Parallel in time molecular dynamics, noncolinear magnetism and orbital free methods

A pot pourri

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Parallel in time molecular dynamics

- Many progress in the field of multiscale simulations in the space domain
- Less is known in the way to perform similar approaches in the time domain
- It is indeed possible to parallelize molecular dynamics simulations
- The technique is mostly useful for *ab initio* simulations.

General framework

- Molecular dynamics: integrate the equations of motion for atoms to follow the evolution of their coordinates $(\vec{R}_1(t), ..., \vec{R}_N(t))$ in time.

$$\begin{aligned} \vec{R}_i(t=0) &= \vec{R}_i^0 \\ \frac{d^2\vec{R}_i(t)}{dt^2} &= -\nabla_{\vec{R}_i} V(\vec{R}_1(t), ..., \vec{R}_N(t)) \end{aligned}$$

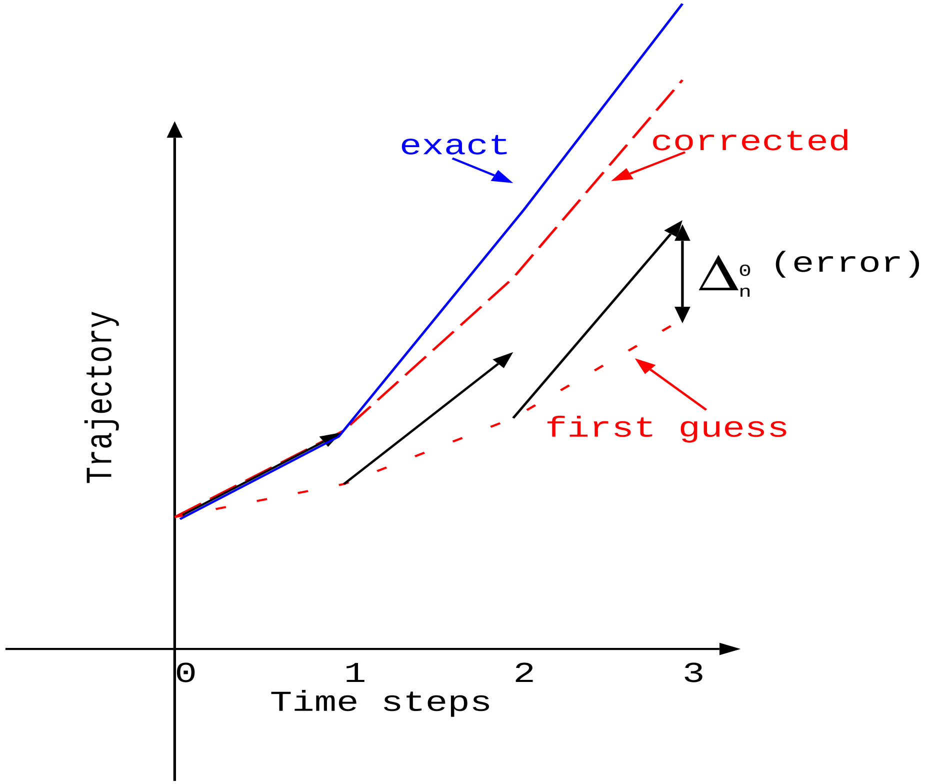
- $V(\vec{R}_1(t), ..., \vec{R}_N(t))$ is the potential function, its gradients, the “force field”
- We solve an initial value problem: How to use parallel machines on it?

Consider now an initial value problem

- Rewrite the equation of motion symbolically as $\frac{du}{dt} = \Lambda(u)$, discretize it using a timestep ΔT.
- If u_n is our position in phase space after n steps, we introduce the propagator: $F_{\Delta T}(u_n) = u_{n+1}$
- $F_{\Delta T}$ describes merely the way we go from one configuration to the next.
- One could introduce, for any set v_n, $J = \sum_i (F_{\Delta T}(v_n) - v_{n+1})^2$, and try to minimize it in parallel.
- Here, we follow another route (parareel algorithm) more akin to a feedback mechanism.

The parareal algorithm (in graphics)

- Use two force fields as a predictor-corrector



The first step is exact

The parareal algorithm (in equations)

- Use two force fields as a predictor-corrector.
- First field:

$$\begin{aligned} u^0_{n+1} &= G_{\Delta T}(u^0_n) \\ u^0_0 &= u_0 \end{aligned}$$

- $G_{\Delta T}(u^0_n)$ is the coarse propagator.
- $F_{\Delta T}(u^0_n)$ is the fine propagator.
- The error is: $\Delta^0_n = F_{\Delta T}(u^0_n) - G_{\Delta T}(u^0_n)$
- Naturally, $F_{\Delta T}(u^0_n)$ can be computed in parallel

The feedback process(Lions et al 2001)

- Propagate a second time using the coarse propagator *corrected* by Δ_n^0

$$u_{n+1}^1 = G_{\Delta T}(u_n^1) + \Delta_{n+1}^0$$

- The process can be iterated, and this defines our successive trajectories, denoted by u_n^k

$$\begin{aligned} u_{n+1}^{k+1} &= G_{\Delta T}(u_n^{k+1}) + F_{\Delta T}(u_n^k) - G_{\Delta T}(u_n^k) \\ u_0^k &= u_0 \end{aligned}$$

- Expensive $F_{\Delta T}$ in // on N processors
- Efficient if $k_{\mathrm{conv}} \ll N$ and $G_{\Delta T} \ll F_{\Delta T}$

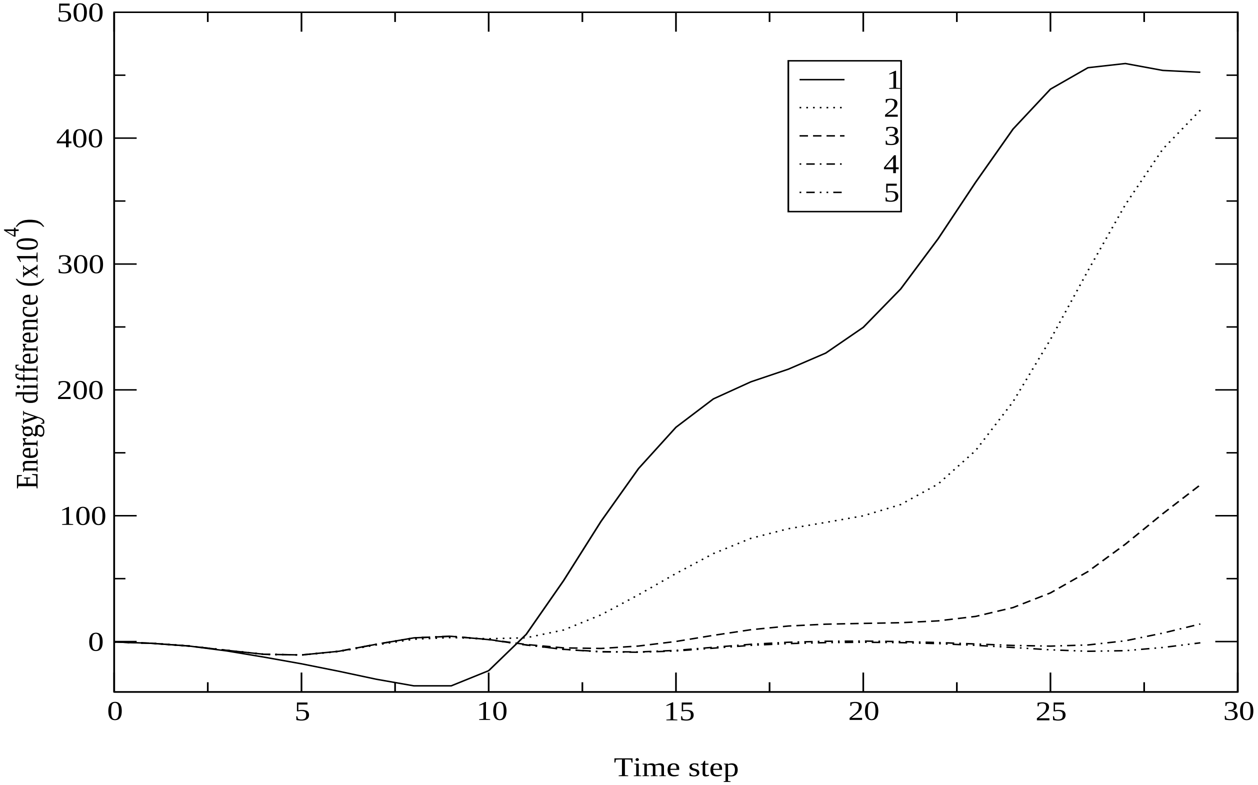
Coupling classical and ab initio

- Simulations of Aluminum (32 atoms)
- Coarse integrator: Glue (==EAM) potential (Ercolessi)

$$\begin{aligned} U &= \frac{1}{2}\Sigma_{i\neq j}\phi(|r_i - r_j|) + \Sigma_j F(\rho_j) \\ \rho_j &= \Sigma_{i\neq j}\rho(|r_i - r_j|) \end{aligned}$$

Results

- Convergence of energy
-



- For $k = 4$, we are converged

Implementation

- A simple *sui generis* classical MD program has been added (light)
- A clone of gstate (pstate) has been written, which calls *ab initio* or classical MD alternatively
- Room for improvement and testing....

(Intermezzo) Non colinear magnetism

- Generally, magnetism is described by $\rho\uparrow$ and $\rho\downarrow$.
- This suppose that magnetisation is along a given direction
- In some cases the magnetisation vector rotates in the cristal, needs $\vec{M}(r)$
- Questions: how to define $\vec{M}(r)$, how to determine it?

$$\Psi(r) = \left\{ \begin{array}{c} \Psi_\alpha(r) \\ \Psi_\beta(r) \end{array} \right\}$$

$\vec{M}_x(r) =< \Psi(r)|\sigma_x|\Psi(r) > \sigma_x$ is the Pauli matrix

Implementation

- Spinor representation of Ψ already ok from spin-orbit (cf M. Torrent)
- Now, $E_{xc}(n(r), \vec{m}(r)) \equiv E_{xc}(n(r), |\vec{m}(r)|)$. Not totally general in GGA.
- Expression of V_{xc} (has now 4 components: normal + magnetic field along $\vec{m}$)

$$V_{xc}(r) \;=\; \frac{\delta E_{xc}}{\delta \rho(r)} \delta_{\alpha,\beta} + \frac{\delta E_{xc}}{\delta |m(r)|} \widehat{m}(r).\sigma$$

- $\rho^{\alpha,\beta} = \sum_n f_n < r|\Psi^\alpha >< \Psi^\beta|r >$ is complex
- Use $\rho^{1,2} + \rho^{2,1}$ which is real (and yields $\rho + m_x$)

Results and discussion

- Implementation not too difficult (but still some work: a good exercice)
- No symmetries taken into account (should do spin groups, brrr..)
- Not many real calculations (a few tests on Fe dimers). None published
- Convergence is slow.
- Seems to be working, but need volunteer for testing (especially in relation with spin orbit: yields magnetic anisotropy)

Orbital-free methods

- Try to perform high temperature calculations
- The Fermi-Dirac occupation imposes many levels.
- Can we avoid to compute the wave functions ?
- Approach 1: use simplified funtionals (e.g. Thomas-Fermi and extensions)
- Approach 2: use recursion methods

Thomas-Fermi

- The free energy functionnal of independent electrons is replaced by a function.
- This is a local density approximation on this functional
- $\frac{\beta F}{V} = \rho\beta\mu - \frac{2}{3} f_{3/2}(f_{1/2}^{-1}(\frac{\rho}{C}))$
- Implementation is very localized: need to change vtorho
- Can perform molecular dynamics. Other functionals in progress (gradient: just use the code!)

Order N method, based on recursion I

- $\rho(\mathbf{r}) =< \mathbf{r}|\frac{1}{1+e^{\beta(\bar{H}-\mu)}}|\mathbf{r} >,\ \mathbf{r}$ Expression of density in terms of density matrix

- $\rho(\mathbf{r}) =< \mathbf{r}|1|\mathbf{r} > - < \mathbf{r}|\frac{1}{1+e^{-\beta(\bar{H}-\mu)}}|\mathbf{r} >= 1- < \mathbf{r}|\frac{1}{1+e^{-\beta(\bar{H}-\mu)}}|\mathbf{r} >$. We change β in $-\beta$

- Second term is diagonal part of inverse: can be computed by recursion.

- Based on: $e^{-\beta(\bar{H}-\mu)}|u_n >= b_n|u_{n-1} > +a_n|u_n > +b_{n+1}|u_{n+1} >$

- Need an easy way to compute $e^{-\beta(\bar{H}-\mu)}|u_n >$

Order N method, based on recursion II

- Decomposition in partial fraction

$$\frac{1}{1+x} = \frac{1}{2p}\sum_{j=1}^{2p-1}\frac{1}{1+z_j x^{\frac{1}{2p}}}\widehat{m}(r).\sigma$$

$$\frac{1}{1+e^{-\beta(\bar{H}-\mu)}} = \frac{1}{2p}\sum_{j=1}^{2p-1}\frac{1}{1+z_j e^{-\frac{\beta(\bar{H}-\mu)}{2p}}}.$$

- Now, use Trotter formula when P large enough $e^{-\beta(V(\mathbf{r})-\triangle/2-\mu)/2p} \sim e^{\beta\mu/2p}e^{-\beta V(\mathbf{r})/4p}.e^{\beta\triangle/4p}.e^{-\beta V(\mathbf{r})/4p}$

Implementation

- We really need to change vtorho. Just branch here, leave the code intact (cool!)
- Work on a grid, apply $\exp(V(r))$ in real space (local pot), $\exp(\Delta)$ in reciprocal space

$\mathbf{n_0}$	β	$\mathbf{p}$
≤ 10	0.01	500
≤ 10	0.05	100
≤ 10	0.5	20

Table 1: Necessary iterations to converge using a DSE with different values of p

Conclusion

- Really an extension of abinit, can leave in a separate directory
- Might want, some day to think on real space methods....
- Still a lot of work to do (e.g. forces)
- Approximate orbital free methods can be used as predictor in parareal approach.
- Useful for alloys, or when one does not have classical potentials available.
- ......

THANK YOU!