Fully-Nonlocal Pseudopotential

12/14/99

— Problem of seminonlocal pseudopotential.

$$\begin{cases} V_{ion}^{PP}(r) = V_{ion,local}^{PP}(r) + \underbrace{\sum_{lm} |lm\rangle \Delta V_l(r) \langle lm|}_{\hat{V}_{NL}} & (1) \\ \Delta V_l(r) = V_{ion,l}^{PP}(r) - V_{ion,local}^{PP}(r) & (2) \end{cases}$$

This is local in r and nonlocal (i.e. separable) in the angular coordinates.

$$\langle \mathbf{k}+\mathbf{G} | \hat{V}_{NL} | \mathbf{k}+\mathbf{G}' \rangle = \sum_l \frac{4\pi(2l+1)}{\Omega} \int dr\, r^2 j_l(|\mathbf{k}+\mathbf{G}|r)\, \Delta V_l(r)\, j_l(|\mathbf{k}+\mathbf{G}'|r) \qquad (3)$$

For each ion, this involves $O(N_{PW}^2)$ radial integration, hence the operation count is $O(N_I N_{PW}^2) \propto O(N_I^3)$ where N_I is the number of ions and N_{PW} is the number of plane waves.

— Idea

If the nonlocal pseudopotential is fully (including the radial part) separable, the source and destination integrals are evaluated independently, hence $O(N_I N_{PW}^2) \longrightarrow O(N_I N_{PW}) \propto O(N_I^2)$.

(cf. Fast multipole method: source—multipole; destination—Taylor.)

Fully nonlocal pseudopotential

[L. Kleinman & D.M. Bylander, Phys. Rev. Lett. 48, 1425 ('82)]

Let's replace the local (in radial coordinate) potential

$$\Delta V_\ell(r) \rightarrow \frac{|\Delta V_\ell R_\ell^{PP}\rangle\langle R_\ell^{PP}\Delta V_\ell|}{\langle R_\ell^{PP}|\Delta V_\ell|R_\ell^{PP}\rangle} \tag{4}$$

where

$$\langle R_\ell^{PP}|\Delta V_\ell|R_\ell^{PP}\rangle = \int dr\, r^2\, |R_\ell^{PP}(r)|^2 \Delta V_\ell(r) \tag{5}$$

$$\boxed{V^{KB}(r) = V_{ion,local}^{PP}(r) + \sum_{\ell m} \frac{|\ell m\rangle|\Delta V_\ell R_\ell^{PP}\rangle\langle R_\ell^{PP}\Delta V_\ell|\langle \ell m|}{\langle R_\ell^{PP}|\Delta V_\ell|R_\ell^{PP}\rangle}} \tag{6}$$

(Prop.) The Kleinman-Bylander pseudopotential is identical to the original seminonlocal pseudopotential, when it operates on the atomic pseudowave function, $R_\ell^{PP}(r)$.

$\because$

$$\frac{|\Delta V_\ell R_\ell^{PP}\rangle\langle\Delta V_\ell R_\ell^{PP}|}{\langle R_\ell^{PP}|\Delta V_\ell|R_\ell^{PP}\rangle}|R_\ell^{PP}\rangle$$

$$= \Delta V_\ell(r) R_\ell^{PP}(r) \times \cancel{\frac{\int dr\, r^2 [R_\ell^{PP}(r)\Delta V_\ell(r)]\, R_\ell^{PP}(r)}{\int dr\, r^2\, |R_\ell^{PP}(r)|^2 \Delta V_\ell(r)}}$$

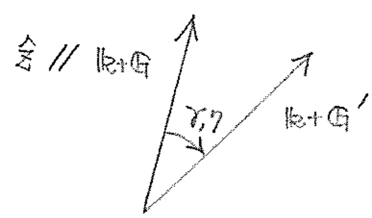
$$= \Delta V_\ell(r) R_\ell^{PP}(r) \quad //$$

- Operation count

$$V^{NL}_{GG'} = \sum_{lm} \frac{\langle k+G|lm\rangle|\Delta V_l R_l^{PP}\rangle\langle \Delta V_l R_l^{PP}|\langle lm|k+G'\rangle}{\langle R_l^{PP}|\Delta V_l|R_l^{PP}\rangle} \tag{7}$$

From 12/14/99, (we absorb the volume factor in the plane-wave basis.)

$$\langle lm|k+G'\rangle = 4\pi i^l j_l(|k+G'|r)\, Y^*_{lm}(\gamma,\eta)\frac{1}{\sqrt{\Omega}} \tag{8}$$



$$\therefore \langle \Delta V_l R_l^{PP}|\langle lm|k+G'\rangle = \frac{4\pi i^l Y^*_{lm}(\gamma,\eta)}{\sqrt{\Omega}}\int dr\, r^2 R_l^{PP}(r)\,\Delta V_l(r)\, j_l(|k+G'|r) \tag{9}$$

Similarly from 12/14/99,

$$\langle k+G|lm\rangle = (-i)^l\sqrt{4\pi(2l+1)}\, j_l(|k+G|r)\,\delta_{m0}\frac{1}{\sqrt{\Omega}} \tag{10}$$

$$\therefore \langle k+G|lm\rangle|\Delta V_l R_l^{PP}\rangle = \frac{(-i)^l\sqrt{4\pi(2l+1)}\,\delta_{m0}}{\sqrt{\Omega}}\int dr\, r^2 j_l(|k+G|r)\,\Delta V_l(r)\, R_l^{PP}(r) \tag{11}$$

Substituting Eqs. (9) and (11) in (7),

$$V^{NL}_{GG'} = \frac{\cancel{\sum_{lm}} \frac{4\pi \cancel{i^l}}{\sqrt{\Omega}} \underbrace{Y^*_{lm}(\gamma,\eta)}_{\cancel{\sqrt{\frac{2l+1}{4\pi}}} P_l(\cos\gamma)} \cancel{(-i)^l} \frac{\cancel{\sqrt{4\pi(2l+1)}\,\delta_{m0}}}{\sqrt{\Omega}} \int dr\, r^2 R_l^{PP}(r)\,\Delta V_l(r)\, j_l(|k+G'|r) \times \int dr\, r^2 j_l(|k+G|r)\,\Delta V_l(r)\, R_l^{PP}(r)}{\int dr\, r^2 |R_l^{PP}(r)|^2 \Delta V_l(r)}$$

$$\boxed{v^{NL}_{\mathbb{G}\mathbb{G}'} = \sum_{\ell} \frac{4\pi(2\ell+1)}{\Omega} P_\ell(\cos\gamma) \frac{\int dr\, r^2 R^{PP}_\ell(r)\Delta V_\ell(r)\, j_\ell(|\mathbb{k}+\mathbb{G}|r) \int dr\, r^2 R^{PP}_\ell(r)\Delta V_\ell(r)\, j_\ell(|\mathbb{k}+\mathbb{G}'|r)}{\int dr\, r^2 |R^{PP}_\ell(r)|^2 \Delta V_\ell(r)}} \tag{12}$$

— Operation count

$(v a)_{\mathbb{k}+\mathbb{G}} \leftarrow 0$

for each ion I,

calculate $A_\ell = \int dr\, r^2 |R^{PP}_\ell(r)|^2 \Delta V_\ell(r)$

for each plane wave $\mathbb{k}+\mathbb{G}$,

calculate $B_\ell(\mathbb{k}+\mathbb{G}) = \int dr\, r^2 R^{PP}_\ell(r)\Delta V_\ell(r)\, j_\ell(|\mathbb{k}+\mathbb{G}|r)$

for each plane wave $\mathbb{k}+\mathbb{G}$,

$$(v a)_{\mathbb{k}+\mathbb{G}} \mathrel{+}= \sum_\ell \frac{4\pi(2\ell+1)}{\Omega} P_\ell(\cos\gamma) \frac{B_\ell(\mathbb{k}+\mathbb{G})\, B_\ell(\mathbb{k}+\mathbb{G}')}{A_\ell}$$

The operation count is still $O(N_I N_{PW}^2)$. However, the number of expensive radial integrations, i.e. the calculation of $B_\ell(\mathbb{k}+\mathbb{G})$ is now $O(N_I N_{PW})$. We precalculate $B_\ell(\mathbb{k}+\mathbb{G})$ before the N_{PW}^2 loop.