Time-dependent Reverse Monte Carlo Modelling

A summary of O Gereben et al (2007) *J. Phys.: Condens. Matter 19 335223*

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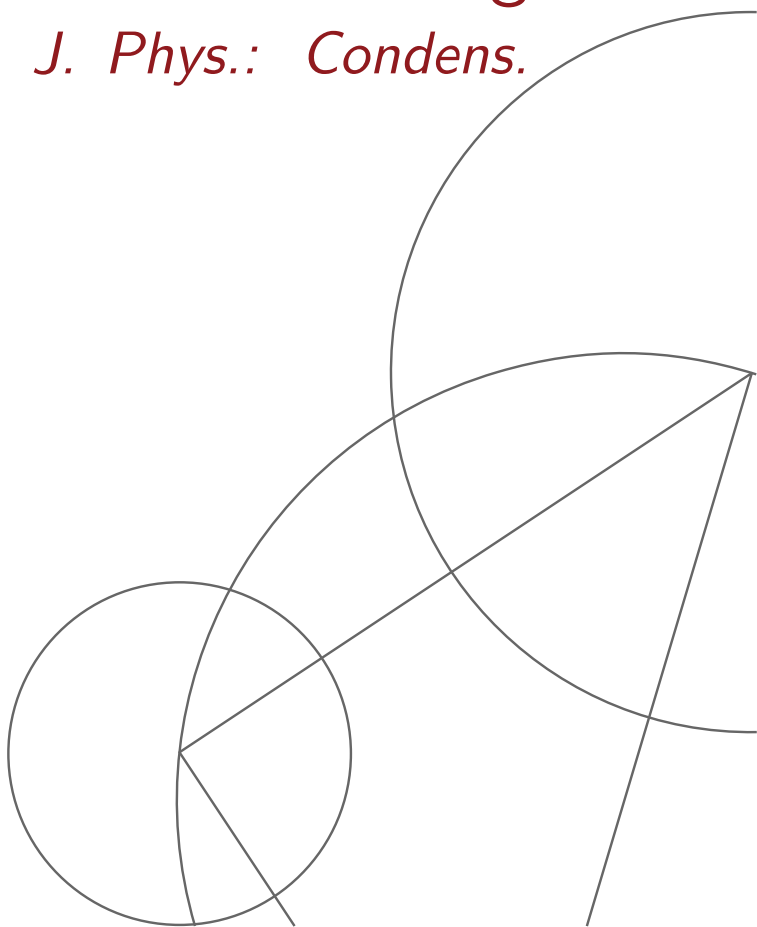

Table of Contents

Basics of RMC

- Statistical method for modelling of atomic-scale structure
- Originally for liquids ([2]), but also applicable to crystalline ([3]), amorphous ([4]) and magnetic ([5]) systems
- Fits atomic structure to experimental data
- Inherently non-parametric

How does it work?

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4. Use MH to accept/reject the proposal given χ^2
5. Repeat step 2-4 until convergence

Overview of RMCt

- Evolution of RMC to model atomic-scale dynamics
- Many ensembles divided by time Δt
- Ensembles have knowledge
- Follows the same steps as RMC

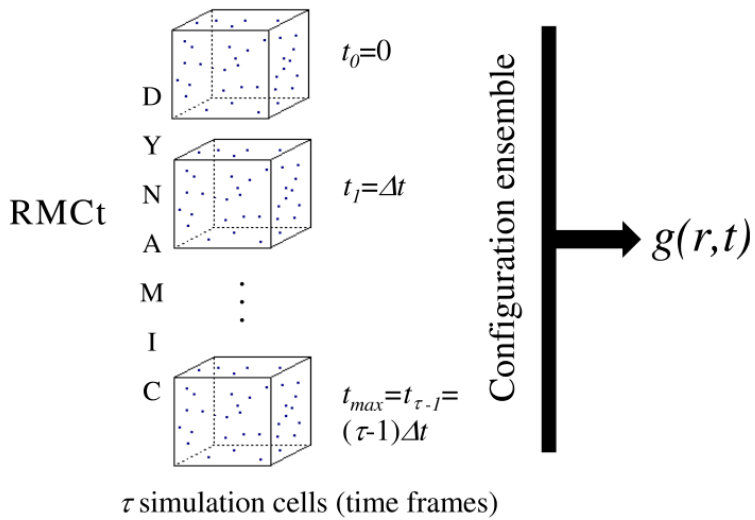


Figure: Sketch of the RMCt method

A statistical issue

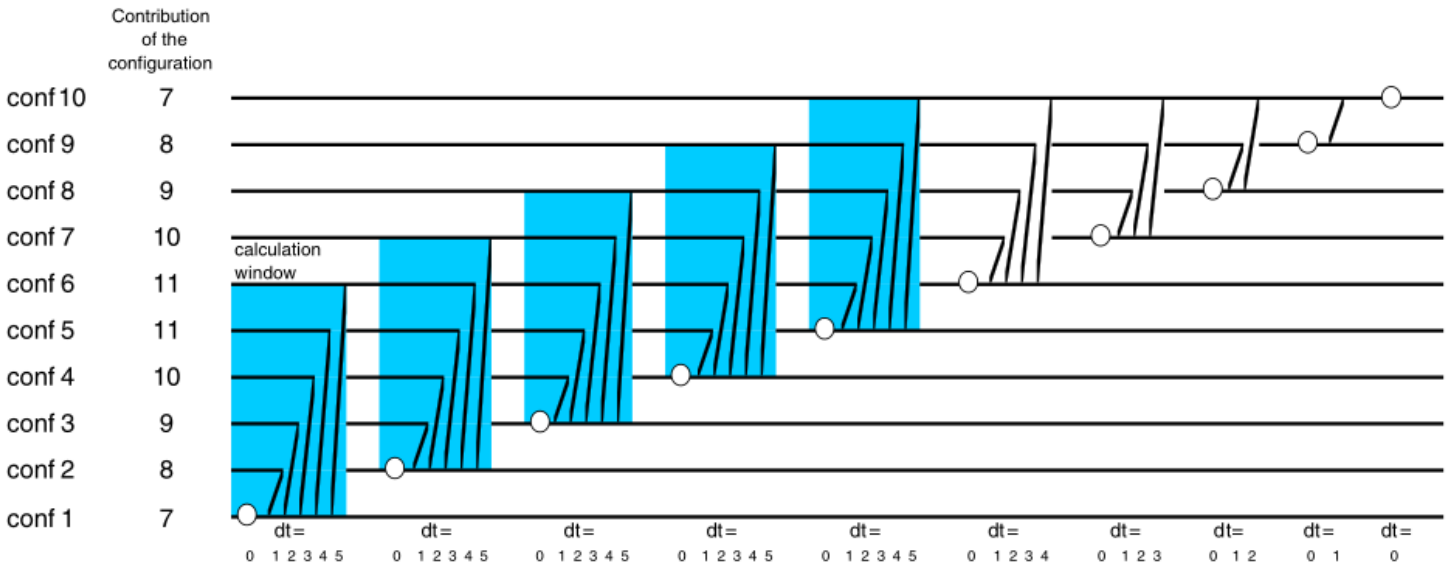


Figure: Sketch of window-of-calculation concept [1]

[1] O Gereben et al, 2007, *J. Phys.: Condens. Matter 19 335223*

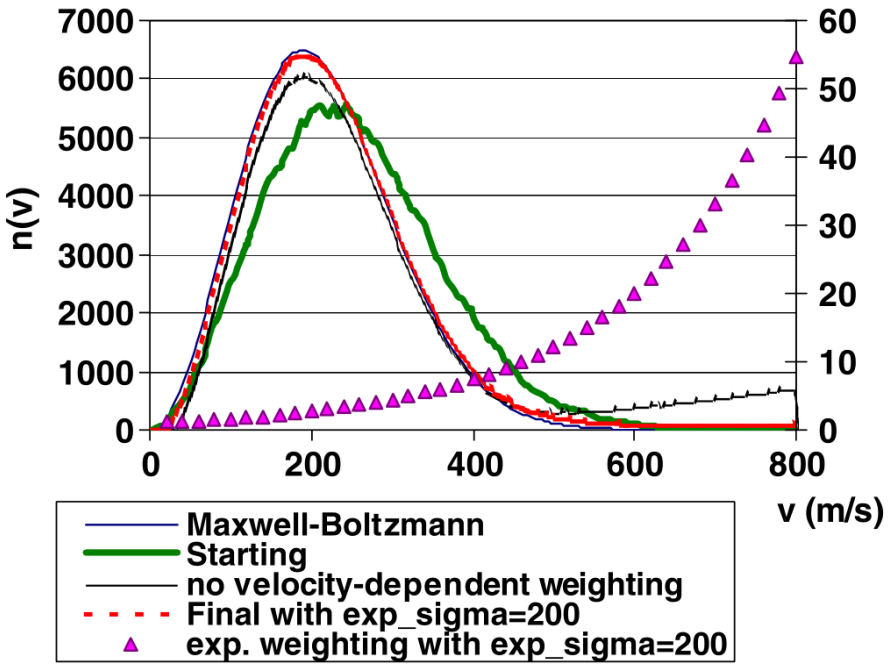
RMCt in the article

- The article[2] describes method - no new physics
- Test data is quasi-experimental, obtained from Molecular Dynamics simulations of liquid Ar
- Dynamics are fitted to three data sets ($g(r,t)$, $S(Q,w)$ and $S(Q)$)
- Constraints are used to facilitate convergence

[2] O Gereben et al, 2007, *J. Phys.: Condens. Matter 19 335223*

Constraints in RMCt

- Necessary for convergence and to produce "real" physics"
- Issues come from lack of information in experimental data
- Adds parameter from *a priori* knowledge
- Can also be physical constraints

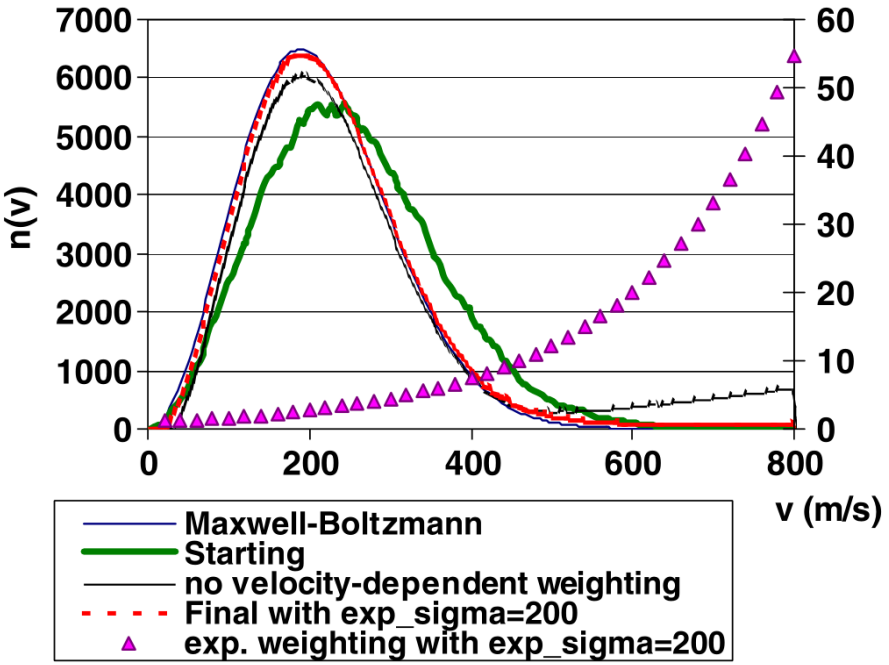


Constraints in practice

Constraints are added by biasing the χ^2. An example:

$$\chi^2_{VD} = \frac{1}{\sigma^2_{VD}} \sum_j (V_j^C - V_j^{MB})^2 w_j(v)$$

$$w_j(v) = \begin{cases} \exp\left(\frac{v_j}{\sigma^{exp}}\right) & \text{if } \sigma^{exp} > 0 \\ 1 & \text{if } \sigma^{exp} \leq 0 \end{cases}$$



Issues

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- Lack of uniqueness can be a problem [4]
- Parameters have to be used anyway (constraints)
- Computationally demanding (huge parameter space, statistical concessions)

Wrap-up

- Non-parametric method for modelling atomic-scale dynamics
- Author succeeded in fitting a model to quasi-experimental data
- Has some issues
  - Lack of uniqueness can be a problem ([4])
  - Constraints require parameters
  - Computationally demanding (statistical concessions, many parameters)
- Despite issues, widely used to great results (especially RMC ([5]))

Thank you for your time!

References

- O Gereben et al, 2007, *J. Phys.: Condens. Matter 19 335223*
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