x^*(\mathbb{k})\, C\, y(\mathbb{k})\right]^*$$

$$= 2\,\mathrm{Re}\left[x^*(\mathbb{k})\, C\, y(\mathbb{k})\right]$$

2ページ前にテストの様子を貼付けた。

energy_module.F (rev1617) の下のほうで、{$\mathbb{k}$}のうち半分で sumation

2012-12-12

K

$$
(\tilde{d}_x \ \tilde{d}_y \ \tilde{d}_z)\begin{pmatrix} k_x & 0 & 0 & 0 & k_z & k_y \\ 0 & k_y & 0 & k_z & 0 & k_x \\ 0 & 0 & k_z & k_y & k_x & 0 \end{pmatrix}
$$

$$
= (\tilde{d}_x k_x \quad \tilde{d}_y k_y \quad \tilde{d}_z k_z \quad \tilde{d}_y k_z + \tilde{d}_z k_y \quad \tilde{d}_z k_x + \tilde{d}_x k_z \quad \tilde{d}_x k_y + \tilde{d}_y k_x)
$$

$$
\begin{pmatrix} k_x & 0 & 0 & 0 & k_z & k_y \\ 0 & k_y & 0 & k_z & 0 & k_x \\ 0 & 0 & k_z & k_y & k_x & 0 \end{pmatrix}
\begin{pmatrix} B_{1xx} & B_{1yy} & B_{1yy} & & & \\ B_{1yy} & B_{1xx} & B_{1yy} & & 0 & \\ B_{1yy} & B_{1yy} & B_{1xx} & & & \\ & & & 2B_{4yz} & 0 & 0 \\ & 0 & & 0 & 2B_{4yz} & 0 \\ & & & 0 & 0 & 2B_{4yz} \end{pmatrix}
$$

$$
= \begin{pmatrix} k_x B_{1xx} & k_x B_{1yy} & k_x B_{1yy} & 0 & 2k_z B_{4yz} & 2k_y B_{4yz} \\ k_y B_{1yy} & k_y B_{1xx} & k_y B_{1yy} & 2k_z B_{4yz} & 0 & 2k_x B_{4yz} \\ k_z B_{1yy} & k_z B_{1yy} & k_z B_{1xx} & 2k_y B_{4yz} & 2k_x B_{4yz} & 0 \end{pmatrix}
$$

Eq. (16)

$$
V^{elas,homo} = \frac{1}{2}(\eta_1 \cdots \eta_6)\begin{pmatrix} B_{11} & B_{12} & B_{12} & & & \\ B_{12} & B_{11} & B_{12} & & 0 & \\ B_{12} & B_{12} & B_{11} & & & \\ & & & B_{44} & 0 & 0 \\ & 0 & & 0 & B_{44} & 0 \\ & & & 0 & 0 & B_{44} \end{pmatrix}\begin{pmatrix} \eta_1 \\ \eta_2 \\ \eta_3 \\ \eta_4 \\ \eta_5 \\ \eta_6 \end{pmatrix}
$$

Eq. (17), (18), (19)

$$
\Phi^{elas,inho}(\mathbb{k}) = \frac{1}{N} K B {}^t K
$$

Eq. (24), (25) $\Rightarrow$ $V^{coup,\left\{\substack{homo \\ inho}\right\}} = -2V^{elas,\left\{\substack{homo \\ inho}\right\}}$

2012-12-12

B. Molecular dynamics

MD simulations with the effective Hamiltonian of Eq. (4) are performed in the canonical ensemble using the Nosé-Poincaré thermostat.[16] This simplectic thermostat is so efficient that we can set the time step to $\Delta t = 2$ fs. In our present simulations, we thermalize the system for 40,000 time steps, after which we average the properties for 10,000 time steps.

In Fig. 1 we roughly illustrate how to calculate the forces exerted on $u_\alpha(\mathbf{R})$ with $\tilde\Phi^{\text{quad}}_{\alpha\beta}(\mathbf{k})$ in Eq. (13) and how the time evolution is simulated. First, $u_\alpha(\mathbf{R})$ is FFTed to $\tilde u_\alpha(\mathbf{k})$, the force $\tilde F_\alpha(\mathbf{k}) = -\Sigma_\beta \tilde\Phi^{\text{quad}}_{\alpha\beta}(\mathbf{k})\tilde u_\beta(\mathbf{k})$ is calculated in reciprocal space, and then the force in real space is obtained by the inverse FFT (IFFT) of $\tilde F_\alpha(\mathbf{k})$. In practice, updates of $u_\alpha(\mathbf{R})$ and $\dot u_\alpha(\mathbf{R}) = \frac{\partial}{\partial t}u_\alpha(\mathbf{R})$ are processed in the manner of the Nosé-Poincaré thermostat.

The homogeneous strain components $\eta_1, \cdots, \eta_6$ are determined by solving

$$\frac{\partial}{\partial \eta_i}[V^{\text{elas,homo}}(\eta_1, \cdots, \eta_6) + V^{\text{coup,homo}}(\{\mathbf{u}\}, \eta_1, \cdots, \eta_6)] = 0 \tag{24}$$

at each time step according to $\{\mathbf{u}\}$ so that $\eta_1, \cdots, \eta_6$ minimize $V^{\text{elas,homo}}(\eta_1, \cdots, \eta_6) + V^{\text{coup,homo}}(\{\mathbf{u}\}, \eta_1, \cdots, \eta_6)$. While the local acoustic displacement $w_\alpha(\mathbf{R})$ could be treated as dynamical variables using the effective mass M^*_{acoustic}, we have instead chosen to integrate out these variables in a manner similar to the treatment of the homogeneous strain. That is, $w_\alpha(\mathbf{R})$ is determined so that $V^{\text{elas,inho}}(\{\mathbf{w}\}) + V^{\text{coup,inho}}(\{\mathbf{u}\},\{\mathbf{w}\})$ becomes minimum at each time step according to $u_\alpha(\mathbf{R})$. Technically, the minimization is performed by solving the linear set of equations

$$\tilde\Phi^{\text{elas,inho}}(\mathbf{k})\tilde{\mathbf{w}}(\mathbf{k}) + \tilde{\mathbf{B}}(\mathbf{k})\tilde{\mathbf{y}}(\mathbf{k}) = 0 \tag{25}$$

for each $\mathbf{k}$ in reciprocal space.

The homogeneous elastic energy $V^{\text{elas,homo}}(\eta_1, \cdots, \eta_6)$ is

$$\begin{aligned} V^{\text{elas,homo}}(\eta_1, \cdots, \eta_6) = {} & \frac{N}{2}B_{11}(\eta_1^2 + \eta_2^2 + \eta_3^2) \\ & + NB_{12}(\eta_2\eta_3 + \eta_3\eta_1 + \eta_1\eta_2) \\ & + \frac{N}{2}B_{44}(\eta_4^2 + \eta_5^2 + \eta_6^2), \end{aligned} \tag{16}$$

where B_{11}, B_{12}, and B_{44} are the elastic constants expressed in energy unit ($B_{11} = a_0^3 C_{11}$, $B_{12} = a_0^3 C_{12}$, and $B_{44} = a_0^3 C_{44}$).

The inhomogeneous elastic energy $V^{\text{elas,inho}}(\{\mathbf{w}\})$ is also calculated in reciprocal space as

(-i) 消える! i

$$V^{\text{elas,inho}}(\{\mathbf{w}\}) = \frac{1}{2}\sum_{\mathbf{k}}\sum_{\alpha,\beta}\tilde w^*_\alpha(\mathbf{k})\tilde\Phi^{\text{elas,inho}}_{\alpha\beta}(\mathbf{k})\tilde w_\beta(\mathbf{k}). \tag{17}$$

For the *force constant* matrix $\tilde\Phi^{\text{elas,inho}}_{\alpha\beta}(\mathbf{k})$, we employed the long-wavelength approximation. For instance, the diagonal part is

$$\tilde\Phi^{\text{elas,inho}}_{\cancel{\alpha\beta}\ xx}(\mathbf{k}) = \frac{1}{N}[k_x^2 B_{11} + k_y^2 B_{44} + k_z^2 B_{44}], \tag{18}$$

and the off-diagonal part is

$$\tilde\Phi^{\text{elas,inho}}_{\cancel{\alpha\beta}\ xy}(\mathbf{k}) = \frac{1}{N}[k_x k_y B_{12} + k_x k_y B_{44}]. \tag{19}$$

The coupling between $\{\mathbf{u}\}$ and homogeneous strain is the same as that given in Ref. 9, i.e.,

$$V^{\text{coup,homo}}(\{\mathbf{u}\}, \eta_1, \cdots, \eta_6) = \frac{1}{2}\sum_{\mathbf{R}}\sum_{i=1}^{6}\sum_{j=1}^{6}\eta_i C_{ij} y_j(\mathbf{R}). \tag{20}$$

Here, $y_1(\mathbf{R}) = u_x^2(\mathbf{R})$, $y_2(\mathbf{R}) = u_y^2(\mathbf{R})$, $y_3(\mathbf{R}) = u_z^2(\mathbf{R})$, $y_4(\mathbf{R}) = u_y(\mathbf{R})u_z(\mathbf{R})$, $y_5(\mathbf{R}) = u_z(\mathbf{R})u_x(\mathbf{R})$, and $y_6(\mathbf{R}) = u_x(\mathbf{R})u_y(\mathbf{R})$,

$$\mathbf{C} = \begin{pmatrix} B_{1xx} & B_{1yy} & B_{1yy} & 0 & 0 & 0 \\ B_{1yy} & B_{1xx} & B_{1yy} & 0 & 0 & 0 \\ B_{1yy} & B_{1yy} & B_{1xx} & 0 & 0 & 0 \\ 0 & 0 & 0 & 2B_{4yz} & 0 & 0 \\ 0 & 0 & 0 & 0 & 2B_{4yz} & 0 \\ 0 & 0 & 0 & 0 & 0 & 2B_{4yz} \end{pmatrix}, \tag{21}$$

and B_{1xx}, B_{1yy}, and B_{4yz} are the coupling coefficients defined in Ref. 9.

The coupling between $\{\mathbf{u}\}$ and inhomogeneous strain is also calculated in reciprocal space as

$$V^{\text{coup,inho}}(\{\mathbf{u}\},\{\mathbf{w}\}) = \frac{1}{2}\sum_{\mathbf{k}}\sum_{\alpha}\sum_{i=1}^{6}\tilde w^*_\alpha(\mathbf{k})\tilde B_{\alpha i}(\mathbf{k})\tilde y_i(\mathbf{k}), \tag{22}$$

where $\tilde w_\alpha(\mathbf{k})$ and $\tilde y_i(\mathbf{k})$ are the Fourier transforms of $w_\alpha(\mathbf{R})$ and $y_i(\mathbf{R})$, respectively. For the 3×6 coupling matrix $\mathbf{B}(\mathbf{k})$, we again employed the long-wavelength approximation

$$\tilde{\mathbf{B}}(\mathbf{k}) = \frac{1}{N}\begin{pmatrix} k_x B_{1xx} & k_x B_{1yy} & k_x B_{1yy} & 0 & 2k_z B_{4yz} & 2k_y B_{4yz} \\ k_y B_{1yy} & k_y B_{1xx} & k_y B_{1yy} & 2k_z B_{4yz} & 0 & 2k_x B_{4yz} \\ k_z B_{1yy} & k_z B_{1yy} & k_z B_{1xx} & 2k_y B_{4yz} & 2k_x B_{4yz} & 0 \end{pmatrix}. \tag{23}$$

$\frac{1}{N}$ is O.K.
See Eq. (14)

In the present MD simulations of $BaTiO_3$, the parameters from Refs. 10 and 11, which are determined by first-principles calculations, are employed. As mentioned in Refs. 10 and 11, this parameter set leads to an underestimation of the Tc.

2012-12-18

On optimize-inho-strain.F and energy_module.F,

if we use $d_\alpha(\mathbb{k})$,

$$V^{elas,inho}(\{\mathbb{d}(\mathbb{k})\}) = \frac{1}{2}\sum_{\mathbb{k}} \tilde{\mathbb{d}}^{+}(\mathbb{k})\,\tilde{\Phi}(\mathbb{k})\,\mathbb{d}(\mathbb{k})$$

$$V^{coup,inho}(\{\mathbb{d}(\mathbb{k})\},\{\mathbb{y}(\mathbb{k})\}) = (-i)\sum_{\mathbb{k}} \tilde{\mathbb{d}}^{+}(\mathbb{k})\left(\frac{1}{2}\tilde{B}(\mathbb{k})\right)\tilde{\mathbb{y}}(\mathbb{k}) .$$

To minimize sum of them, $V^{elas,inho}(\{\mathbb{d}(\mathbb{k})\}) + V^{coup,inho}(\{\mathbb{d}(\mathbb{k})\},\{\mathbb{y}(\mathbb{k})\})$,

$$\tilde{\Phi}(\mathbb{k})\,\mathbb{d}(\mathbb{k}) - i\left(\frac{1}{2}\tilde{B}(\mathbb{k})\right)\tilde{\mathbb{y}}(\mathbb{k}) = 0 . \qquad (25)'$$

Now, let's define $\tilde{w}_\alpha(\mathbb{k})$ with

$$\tilde{d}_\alpha(\mathbb{k}) = \left[-i\,\tilde{w}_\alpha(\mathbb{k})\right] ,$$

$$\tilde{d}_\alpha^*(\mathbb{k}) = i\,\tilde{w}_\alpha^*(\mathbb{k}) .$$

In other words

$$d_\alpha(\mathbb{R}) = \frac{1}{N}\sum_{\mathbb{k}}\left[-i\,\tilde{w}_\alpha(\mathbb{k})\right] e^{i\mathbb{k}\cdot\mathbb{R}}$$

and

$$\left[-i\tilde{w}(\mathbb{k})\right] = \sum_{\mathbb{R}} d_\alpha(\mathbb{R})\, e^{-i\mathbb{k}\cdot\mathbb{R}} .$$

Using this $\tilde{w}_\alpha(\mathbb{k})$, Eq. ☆1 and ☆2 become

$$\tilde{\varepsilon}_{xx}(\mathbb{k}) = \tilde{e}_{xx}(\mathbb{k}) = \eta_1(\mathbb{k}) = \tilde{w}_x(\mathbb{k})\,k_x$$

and

$$\tilde{e}_{xy}(\mathbb{k}) = \tilde{\eta}_6(\mathbb{k})$$

$$= \tilde{\varepsilon}_{xy}(\mathbb{k}) + \tilde{\varepsilon}_{yx}(\mathbb{k}) = \tilde{w}_x(\mathbb{k})\,k_y + \tilde{w}_y(\mathbb{k})\,k_x .$$

Eq. (22)' becomes Eq. (22) in [Nishimatsu 2008].

And, Eq. (25)' becomes that of Eq. (25).

Up until now (and from now on), optimize-info-strain.F and energy_module.F are inplemented with this $\tilde{w}_\alpha(\mathbb{k})$, acouK(ix,iy,iz,$\alpha$).