XL-BOMD for HF/DFT + U

$$\Omega(\mathbf{R}) = \min_{D\in\mathcal{N}} \{2\mathrm{Tr}[hD] + \mathrm{Tr}[DG(D)] + \mathrm{Tr}[U(DS - DSDS)] - 2T_e\mathcal{S}[D]\} + V_{nn}(\mathbf{R})$$

$$\Omega(\mathbf{R}, P) = \min_{D\in\mathcal{N}} \{2\mathrm{Tr}[hD] + \mathrm{Tr}[(2D - P)G(P)] + \mathrm{Tr}[U(DS - PSDS - DSPS + PSPS)] - 2T_e\mathcal{S}[D]\} + V_{nn}(\mathbf{R})$$

$$H = h + G(P) + \frac{1}{4}(SU - SUPS - SPSU + US - SPUS - USPS)$$

$$D^{\perp} = \left[e^{\beta(H^{\perp}-\mu I)} + I\right]^{-1} \qquad H^{\perp} = Z^T H Z, \quad D = ZD^{\perp}Z^T, \quad P = ZP^{\perp}Z^T, \quad Z^T S Z = I$$

$$\mathcal{L}(\mathbf{R}, \dot{\mathbf{R}}, P, \dot{P}) = \frac{1}{2}\sum_I M_I \dot{R}_I^2 - \Omega(\mathbf{R}, P) + \frac{1}{2}\mu\mathrm{Tr}[\dot{P}^{\perp^2}] - \frac{1}{2}\mu\omega^2\mathrm{Tr}\left[(D^{\perp} - P^{\perp})^T\mathcal{K}^T\mathcal{K}(D^{\perp} - P^{\perp})\right]$$

$$M_I\ddot{R}_I = -\left.\frac{\partial\Omega(\mathbf{R}, P)}{\partial R_I}\right|_P \qquad c \in [0, 1]$$

$$\ddot{P}^{\perp} = -\omega^2\mathcal{K}(D^{\perp} - P^{\perp}) \approx \omega^2 c\Delta D + \frac{\omega^2 c^2\|\Delta D\|_{\mathrm{F}}}{1 - c\ \mathrm{Tr}[\overline{\Delta D}^T V]}V$$

$$V = \left.\frac{\partial\left[e^{\beta(H^{\perp}[D^{\perp}+\lambda\overline{\Delta D}]-\mu I)} + I\right]^{-1}}{\partial\lambda}\right|_{\lambda=0} + \frac{1-c}{c}\,\overline{\Delta D} \qquad \Delta D = D^{\perp} - P^{\perp} \qquad \overline{\Delta D} = \Delta D/\|\Delta D\|_F$$

$$\Omega(\mathbf{R}, P) = \min_{D \in \mathcal{N}} \{2\mathrm{Tr}[hD] + \mathrm{Tr}[(2D - P)G(P)] + \mathrm{Tr}[U(DS - PSDS - DSPS + PSPS)] - 2T_e\mathcal{S}[D]\} + V_{nn}(\mathbf{R})$$