Toward sharp error analysis of extended Langrangian molecular dynamics for polarizable force field simulation

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Ki-Net Young Researchers Workshop
UMD, October 2019

Background

- In molecular dynamics (MD), interested in evolution of state vector $r = (r^{(1)}, \ldots, r^{(N)}) \in \mathbb{R}^d$ of atomic positions $r^{(i)} \in \mathbb{R}^3$ (so $d = 3N$).
- Often a *latent variable* $x = \Phi(r) \in \mathbb{R}^{d'}$ needs to be computed as auxiliary step to perform the evolution, i.e., dynamics given by

$$\ddot{r}(t) = G(r(t), x(t)), \quad \text{where} \quad x(t) = \Phi(r(t)).$$

- Φ can be complicated, e.g., in *ab initio* MD simulations, one must solve a quantum many-body problem at each time step to compute the forces!
  - In practice, within DFT approximation, must solve nonlinear eigenvalue problem at each time step.
- Spoiler: we will study case $x(t) = A(r)^{-1}\, b(r)$, to be motivated later. Must solve linear system at each time step.
- Maintain general perspective for the time being.

Background

- More structure: dynamics of (r, x) are Hamiltonian with respect to

$$H(r, x, p, \phi) = \frac{1}{2}|p|^2 + U(r) + Q(r, x),$$

where ϕ is the momentum variable for x, viewed as having zero mass. (Mass of all particles taken to be 1 for simplicity.)

- What I really mean is that the auxiliary variable x has $\varepsilon \to 0$ mass, so consider

$$H_\varepsilon(r, x, p, \phi) = \frac{1}{2}|p|^2 + \frac{\varepsilon}{2}|\phi|^2 + U(r) + Q(r, x).$$

- This yields dynamics:

$$\ddot{r}_\varepsilon(t) = -\frac{\partial U}{\partial r}(r_\varepsilon(t)) - \frac{\partial Q}{\partial r}(r_\varepsilon(t), x_\varepsilon(t))$$
$$\varepsilon\ddot{x}_\varepsilon(t) = -\frac{\partial Q}{\partial x}(r_\varepsilon(t), x_\varepsilon(t)).$$

Background

- Take limit:

$$\ddot{r}(t) = -\frac{\partial U}{\partial r}(r(t)) - \frac{\partial Q}{\partial r}(r(t), x(t))$$
$$0 = -\frac{\partial Q}{\partial x}(r(t), x(t)).$$

- Map $x = \Phi(r)$ implicitly defined by solving $\frac{\partial Q}{\partial x}(r, x) = 0$ for given r.
- Don't think of this as a 'derivation' of our model...will define specific model shortly.
- But it is a way to understand the structure, and the practical method that we consider we will *reverse* these steps.

Model

- Classical molecular dynamics with polarizable force field.
- Potential energy landscape $U(r)$ includes:
  - 'nonbonded terms' (e.g., permanent electrostatic and van der Waals interactions).
  - 'bonded valence terms' (bond-stretching, angle-bending, etc.).
- $x = (x^{(1)}, \cdots, x^{(N)}) \in \mathbb{R}^d$ is the vector of induced dipoles associated to each atom (so $d = d' = 3N$).
- Contribution of induced dipole given by

$$Q(r, x) = \frac{1}{2} x^\top A(r) x - b(r)^\top x,$$

  where
  - $b^{(i)}(r)$ indicates the electric field felt by the i-th atom due to the permanent electrostatics of the system in configuration r.
  - $A(r) \succeq 0$ specifies the interaction of the induced dipoles with themselves and each other.
    - $A^{(i,j)}(r) = \delta_{ij}[\alpha^{(i)}]^{-1} I_3 + T^{(i,j)}(r)$, where $\alpha^{(i)}$ is the atomic polarizability of the i-th atom, $T^{(i,j)} \in \mathbb{R}^{3\times 3}$ is the induced dipole interaction matrix.

Model summary

- Plugging in Q to our earlier formulas, the ('massless') dynamics are given by differential-algebraic system

$$\ddot{r}(t) = G(r(t), x(t)))$$
$$0 = b(r(t)) - A(r(t))x(t),$$

where

$$G(r, x) = -\frac{\partial U}{\partial r}(r) - \frac{1}{2}x^\top \frac{\partial A}{\partial r}(r)x - \frac{\partial b^\top}{\partial r}(r)x.$$

- Computational bottleneck: apparently have to invert $A(r)$ at each time-step in numerical integration.
- Aside: our main analysis will work for quite general G (and even $d' \neq d$), but...
  - We do use Hamiltonian structure to guarantee some useful global-in-time *a priori* bounds.
  - For numerical discretization, Hamiltonian structure exploited by symplectic integrators for long-time stability.

Extended Lagrangian scheme

- *Extended Lagrangian* approach solves the $\varepsilon > 0$ ('massive') dynamics from before:

$$\ddot{r}_\varepsilon(t) = G(r_\varepsilon(t), x_\varepsilon(t)))$$
$$\varepsilon\ddot{x}_\varepsilon(t) = b(r_\varepsilon(t)) - A(r_\varepsilon(t))x_\varepsilon(t).$$

- In this setting, called the 'inertial extended Lagrangian SCF-free' or 'iEL/0-SCF' method [Albaugh, Niklasson, Head-Gordon 2017].
  - SCF = self-consistent field iteration.
  - 0-SCF means here that you don't have solve linear systems at each step.
- Can consider extended Lagrangian molecular dynamics (XLMD) for more general Q (e.g., XL Born-Oppenheimer MD (XL-BOMD) for *ab initio* simulations [Niklasson et al 2006; Niklasson 2008; Niklasson, Cawkwell 2012]).
- Auxiliary dynamics oscillatory on time scale $\sim \sqrt{\varepsilon}$, hence must take time steps at least this small.
- With careful choice of ε, iEL/0-SCF (and related methods) can outperform discretizations of the original dynamics in terms of efficiency and long-time stability while still maintaining the accuracy for the atomic trajectories.

Assumptions

- We consider a fixed time interval $[0, T]$ (independent of ε).
- Let $(r_\star, p_\star)$, where $p_\star := \dot{r}_\star$, be the solution of the massless equations, given fixed initial conditions $r_\star(0)$, $p_\star(0)$.
- Let $(r_\varepsilon, p_\varepsilon, x_\varepsilon, \dot{x}_\varepsilon)$ be the solution of the ε-massive equation, given initial conditions $r_\varepsilon(0) = r_\star(0)$, $p_\varepsilon(0) = p_\star(0)$, as well as $x_\varepsilon(0), \dot{x}_\varepsilon(0)$ arbitrary (for now) other than being bounded independent of ε.
- Technical assumptions:
  1. $A : \mathbb{R}^d \to \mathcal{S}^d_{++}$ is a C^3 map, and there exists $C > 0$ such that $A(r) \succeq C^{-1}$ for all $r \in \mathbb{R}^d$.
  2. $b : \mathbb{R}^d \to \mathbb{R}^d$ is a bounded C^3 map.
  3. U is bounded below, and $F := -\frac{\partial U}{\partial r} : \mathbb{R}^d \to \mathbb{R}^d$ is a locally Lipschitz C^0 map.

Proposition (*a priori* bounds)

Under assumptions $(1), (2), (3)$*, there exists* C *such that both* $|x_\varepsilon| \leq C$ *and* $\left|\frac{d^k r_\star}{dt^k}\right|, \left|\frac{d^k r_\varepsilon}{dt^k}\right| \leq C$ *for* $k = 0, 1, 2$ *and on the entire time interval* $[0, T]$*.*

Goals and results

- Want to study the convergence of $(r_\varepsilon, p_\varepsilon)$ to $(r_\star, p_\star)$ as $\varepsilon \to 0$, as well as the convergence $x_\varepsilon \to x_\star$, where $x_\star(t) := A(r_\star(t))^{-1} b(r_\star(t))$.
- What about the initial condition for $x_\varepsilon, \dot{x}_\varepsilon$ again? Three cases:
  - *Compatible*: choose $x_\varepsilon(0) = x_\star(0) = A(r_\star(0))^{-1} b(r_\star(0))$, but can take $\dot{x}_\varepsilon(0)$ arbitrary.
  - *Optimally compatible*: compatible but also take $\dot{x}_\varepsilon(0) = \dot{x}_\star(0)$ (can get explicit formula).
  - *Incompatible*: otherwise.
- Mostly care about accuracy of r_ε. Numerical experiments say:
  - *Compatible*: $r_\varepsilon - r_\star = O(\varepsilon)$, $x_\varepsilon - x_\star = O(\sqrt{\varepsilon})$.
  - *Optimally compatible*: $r_\varepsilon - r_\star = O(\varepsilon)$, $x_\varepsilon - x_\star = O(\varepsilon)$.
  - *Incompatible*: no convergence of anything.

Theorem

For compatible initial condition, it holds that $r_\varepsilon - r_\star, x_\varepsilon - x_\star = O(\sqrt{\varepsilon})$. In dimension $d = 1$, all the sharp rates mentioned above hold.

Remark

Sharp analysis in $d = 1$ should extend, but it's at least a bit more difficult. (It is only the dimension of the auxiliary variable x that matters.)

Numerical example

$$U(r) = r_1^2 + r_2^2 = |r|^2, \quad A(r) = \begin{pmatrix} 2+|r|^2 & |r|^2 \\ |r|^2 & 1+|r|^2 \end{pmatrix}$$

$$b(r) = (\sin(r_1 + r_2), \cos(r_1 - 2r_2))^\top$$

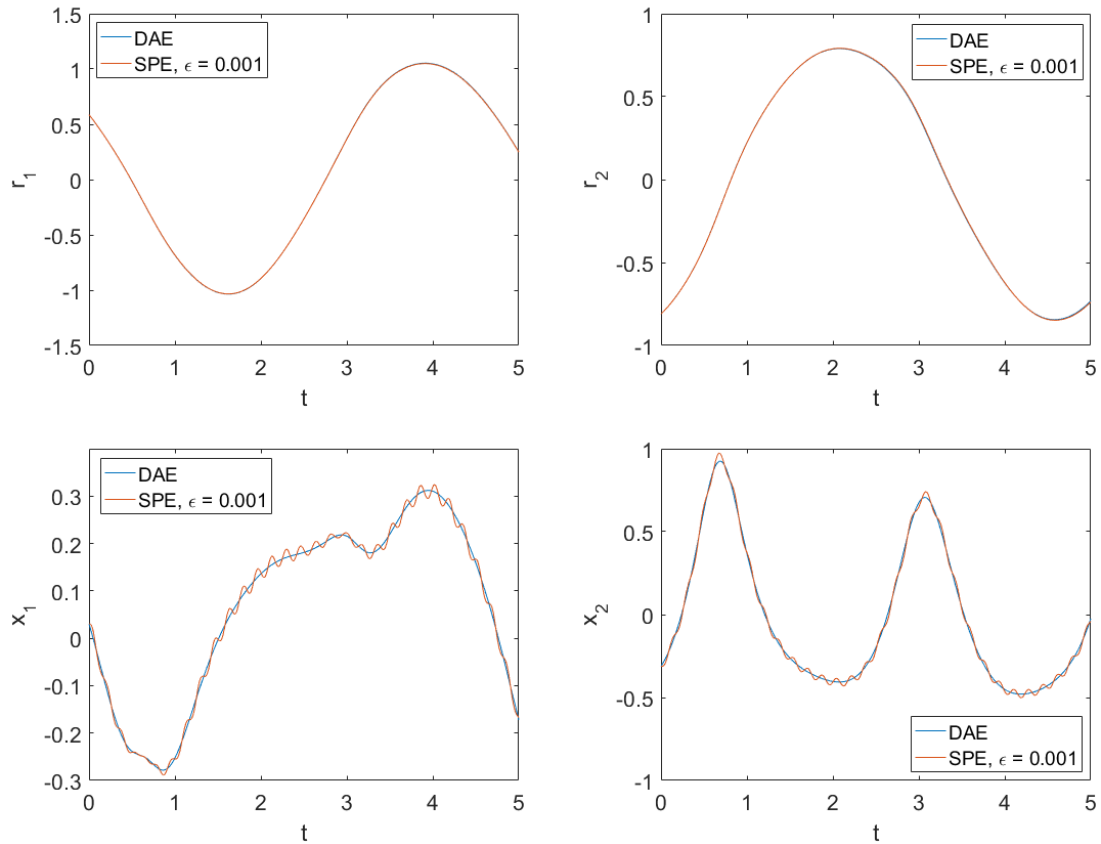

Figure: Example ($d = 2$) of DAE (massless) and SPE (massive, $\varepsilon = 0.001$) dynamics with compatible initial condition.

Related work

- To our knowledge, there are no convergence results for such extended Lagrangian schemes, except in setting (linear-response regime) where dynamics are linear in both r and x and the system can be directly diagonalized [L. Lin, J. Lu, S. Shao 2014].
- An interesting related work [D. An, T. Head-Gordon, L. Lin, J. Lu 2019] studies the related stochastic dynamics:

$$\ddot{r}_\varepsilon(t) = G(r_\varepsilon(t), x_\varepsilon(t)))$$
$$\varepsilon\ddot{x}(t) = b(r_\varepsilon(t)) - A(r_\varepsilon(t))x_\varepsilon(t) - \sqrt{\varepsilon}\,\gamma\,\dot{x}_\varepsilon + \sqrt{2\gamma/\beta}\,\varepsilon^{1/4}\,\dot{W}(t),$$

  where β is an inverse temperature, γ is a damping parameter, and W is a Brownian motion.
- Choosing $\beta^{-1} = O(\varepsilon^{1/2})$ yields $O(\sqrt{\varepsilon})$ error for both $r_\varepsilon, x_\varepsilon$.
- These results are sharp in this setting, but the setting itself (even without noise) loses the $O(\varepsilon)$ error.

Residual dynamics

- Recall the dynamics

$$\ddot{r}_\varepsilon(t) = G(r_\varepsilon(t), x_\varepsilon(t)))$$
$$\varepsilon\ddot{x}_\varepsilon(t) = b(r_\varepsilon(t)) - A(r_\varepsilon(t))x_\varepsilon(t).$$

- Think of path r_ε as being fixed, then study the behavior of x_ε.
  - Governed by linear inhomogeneous ODE with time-dependent coefficients.
- Want to think of x_ε as perturbation of $A(r_\varepsilon(t))^{-1}b(r_\varepsilon(t))$, so consider instead dynamics for the residual $y_\varepsilon := x_\varepsilon - A(r_\varepsilon(t))^{-1}b(r_\varepsilon(t))$:

$$\varepsilon\ddot{y}_\varepsilon = -A(r_\varepsilon(t))^{-1}y_\varepsilon(t) + \varepsilon\psi_\varepsilon(t),$$

  where

$$\psi_\varepsilon := -\frac{d^2}{dt^2}\left[A(r_\varepsilon)^{-1}b(r_\varepsilon)\right].$$

- *A priori* bounds guarantee that $|\psi_\varepsilon| \leq C$ uniformly in ε, t.
- Initial conditions for y_ε and $\dot{y}_\varepsilon$ are given by

$$y_\varepsilon(0) = 0,\ \dot{y}_\varepsilon(0) = z_0 := \dot{x}_\varepsilon(0) - \dot{x}_\star(0).$$

Warm-up

- As a warm-up, consider the constant-coefficient, homogeneous version of the residual dynamics:

$$\begin{cases} \varepsilon \ddot{y}_\varepsilon(t) = -A y_\varepsilon(t) \\ y_\varepsilon(0) = 0, \ \dot{y}_\varepsilon(0) = z_0, \end{cases}$$

  where A is constant.
- Then we have the explicit solution

$$y_\varepsilon(t) = \sqrt{\varepsilon} \sin\left(\sqrt{A}\frac{t}{\sqrt{\varepsilon}}\right) A^{-1/2} z_0.$$

- $|y_\varepsilon| \leq C\sqrt{\varepsilon}$ for arbitrary z_0, and moreover $\dot{y}_\varepsilon$ is $O(1)$ but oscillatory on time scale $\sim \sqrt{\varepsilon}$.
- If $z_0 = 0$, then in fact we have $y_\varepsilon \equiv 0$, which is of course much stronger than real case (will get $|y_\varepsilon| \leq C\varepsilon$).

Homogeneous system

- First study homogeneous equation

$$\varepsilon \ddot{\widetilde{y}}_\varepsilon = -A(r_\varepsilon)\widetilde{y}_\varepsilon, \tag{1a}$$

$$\widetilde{y}_\varepsilon(s) = 0,\ \dot{\widetilde{y}}_\varepsilon(s) = \xi_0, \tag{1b}$$

  where the starting time $s < T$ and initial value ξ_0 is arbitrary.

- For future reference, we define the flow map for the homogeneous system (1) by

$$\Phi_\varepsilon^{s,t}(0, \xi_0) = \begin{pmatrix} \widetilde{y}_\varepsilon(t) \\ \dot{\widetilde{y}}_\varepsilon(t) \end{pmatrix}$$

  for $t \geq s$, where $\widetilde{y}_\varepsilon$ is the solution of (1).

Homogeneous system

- Define
$$K_\varepsilon(t) = A(r_\varepsilon(t))^{1/2}.$$
- Let
$$U^s_{\varepsilon,+}(t) = \mathcal{T} e^{\imath\varepsilon^{-1/2}\int_s^t K_\varepsilon(t')\,dt'},$$
where $\mathcal{T}e^{[\cdot]}$ denotes is 'the time-ordered exponential.'
- This just means that $U^s_{\varepsilon,+}(t)$ is defined to be the solution of
$$\dot{U}^s_{\varepsilon,+}(t) = \imath\varepsilon^{-1/2} K_\varepsilon(t) U^s_{\varepsilon,+}(t), \quad U^s_{\varepsilon,+}(s) = I_d.$$
- Likewise, let
$$U^s_{\varepsilon,-}(t) = \mathcal{T} e^{-\imath\varepsilon^{-1/2}\int_s^t K_\varepsilon(t')\,dt'}.$$
- By construction $U^s_{\varepsilon,+}$ and $U^s_{\varepsilon,-}$ are unitary matrices for all t.

Homogeneous system

Lemma

(i) $\Phi_\varepsilon^{s,t}(0,\xi_0)$ *can be written in the form*

$$\Phi_\varepsilon^{s,t}(0,\xi_0) = \begin{pmatrix} U_{\varepsilon,+}^s(t)c_{\varepsilon,+}^s(t) + U_{\varepsilon,-}^s(t)c_{\varepsilon,-}^s(t) \\ \imath\varepsilon^{-1/2}K_\varepsilon(t)\left[U_{\varepsilon,+}^s(t)c_{\varepsilon,+}^s(t) - U_{\varepsilon,-}^s(t)c_{\varepsilon,-}^s(t)\right] \end{pmatrix}.$$

(ii) $c_{\varepsilon,+}^s(t)$ *and* $c_{\varepsilon,-}^s(t)$ *allow the estimate*

$$|c_{\varepsilon,+}^s(t)|, |c_{\varepsilon,-}^s(t)| \leq C\varepsilon^{1/2}|\xi_0|,$$

where C *is independent of* ε *and* ξ_0.
(iii) Φ *allows the estimate*

$$\Phi_\varepsilon^{s,t}(0,\xi_0) = \begin{pmatrix} \mathcal{O}(\varepsilon^{1/2}|\xi_0|) \\ \mathcal{O}(|\xi_0|) \end{pmatrix}.$$

Inhomogeneous system

- Now we return to the residual system

$$\ddot{y}_\varepsilon = -A(r_\varepsilon(t))^{-1} y_\varepsilon(t) + \varepsilon\psi_\varepsilon(t), \quad y_\varepsilon(0) = 0,\ \dot{y}_\varepsilon(0) = z_0.$$

- Introduce the variable $z_\varepsilon := \dot{y}_\varepsilon$ to obtain:

$$\begin{pmatrix} \dot{y}_\varepsilon \\ \dot{z}_\varepsilon \end{pmatrix} = \begin{pmatrix} z_\varepsilon \\ -\varepsilon^{-1} A y_\varepsilon \end{pmatrix} + \begin{pmatrix} 0 \\ \psi_\varepsilon(t) \end{pmatrix}.$$

- Then by Duhamel's principle,

$$\begin{pmatrix} y_\varepsilon(t) \\ z_\varepsilon(t) \end{pmatrix} = \Phi_\varepsilon^{0,t}(0, z_0) + \int_0^t \Phi_\varepsilon^{s,t}(0, \psi_\varepsilon(s))\, ds.$$

- Recall from our Lemma:

$$\Phi_\varepsilon^{s,t}(0, \xi_0) = \begin{pmatrix} \mathcal{O}(\varepsilon^{1/2}|\xi_0|) \\ \mathcal{O}(|\xi_0|) \end{pmatrix}.$$

- Then it directly follows that $|y_\varepsilon| \leq C\varepsilon^{1/2}$, $|\dot{y}_\varepsilon| \leq C$.

Completing the proof of the coarse estimate

- Recall dynamics:
$$\dot{r}_\varepsilon = p_\varepsilon, \quad \dot{p}_\varepsilon = G(r_\varepsilon, x_\varepsilon).$$
- We just proved $y_\varepsilon = x_\varepsilon - A(r_\varepsilon)^{-1} b(r_\varepsilon) = \mathcal{O}(\varepsilon^{1/2})$, so by assumptions / *a priori* bounds
$$\dot{r}_\varepsilon = p_\varepsilon, \quad \dot{p}_\varepsilon = G(r_\varepsilon, A(r_\varepsilon)^{-1} b(r_\varepsilon)) + e_\varepsilon,$$
where $e_\varepsilon = \mathcal{O}(\varepsilon^{1/2})$.
- Hence $(r_\varepsilon, p_\varepsilon)$ almost satisfy an $\mathcal{O}(\varepsilon^{1/2})$-perturbation of the *equations* defining $(r_\star, p_\star)$.
- This is enough to imply that $r_\varepsilon = r_\star + \mathcal{O}(\varepsilon^{1/2})$, $p_\varepsilon = p_\star + \mathcal{O}(\varepsilon^{1/2})$.
  - Several ways to see this, e.g., go back to Picard iteration.

Sharp estimates in 1D

- We focus on the case $d = 1$.
- Actually we will have to bootstrap from our coarse estimate!
- Retain definitions from above.
- Since $d = 1$, we denote $k_\varepsilon = K_\varepsilon$ to emphasize that this is a scalar quantity.
- Since $d = 1$, time-ordered exponential takes simple form

$$U^s_{\varepsilon,\pm}(t) = e^{\pm \imath(\kappa_\varepsilon(t)-\kappa_\varepsilon(s))/\sqrt{\varepsilon}}, \quad \kappa_\varepsilon(t) = \int_0^t k_\varepsilon(s)\, ds.$$

- By assumption $k \geq C^{-1}$, so κ_ε is strictly increasing and inverse mapping κ_ε^{-1} is well-defined.
- *A priori* bounds imply $|\dot{\kappa}|, |\ddot{\kappa}| \leq C$.

Sharp homogeneous oscillator asymptotics

Lemma

With the flow map $\Phi^{t,s}$ defined as before,

$$\Phi^{s,t}(0,\xi_0) = \begin{pmatrix} \varepsilon^{1/2} k_\varepsilon(t)^{-1/2} k_\varepsilon(s)^{-1/2} \sin\left(\frac{\kappa_\varepsilon(t)-\kappa_\varepsilon(s)}{\sqrt{\varepsilon}}\right)\xi_0 \\ k_\varepsilon(t)^{1/2} k_\varepsilon(s)^{-1/2} \cos\left(\frac{\kappa_\varepsilon(t)-\kappa_\varepsilon(s)}{\sqrt{\varepsilon}}\right)\xi_0 \end{pmatrix} + \begin{pmatrix} \mathcal{O}(\varepsilon) \\ \mathcal{O}(\varepsilon^{1/2}) \end{pmatrix}$$

- Compare: proof of asymptotics of Hermite polynomials from Tao's book on RMT.
- In earlier proof, obtained the system for $c_\pm$ (omit ε from notation):

$$\dot{c}_+ = -\frac{1}{2}U_+^{-1}K^{-1}\dot{K}U_+c_+ + \frac{1}{2}U_+^{-1}K^{-1}\dot{K}U_-c_-$$
$$\dot{c}_- = \frac{1}{2}U_-^{-1}K^{-1}\dot{K}U_+c_+ - \frac{1}{2}U_-^{-1}K^{-1}\dot{K}U_-c_-.$$

- Since $d = 1$, we can now commute operators to obtain

$$\dot{c}_+ = -\frac{\dot{k}}{2k}c_+ + \frac{\dot{k}}{2k}e^{-2\imath\kappa/\sqrt{\varepsilon}}c_-, \quad \dot{c}_- = -\frac{\dot{k}}{2k}c_- + \frac{\dot{k}}{2k}e^{2\imath\kappa/\sqrt{\varepsilon}}c_+.$$

Sharp inhomogeneous asymptotics

Lemma

Let y_ε be the solution to the residual system $\varepsilon \ddot{y}_\varepsilon = -Ay_\varepsilon + \psi_\varepsilon$, with $y_\varepsilon(0) = 0, \dot{y}_\varepsilon(0) = z_0$. Then for $t \in [0, T]$,

$$\begin{pmatrix} y_\varepsilon(t) \\ \dot{y}_\varepsilon(t) \end{pmatrix} = \begin{pmatrix} \varepsilon^{1/2} k(t)^{-1/2} k(0)^{-1/2} \sin\left(\frac{\kappa(t)}{\sqrt{\varepsilon}}\right) z_0 \\ k(t)^{1/2} k(0)^{-1/2} \cos\left(\frac{\kappa(t)}{\sqrt{\varepsilon}}\right) z_0 \end{pmatrix} + \begin{pmatrix} O(\varepsilon) \\ O(\varepsilon^{1/2}) \end{pmatrix}.$$

- Reformulate inhomogeneous as the first-order system

$$\begin{pmatrix} \dot{y}_\varepsilon(t) \\ \dot{z}_\varepsilon(t) \end{pmatrix} = \begin{pmatrix} z_\varepsilon(t) \\ -\varepsilon^{-1} A(r_\varepsilon(t)) y_\varepsilon(t) \end{pmatrix} + \begin{pmatrix} 0 \\ \psi_\varepsilon(t) \end{pmatrix}.$$

- Duhamel's principle yields

$$\begin{pmatrix} y_\varepsilon(t) \\ z_\varepsilon(t) \end{pmatrix} = \Phi_\varepsilon^{0,t}(0, z_0) + \int_0^t \Phi_\varepsilon^{s,t}(0, \psi_\varepsilon(s))\, ds.$$

- Want to show integral term is $\mathcal{O}(\varepsilon)$ (oscillatory integral argument).

Completion of proof of sharp estimate

- Recall the ε-massive dynamics

$$\ddot{r}_\varepsilon = G(r_\varepsilon, x_\varepsilon)$$
$$\varepsilon \ddot{x}_\varepsilon = b(r_\varepsilon) - A(r_\varepsilon) x_\varepsilon$$

  and the massless dynamics

$$\ddot{r}_\star = G\left(r_\star, A(r_\star)^{-1} b(r_\star)\right).$$

- Already know that $|x_\varepsilon - A(r_\varepsilon)^{-1} b(r_\varepsilon)| \leq C\varepsilon^{1/2}$.
- Also know that $|r_\varepsilon - r_\star| \leq C\varepsilon^{1/2}$ as well by bootstrapping from the coarse estimate.
- Combining previous two lines, get $|x_\varepsilon - A(r_\star)^{-1} b(r_\star)| \leq C\varepsilon^{1/2}$.
- Hence can Taylor-expand the ODE and retain only linear terms, with $\mathcal{O}(\varepsilon)$ error
- Defining new variable $\theta_\varepsilon := r_\varepsilon - r_\star$, which measures the error in the r variable, can obtain:

$$\ddot{\theta}_\varepsilon = \Upsilon \theta_\varepsilon + \Gamma y_\varepsilon + O(\varepsilon), \quad \theta_\varepsilon(0) = 0,\ \dot{\theta}_\varepsilon(0) = 0.$$

Completion of proof of sharp estimate

- We view the ODE for θ_ε as a forced modification of the homogeneous ODE
$$\ddot{\widetilde{\theta}}_\varepsilon = \Upsilon \widetilde{\theta}_\varepsilon.$$
- Let $\Theta^s(t) \in \mathbb{R}^{2\times 2}$ be the flow map for this homogeneous ODE.
- Then by Duhamel's principle and oscillatory integral argument (forcing is of magnitude $\sim \sqrt{\varepsilon}$ and oscillation $\sim \sqrt{\varepsilon}$ show forcing contributes only $\mathcal{O}(\varepsilon)$.
- In optimally compatible case, forcing is of magnitude $\mathcal{O}(\varepsilon)$ to begin with.

Conclusions and further directions

- We have proved first convergence results for iEL/0-SCF method.
- Can get sharp asymptotics in case $d = 1$. Qualitatively, the analysis seems to capture what is happening for $d \geq 1$.
- Nonetheless, how to extend the analysis?
  - There should probably be an extra $\mathcal{O}(\varepsilon)$ non-oscillatory drift term appearing in the homogeneous oscillator dynamics, depending only on commutators that are equal to zero in $d = 1$.
- Can one extend to more general $Q(r, x)$? Schemes for *ab initio* MD?
- Any practical insights to be gained from the analysis?