Quantum optimization through path-integral molecular dynamics

Alfredo Fiorentino[1], Nicola Marzari[1,2]

[1] PSI Center for Scientific Computing, Theory, and Data, Villigen, Switzerland
[2] Theory and Simulation of Materials (THEOS), Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

PSI Center for Scientific Computing, Theory and Data

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1. Introduction

- **Motivation**
  Global optimization of atomic degrees of freedom is a key problem in physics whenever the number of candidate structures (local minima) is combinatorially large: cluster physics, polymer folding, interfaces...
- **The idea**
  Novel implementation of **Quantum Annelealing** (QA) using **Path-Integral Molecular Dynamics** (PIMD), labeled QA-PIMD. While **Classical Annealing** (CA) involves melting and slowly cooling through a temperature schedule, QA uses quantum delocalization.

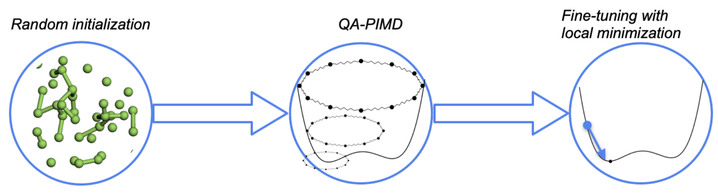


Figure: Summary of the workflow

2. Methods

QA-PIMD is obtained by combining two powerful methods:

- **Quantum annealing: exploration through quantum delocalization**

$$\hat{H}(t) = -\alpha(t) \sum_{i=1}^{N} \frac{\hbar^2}{2m_i}\nabla^2 + U(\hat{\mathbf{x}}_1, \ldots, \hat{\mathbf{x}}_N)$$

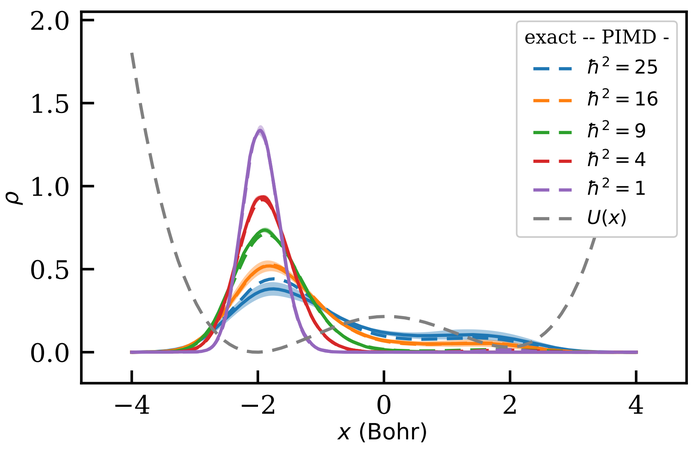
  - $\alpha(t=0) \gg 1$, $\alpha(t_max) \ll 1$
  - $\lim_{t_{max}\to\infty} Tr[\rho(t_{max})U] = \min U$
- **Path-Integral Molecular Dynamics**
  Equilibrium nuclear quantum effects (NQEs) from P classical simulations ($P \sim 10-100$.

$$H_P = \sum_{i=1}^{N}\sum_{j=1}^{P}\left[\frac{1}{2m_i}|\mathbf{p}_i^{(j)}|^2 + \frac{1}{2}m_i\omega_P^2|\mathbf{x}_i^{(j)} - \mathbf{x}_i^{(j+1)}|^2\right] + \sum_{j=1}^{P} U(\mathbf{x}_{1j}, \ldots, \mathbf{x}_{Nj}),$$

$$\omega_P = \frac{Pk_BT}{\hbar}$$

3. Benchmark: asymmetric double well

The local density from 200-ps PIMD simulations follows adiabatically the exact instantaneous quantum density. $T = 100$ K.



4.1 Lennard-Jones clusters

LJ clusters: millions of estimated local minima with just a few tens of atoms!

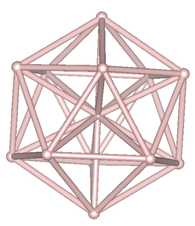


Figure: Global minimum of a LJ cluster with $N = 13$.

4.2 LJ clusters: classical vs quantum

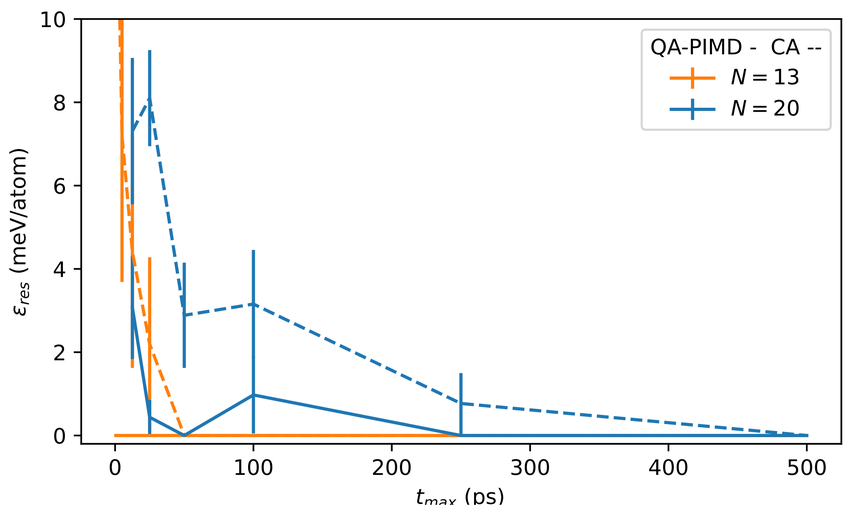


Figure: Comparison of QA and CA residual energy against annealing time, for two size of LJ cluster.

5.1 Missing hydrogens' sites

Hydrogen's positions are hardly detected in X-ray scattering, resulting in a large dataset of structures missing hydrogen positions.
The 10 materials are simulated with MACE's foundational **MLIP**

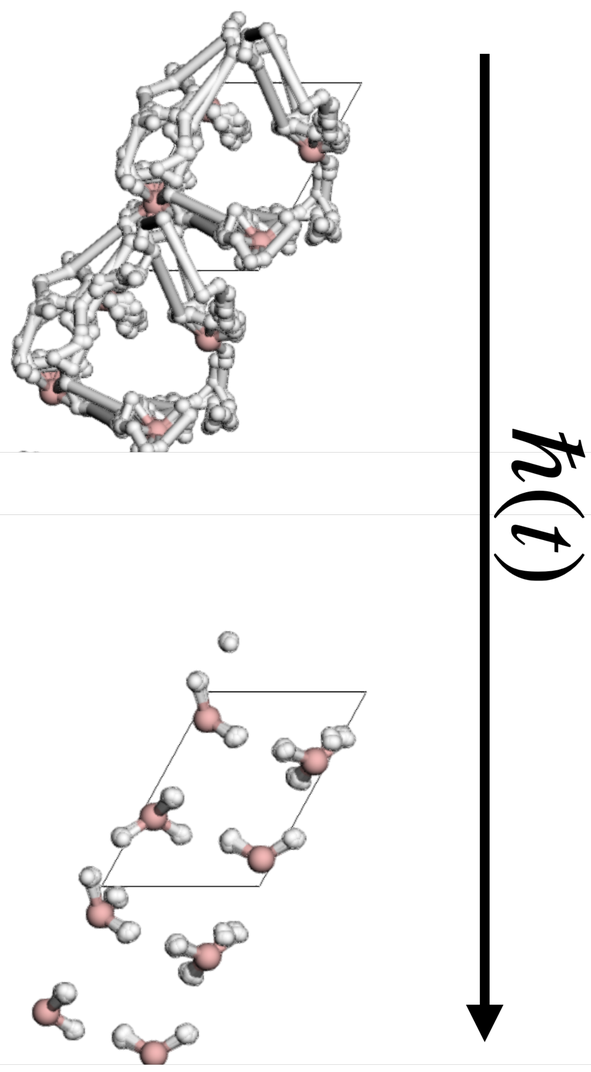


Figure: Example of finding, through QA-PIMD, the hydrogens' sites for a known crystal (B_4H_{12}). Check the QR code for the video!

5.2 Results *missing hydrogens* problem

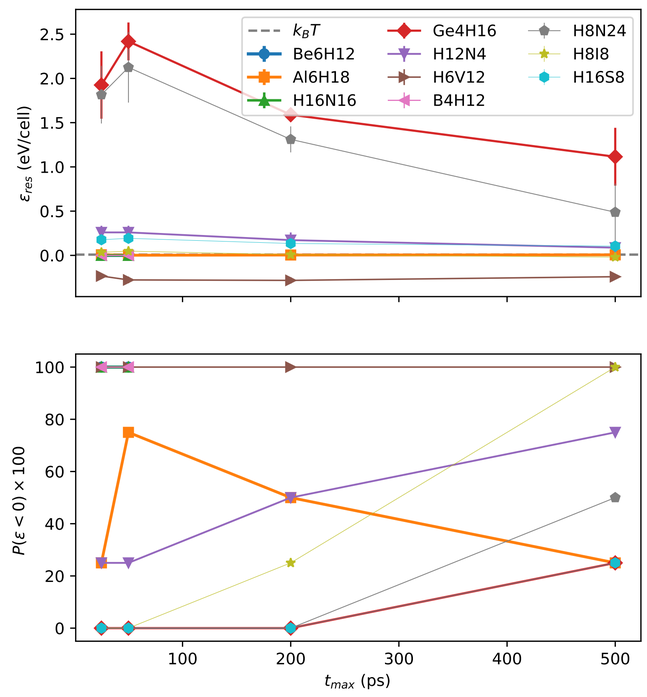


Figure: Top panel: residual energy with respect of MC3D database against annealing time. Lower panel: probability of finding an equal of lower energy.

6. Conclusion

- QA-PIMD is an unbiased and general novel global optimization algorithm using quantum fluctuations.
- It can outperform classical annealing. E.g., shorter annealing time needed for LJ clusters.
- Material discovery: new candidate stable crystals from the MC3D database

Open questions and challenges

- Small, but **finite**, temperature. Not a ground-state method.
- PIMD time-evolution is not exactly quantum, dynamically $\mathrm{QA} - \mathrm{PIMD} \neq \mathrm{QA}$. Work in progress.

7. References

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