Quantum Monte Carlo

- Premise: need to use simulation techniques to "solve" many-body quantum problems just as you need them classically.
- Both the wavefunction and expectation values are determined by the simulations. Correlation built in from the start.
- QMC gives most accurate method for general quantum many-body systems, and a standard for approximate DFT calculations.
- Provides a new understanding of quantum phenomena

QMC methods in the continuum

- Variational Monte Carlo (VMC) (single state)
- Projector Monte Carlo methods for T=0:
  - **Diffusion Monte Carlo (DMC)**
  - Reptation MC (RQMC)
  - Auxiliary field QMC (AFQMC)
- Path Integral Monte Carlo for T>0 (PIMC)
- Coupled Electron-Ion Monte Carlo T>0 (CEIMC)

Notation

- Individual coordinate of a particle = r_i
- All 3N coordinates R= (r_1,r_2, …. r_N)

- Total potential energy = V(R)

- Kinetic energy $\hat{T} = -\lambda \sum_{i=1}^{N} \nabla_i^2$ where $\lambda \equiv \frac{\hbar^2}{2m}$

- Hamiltonian $\hat{H} = \hat{T} + \hat{V}$

Variational Monte Carlo (VMC)

- Variational Principle. Given an appropriate trial function:
  - Continuous
  - Proper symmetry
  - Normalizable
  - Finite variance

$$E_V = \frac{\int dR \left\langle \psi \middle| \hat{H} \middle| \psi \right\rangle}{\int dR \left\langle \psi\psi \right\rangle} \geq E_0$$

$$\sigma^2 = \frac{\int dR \left\langle \psi \middle| \hat{H}^2 \middle| \psi \right\rangle}{\int dR \left\langle \psi\psi \right\rangle} - E_V^2$$

- Quantum chemistry uses a product of single particle functions
- With MC we can use any "computable" function.
  - Sample R from $|\psi|^2$ using MCMC.
  - Take average of local energy:
  - Optimize ψ to get the best upper bound

$$E_L(R) = \Re\left[\psi^{-1}(R)\hat{H}\psi(R)\right]$$

$$E_V = \left\langle E_L(R) \right\rangle_{\psi^2} \geq E_0$$

- Better wavefunction, lower variance! "Zero variance" principle. (non-classical)

Problems with Variational MC

- Powerful method since you can use any trial function
- Scaling (computational effort vs. size) is almost classical
- Learn directly about what works in wavefunctions
- No sign problem

- Optimization is time consuming
- Energy is insensitive to order parameter
- Non-energetic properties are less accurate. O(1) vs. O(2) for energy.
- Difficult to find out how accurate results are.
- Favors simple states over more complicated states, e.g.
  - Solid over liquid
  - Polarized over unpolarized

What goes into the trial wave function comes out! "GIGO"

We need a more automatic method! Projector Monte Carlo

Projector Monte Carlo

•Originally suggested by Fermi and implemented in 1950 by Donsker and Kac for H atom.

•Practical methods and application developed by Kalos:

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Helium at zero temperature with hard-sphere and other forces

M. H. Kalos*
Courant Institute of Mathematical Sciences, New York University, New York, New York 10012

D. Levesque and L. Verlet
Laboratoire de Physique Théorique et Hautes Energies, Orsay, France †
(Received 22 August 1973)

Various theoretical and numerical problems relating to heliumlike systems in their ground states are treated. New developments in the numerical solution of the Schrödinger equation permit the solution of 256-body systems with hard-sphere forces. Using periodic boundary conditions, fluid and crystal states can be described; results for the energy and radial-distribution functions are given. A new method of correcting for low-lying phonon excitations

Projector Monte Carlo

(variants: Green's function MC, Diffusion MC, Reptation MC)

- Project single state using the Hamiltonian

$$\phi(t) = e^{-(\mathrm{H}-\mathrm{E_T})\mathrm{t}}\phi(0)$$

- We show that this is a diffusion + branching operator if we can interpret as a probability. But is it?
- **Yes!** for bosons since ground state can be made real and non-negative.
- **But** all excited states must have sign changes. This is the "sign problem."
- For efficiency we do "importance sampling."
- Avoid sign problem with the fixed-node method.

Diffusion Monte Carlo

- How do we analyze this operator?
- Expand into exact eigenstates of H.
- Then the evolution is simple in this basis.
- Long time limit is lowest energy state that overlaps with the initial state, usually the ground state.
- How to carry out on the computer?

$$\psi(R,t) = e^{-(\hat{H}-E_T)t}\psi(R,0)$$

$$\hat{H}\phi_\alpha = E_\alpha\phi_\alpha$$

$$\psi(R,0) = \sum_\alpha \phi_\alpha(R)\left\langle \phi_\alpha \middle| \psi(0)\right\rangle$$

$$\psi(R,t) = \sum_\alpha \phi_\alpha(R)e^{-t(E_\alpha-E_T)}\left\langle \phi_\alpha \middle| \psi(0)\right\rangle$$

$$\lim_{t\to\infty}\psi(R,t) = \phi_0(R)e^{-t(E_0-E_T)}\left\langle \phi_0 \middle| \psi(0)\right\rangle$$

$$E_0 \approx E_T \Rightarrow \textit{normalization fixed}$$

Monte Carlo process

- Now consider the variable "t" as a continuous time *(it is really imaginary time).*
- Take derivative with respect to time to get evolution.
- This is a diffusion + branching process.
- Justify in terms of Trotter's theorem.

Requires interpretation of the wavefunction as a probability density.

But is it? Only in the boson ground state. Otherwise there are nodes. Come back to later.

$$-\frac{\partial \psi(R,t)}{\partial t} = (H - E_T)\psi(R,t)$$

$$H = -\sum_i \frac{\hbar^2}{2m_i}\nabla_i^2 + V(R)$$

$$\begin{cases} -\dfrac{\partial \psi(R,t)}{\partial t} = -\displaystyle\sum_i \frac{\hbar^2}{2m_i}\nabla_i^2 \psi(R,t) \\ -\dfrac{\partial \psi(R,t)}{\partial t} = (V(R) - E_T)\psi(R,t) \end{cases}$$

Trotter's formula

- How do we find the solution of:

$$\frac{d\hat{\rho}}{dt} = (A+B)\hat{\rho}$$

- The operator solution is:

$$\hat{\rho} = e^{(A+B)t}$$

- Trotter's formula (1959):

$$\hat{\rho} = \lim_{n\to\infty}\left[e^{\frac{t}{n}\hat{A}}e^{\frac{t}{n}\hat{B}}\right]^n$$

- Assumes that A,B and A+B are reasonable operators.

$$\left\langle R_0\left|\left[e^{\frac{t}{n}\hat{A}}e^{\frac{t}{n}\hat{B}}\right]^n\right|R_n\right\rangle = \left\langle R_0\left|e^{\frac{t}{n}\hat{A}}\right|R'_1\right\rangle\left\langle R'_1\left|e^{\frac{t}{n}\hat{B}}\right|R_1\right\rangle....\left\langle R_{n-1}\left|e^{\frac{t}{n}\hat{A}}\right|R'_n\right\rangle\left\langle R'_n\left|e^{\frac{t}{n}\hat{B}}\right|R_n\right\rangle$$

- This means we just have to figure out what each operator does independently and then alternate their effect. This is rigorous in the limit as n➔∞.
- In the DMC case A is diffusion operator, B is a branching operator.
- Just like "molecular dynamics" At small time we evaluate each operator separately.

Basic DMC algorithm

- Construct an ensemble (population P(0)) sampled from the trial wavefunction. $\{R_1, R_{2,\ldots}, R_P\}$
- Go through ensemble and diffuse each one (timestep τ)

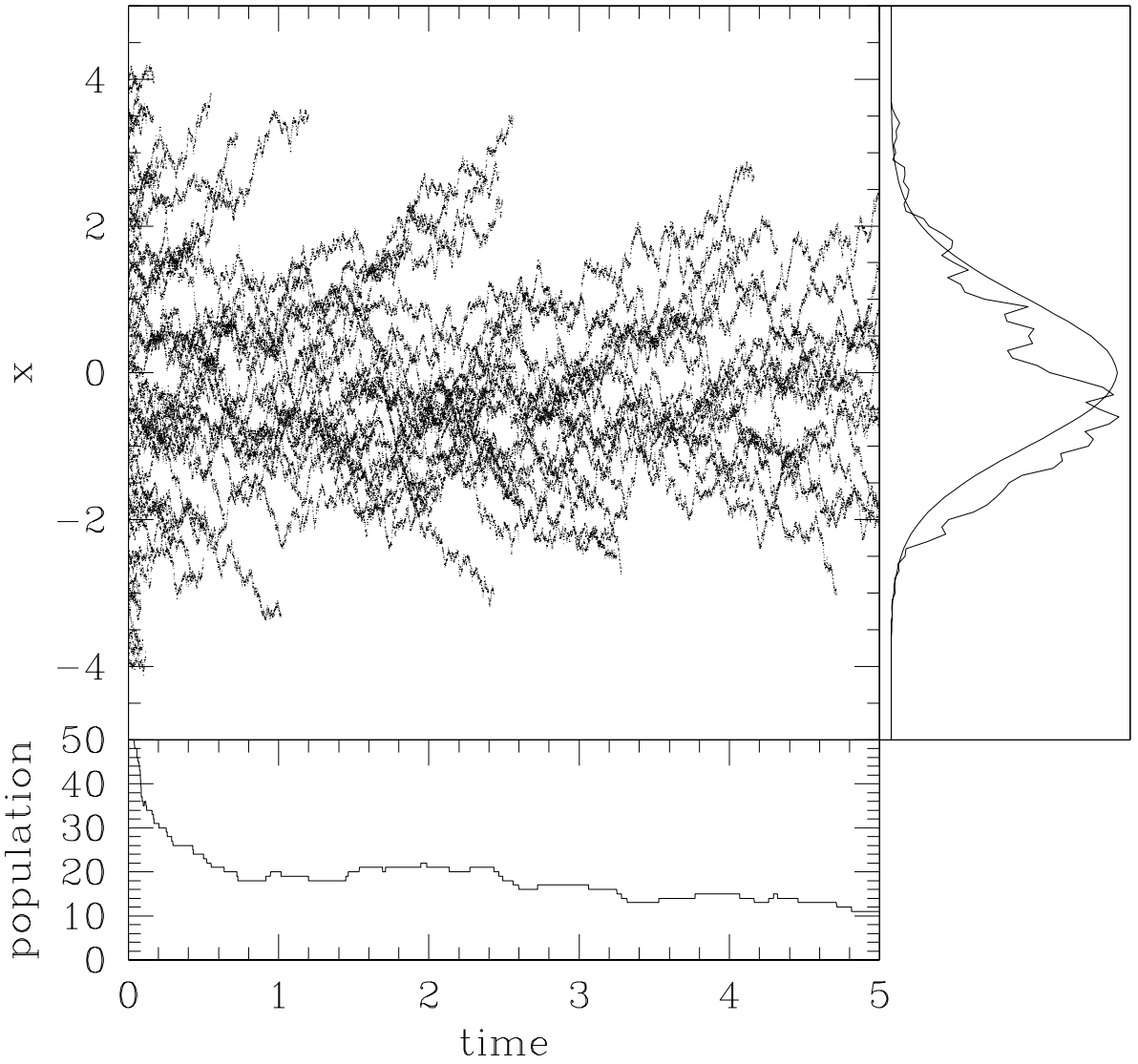
$$R'_k = R_k + \sqrt{2\lambda\tau}\,\zeta(t)$$ ← ndrn

- number of copies= $\underline{e^{-\tau(V(R)-E_T)} + u}$ ← uprn, floor function
- Trial energy E_T adjusted to keep population fixed.

$$E_0 = \lim_{t\to\infty} \frac{\int dRH\phi(R,t)}{\int dR\phi(R,t)} \approx \langle V(R)\rangle_{\phi(\infty)}$$

- Problems:
  - Branching is uncontrolled
  - Population unstable
  - What do we do about fermi statistics?

Harmonic Oscillator DMC



Population Bias

- Having the right trial energy guarantees that population will on the average be stable, but fluctuations will always cause the population to either grow too large or too small.
- Various ways to control the population
- Suppose P_0 is the desired population and P(t) is the current population. How much do we have to adjust E_T to make $P(t+T)=P_0$?

$$P(t+T) = e^{-T(-\delta E_T)} P(t) = P_0$$

$$\delta E_T = -\frac{\ln(P(t)/P_0)}{T}$$

- Feedback procedure:

$$\boxed{E_T = E_{T0} - \kappa \ln\left(P/P_0\right)}$$

- There will be a (small) bias in the energy caused by a limited population.

Importance Sampling

Kalos 1970, Ceperley 1979

- Why should we sample the wavefunction? The physically correct pdf is $|\varphi_0|^2$.
- Importance sample (multiply) by trial wave function.

$$f(R,t) \equiv \psi_T(R)\phi(R,t) \qquad \lim_{t\to\infty} f(R,t) \equiv \psi_T(R)\phi_0(R)$$

$$-\frac{\partial f(R,t)}{\partial t} = \psi_T(R) H\left[f(R,t)/\psi_T(R)\right] \qquad \text{Commute } \Psi \text{ through H}$$

$$-\frac{\partial f(R,t)}{\partial t} = -\lambda\nabla^2 f - \lambda\nabla\left(2f\nabla\ln\psi_T(R)\right) + \left(\psi_T{}^{-1} H \psi_T\right) f(R,t)$$

Evolution = diffusion + drift + branching

- Use accept/reject step for more accurate evolution.
  make acceptance ratio>99% . Determines time step.
- We have three terms in the evolution equation.
  Trotter's theorem still applies.

- To the pure diffusion algorithm we have added a drift step that pushes the random walk in directions of increasing trial function:
$$R' = R + 2\lambda\tau\nabla \ln \psi_T(R)$$
- Branching is now controlled by the local energy
$$E_L(R) - E_T = \psi_T^{-1}(R)\hat{H}\psi_T(R) - E_T$$
- Because of zero variance principle, fluctuations are controlled.
- Cusp condition can limit infinities coming from singular potentials.
- We still determine E_T by keeping asymptotic population stable.
$$E_0 = \lim_{t\to\infty} \frac{\int dR\phi(R,t)H\psi_T(R)}{\int dRf(R,t)} \approx \left\langle E_\psi(R)\right\rangle_{f(\infty)}$$
- Must have accurate “time” evolution. Adding accept/reject step is a major improvement.

Fermions?

- How can we do fermion simulations? The initial condition can be made real but not positive (for more than 1 electron in the same spin state)

- In *transient estimate* or *released-node* methods one carries along the sign as a weight and samples the modulus.

$$\phi(t) = e^{-(\hat{H}-E_T)t}\,\mathrm{sign}(\phi(R,0))\,|\,\phi(R,0)\,|$$

- Do not forbid crossing of the nodes, but carry along sign when walks cross.
- What's wrong with node release:
  - Because walks don't die at the nodes, the computational effort increases (bosonic noise)
  - The signal is in the cancellation which dominates

Monte Carlo can add but not subtract

Transient Estimate Approach

$$\Psi(\beta) = e^{-\frac{\beta}{2}H}\Psi$$

$$Z(\beta) = \langle \Psi(\beta)\Psi(\beta)\rangle = \langle \Psi e^{-\beta H}\Psi\rangle = \int dR_0 \ldots dR_p \Psi(R_0)\langle R_0 e^{-\tau H} R_1\rangle \ldots \langle R_{p-1} e^{-\tau H} R_p\rangle \Psi(R_p)$$

$$E(\beta) = \frac{\langle \Psi(\beta) H \Psi(\beta)\rangle}{\langle \Psi(\beta)\Psi(\beta)\rangle} = \langle E_L(R_0)\rangle_\beta \qquad \tau = \frac{\beta}{p}$$

- Ψ(β) converges to the exact ground state
- E(β) is an upper bound converging to the exact answer monotonically
- Define the sign of a walker: $\sigma(R) = \dfrac{\Psi(R)}{|\Psi(R)|}$

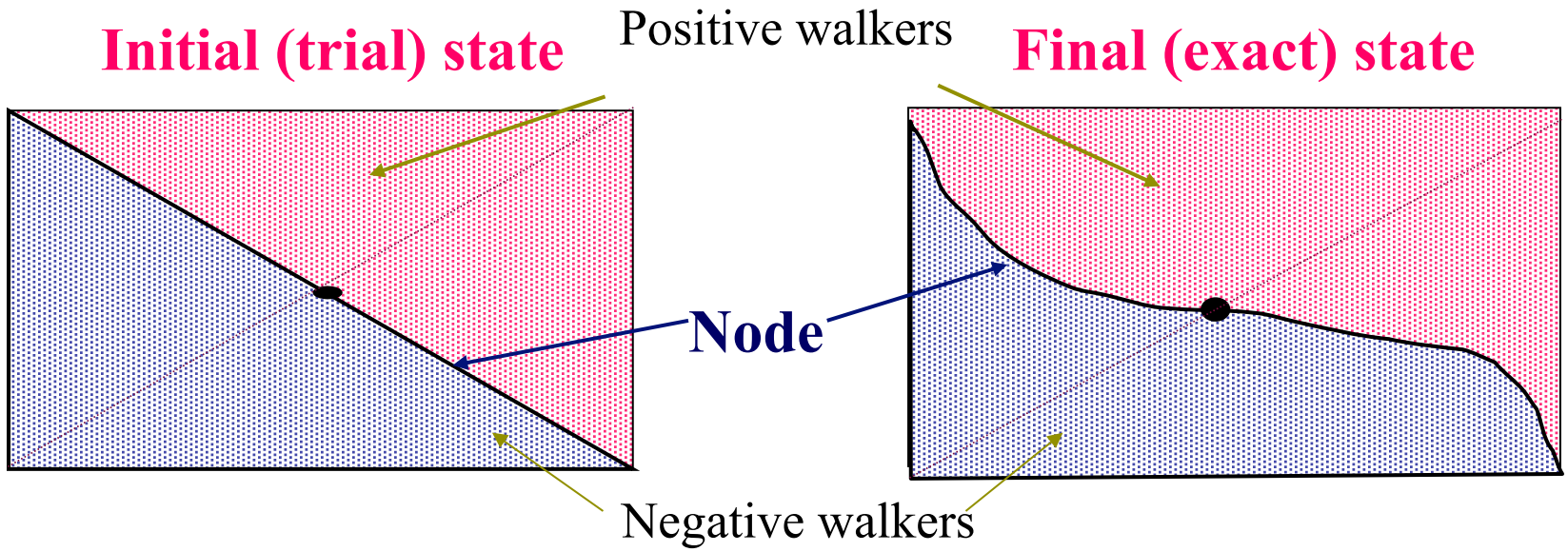
$$Z(\beta) = \int dR_0 \ldots dR_p |\Psi(R_0)|\langle R_0 e^{-\tau H} R_1\rangle \ldots \langle R_{p-1} e^{-\tau H} R_p\rangle |\Psi(R_p)| \sigma(R_0)\sigma(R_P)$$

$$\frac{Z_{\text{fermi}}}{Z_{\text{bose}}} = \langle \sigma(R_0)\sigma(R_P)\rangle$$

Model fermion problem: Particle in a box

Symmetric potential: V(**r**) =V(-**r**)

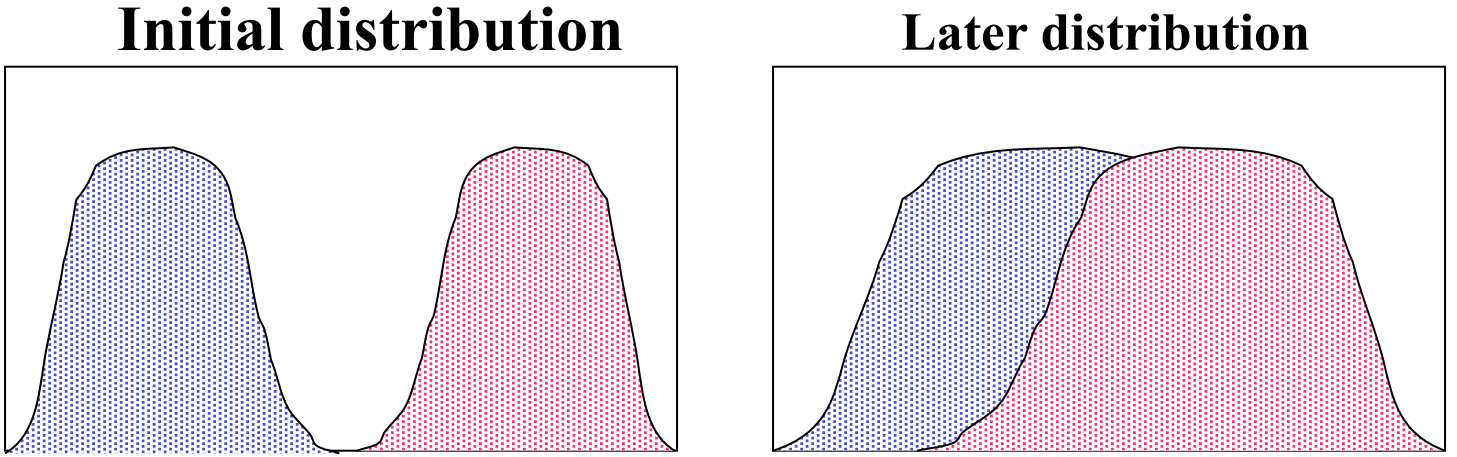
Antisymmetric state: $\phi(\mathbf{r})=-\varphi(-\mathbf{r})$



Sign of walkers fixed by initial position. They are allowed to diffuse freely. f(r)= number of positive-negative walkers. Node is dynamically established by diffusion process. (cancellation of positive and negative walkers.)

$$\langle E(t)\rangle = \frac{\sum \sigma(0)\sigma(t)E(t)}{\sum \sigma(0)\sigma(t)}$$

Scaling in Released-Node



- At any point, positive and negative walkers will tend to cancel so the signal is drown out by the fluctuations.
- Signal/noise ratio is : $e^{-t[E_F - E_B]}$ **t**=projection time

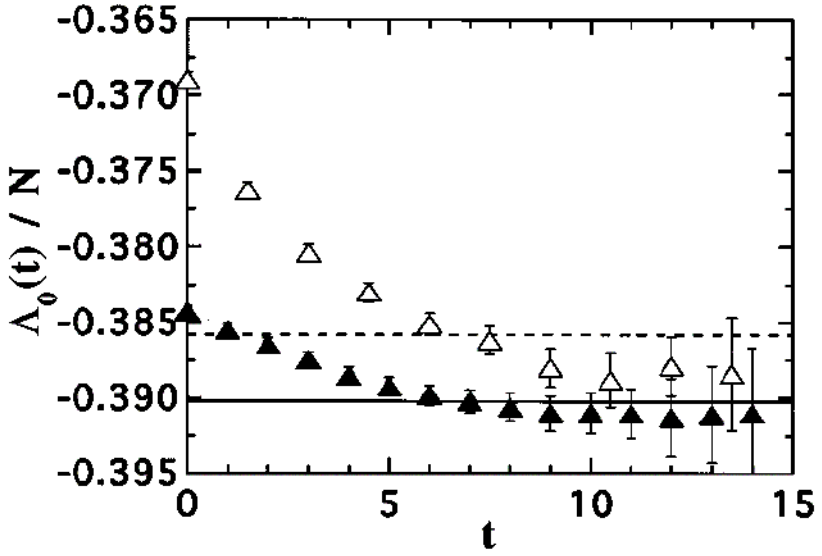
  E_F and E_B are Fermion, Bose energy (proportional to **N**)
- Converges but at a slower rate. Higher accuracy, larger **t**.
- For general excited states:

  Exponential complexity! $\mathrm{CPUtime} \propto \varepsilon^{-2(1+\frac{E_F}{E_g})} \approx \varepsilon^{-2N\frac{e_F}{E_g}}$
- Not a fermion problem but an excited state problem.
- Cancellation is difficult in high dimensions.

Exact fermion calculations

- Possible for the electron gas for up to 60 electrons.
- 2DEG at rs=1 N=26
- Transient estimate calculation with SJ and BF-3B trial functions.

$$\left\langle \Psi_T \left| e^{-tH} \right| \Psi_T \right\rangle$$



General statement of the "fermion problem"

- Given a system with **N** fermions and a known Hamiltonian and a property **O.** (usually the energy).
- How much time **T** will it take to estimate **O** to an accuracy **ε**? How does **T** scale with **N** and **ε**?
- If you can map the quantum system onto an equivalent problem in classical statistical mechanics then:

$$T \propto N^{\alpha} \varepsilon^{-2} \qquad \text{With } 0 < \alpha < 4$$

This would be a "solved" quantum problem!

- All approximations must be controlled!
- Algebraic scaling in N!

e.g. properties of Boltzmann or Bose systems in equilibrium.

“Solved Problems”

- 1-D problem. (simply forbid exchanges)
- Bosons and Boltzmanons at any temperature
- Some lattice models: Heisenberg model, 1/2 filled Hubbard model on bipartite lattice (Hirsch)
- Spin symmetric systems with purely attractive interactions: u<0 Hubbard model, nuclear Gaussian model.
- Harmonic oscillators or systems with many symmetries.
- Any problem with $\langle i|H|j\rangle \leq 0$
- Fermions in special boxes
- Other lattice models

The sign problem

- The fermion problem is intellectually and technologically very important.
- Progress is possible but ***danger***-the problem maybe more subtle than you first might think. New ideas are needed.
- No fermion methods are perfect but QMC is competitive with other methods and more general.
- The fermion problem is one of a group of related problems in quantum mechanics (**e.g** dynamics).
- Feynman argues that general many-body quantum simulation is exponentially slow on a classical computer.

Fixed-node method

- Initial distribution is a pdf.
  It comes from a VMC simulation. $f(R,0) = |\psi_T(R)|^2$
- Drift term pushes walks away from the nodes.
- Impose the condition: $\phi(R) = 0 \;\text{ when }\; \psi_T(R) = 0.$
- This is the fixed-node BC

- Will give an *upper bound* to the exact energy, the best upper bound consistent with the FNBC.

$$E_{FN} \geq E_0$$

$$E_{FN} = E_0 \;\text{ if }\; \phi_0(R)\psi(R) \geq 0 \;\text{ all } R$$

- f(R,t) has a discontinuous gradient at the nodal location.
- Accurate method because Bose correlations are done exactly.
- Scales well, like the VMC method, as N^3. Classical complexity.
- Can be generalized from the continuum to lattice finite temperature, magnetic fields, …
- One needs trial functions with accurate nodes.

Nodal Properties

If we know the sign of the exact wavefunction (the nodes), we can solve the fermion problem with the fixed-node method.

- If $\varphi(R)$ is real, nodes are $\varphi(R)=0$ where R is the 3N dimensional vector.
- Nodes are a 3N-1 dimensional surface. (Do not confuse with single particle orbital nodes!)
- Coincidence points $\mathbf{r}_i = \mathbf{r}_j$ are 3N-3 dimensional hyper-planes
- In 1 spatial dimension these "points" exhaust the nodes: *fermion problem is easy to solve in 1D* with the "no crossing rule."
- Coincidence points (and other symmetries) only constrain nodes in higher dimensions, they do not determine them.
- The nodal surfaces define nodal volumes. How many nodal volumes are there? **Conjecture**: there are typically *only* 2 different volumes (+ and -) except in 1D. (but only demonstrated for free particles.)

Fixed-Phase method

Ortiz, Martin, DMC 1993

- Generalize the FN method to complex trial functions: $\Psi(R) = e^{-U(R)}$
- Since the Hamiltonian is Hermitian, the variational energy is real:

$$E_V = \frac{\int dR\ e^{-2\Re U(R)} \left[V(R) + \lambda \nabla^2 U(R) - \lambda \left[\Re \nabla U(R) \right]^2 + \lambda \left[\Im \nabla U(R) \right]^2 \right]}{\int dR\ e^{-2\Re U(R)}}$$

- We see only one place where the energy depends on the phase of the wavefunction.
- We fix the phase, then we add this term to the potential energy. In a magnetic field we get also the vector potential.

$$\text{effective potential} = V(R) + \sum_i \lambda_i \left[A(r_i) + \Im \nabla_i U(R) \right]^2$$

- We can now do VMC or DMC and get upper bounds as before.
- The imaginary part of the local energy will not be zero unless the right phase is used.
- Used for twisted boundary conditions, magnetic fields, vortices, phonons, spin states, …

Fermions: antisymmetric trial function

- At mean field level the wavefunction is a Slater determinant. Orbitals for homogenous systems are a filled set of plane waves.
- We can compute this energy analytically (HF).
- To include correlation we multiply by a "jastow". We need MC to evaluate properties.
- New feature: how to compute the derivatives of a deteminant and sample the determinant. Use tricks from linear algebra.
- Reduces complexity to $O(N^2)$.

$$\Psi_s(R) = Det\left\{e^{ik_i r_j}\eta_i\left(\sigma_j\right)\right\}$$

$$\text{PBC:}\quad k \cdot L = 2\pi n + \{\theta\}$$

$$\Psi_{SJ}(R) = Det\{e^{ik_i r_j}\}e^{-\sum_{i<j} u(r_{ij})}$$

Slater-Jastrow trial function.

$$\det\left(\phi_k\left(r_j^T\right)\right) = \det\left(\phi_k\left(r_j\right)\right)\sum_k \phi_k\left(r_j^T\right)M^{-1}_{k,i}$$

$$\frac{1}{\det(M)}\frac{\partial \det(M)}{\partial a} = Tr\left\{M^{-1}\frac{\partial M}{\partial a}\right\}$$

Jastrow factor for the e-gas

- Look at local energy either in r space or k-space:
- r-space as 2 electrons get close gives cusp condition: $du/dr|_0=-1$
- K-space, charge-sloshing or plasmon modes.

$$2\rho u_k = \sqrt{\frac{V_k}{\lambda k^2}} \propto \frac{1}{k^2}$$

- Can combine 2 exact properties in the Gaskell form. Write E_V in terms structure factor making "random phase approximation." (RPA).

$$2\rho u_k = -\frac{1}{S_k} + \sqrt{\frac{1}{S_k^2} + \frac{V_k}{\lambda k^2}} \qquad S_k = \text{ideal structure factor}$$

- Optimization can hardly improve this form for the e-gas in either 2 or 3 dimensions. RPA works better for trial function than for the energy.
- NEED EWALD SUMS because potential trial function is long range, it also decays as 1/r, but it is not a simple power.

$$\lim_{r\to\infty} u(r) = \begin{cases} r^{-1} & \text{3D} \\ r^{-1/2} & \text{2D} \\ \log(r) & \text{1D} \end{cases}$$

Long range properties important

- Give rise to dielectric properties
- Energy is insensitive to u_k at small k
- Those modes converge $t\sim 1/k^2$

Wavefunctions beyond Jastrow

- Use method of residuals construct a sequence of increasingly better trial wave functions. Justify from the Importance sampled DMC.
- Zeroth order is Hartree-Fock wavefunction
- First order is Slater-Jastrow pair wavefunction (RPA for electrons gives an analytic formula)
- Second order is 3-body backflow wavefunction
- Three-body form is like a squared force. It is a bosonic term that does not change the nodes.

smoothing

$$\phi_{n+1}(R) \approx \phi_n(R) e^{-\tau <\phi_n^{-1} H \phi_n>}$$

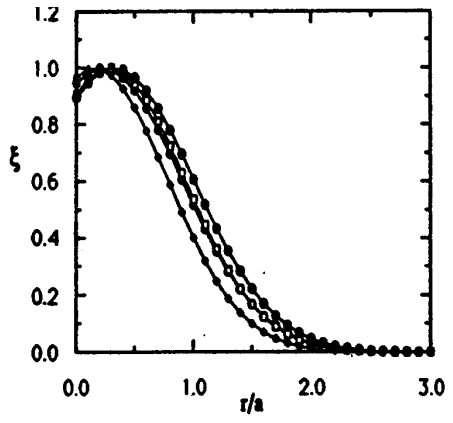
$$\phi_0 = e^{i\sum_j \mathbf{k}_j \bullet \mathbf{r}_j}$$

$$E_0 = V(R)$$

$$\phi_1 = \phi_0 e^{-U(R)}$$

$$E_1 = U(R) - \left[\nabla W(R)\right]^2 + i\sum_j \mathbf{k}_j \bullet \left(\mathbf{r}_j - \nabla_j Y(R)\right)$$

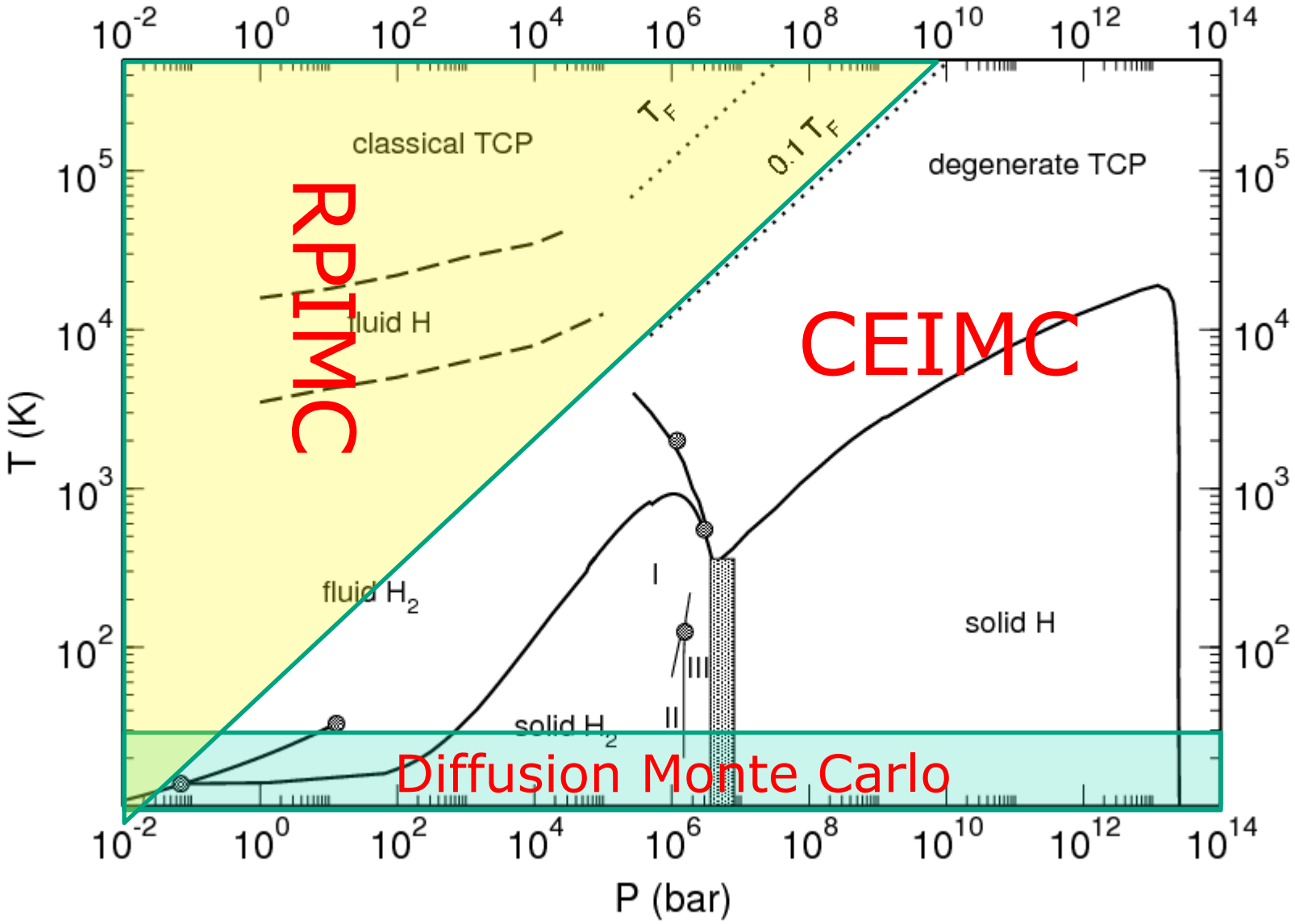
$$\exp\{\sum_i [\sum_j \xi_{ij}(r_{ij})(\mathbf{r}_i - \mathbf{r}_j)]^2\}$$



Why study dense Hydrogen?

- Applications:
  - Astrophysics: giant planets, exoplanets
  - Inertially confined fusion: NIF
  - H is an ubiquitous atom!
- Fundamental physics:
  - Which phases are stable?
  - Superfluid/ superconducting phases?
  - “Holy grail” of high pressure physics
- Benchmark for simulation:
  - “Simple” electronic structure; no core states
  - But strong quantum effects from its nuclei
  - If we can’t simulate dense hydrogen what chance do we have for heavier elements?

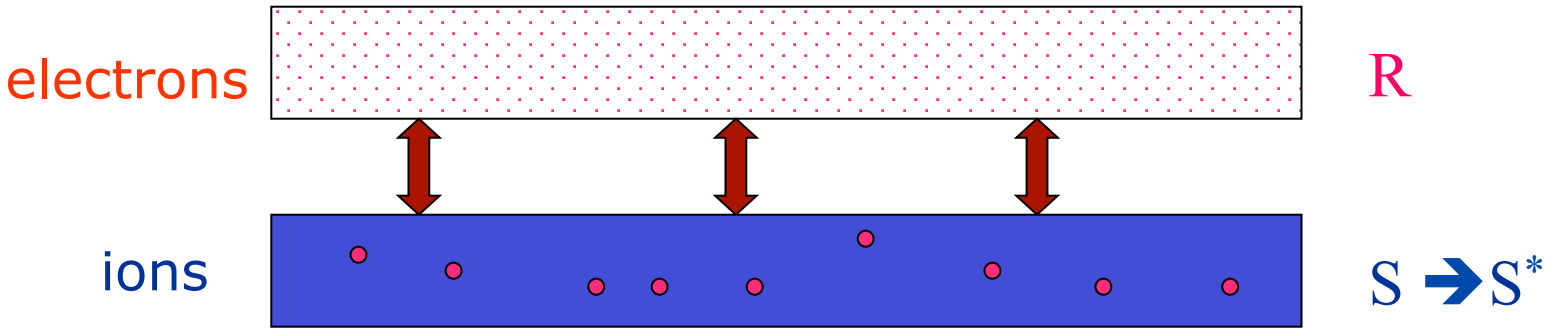
Regimes for Quantum Monte Carlo



Coupled Electron-Ionic Monte Carlo:CEIMC

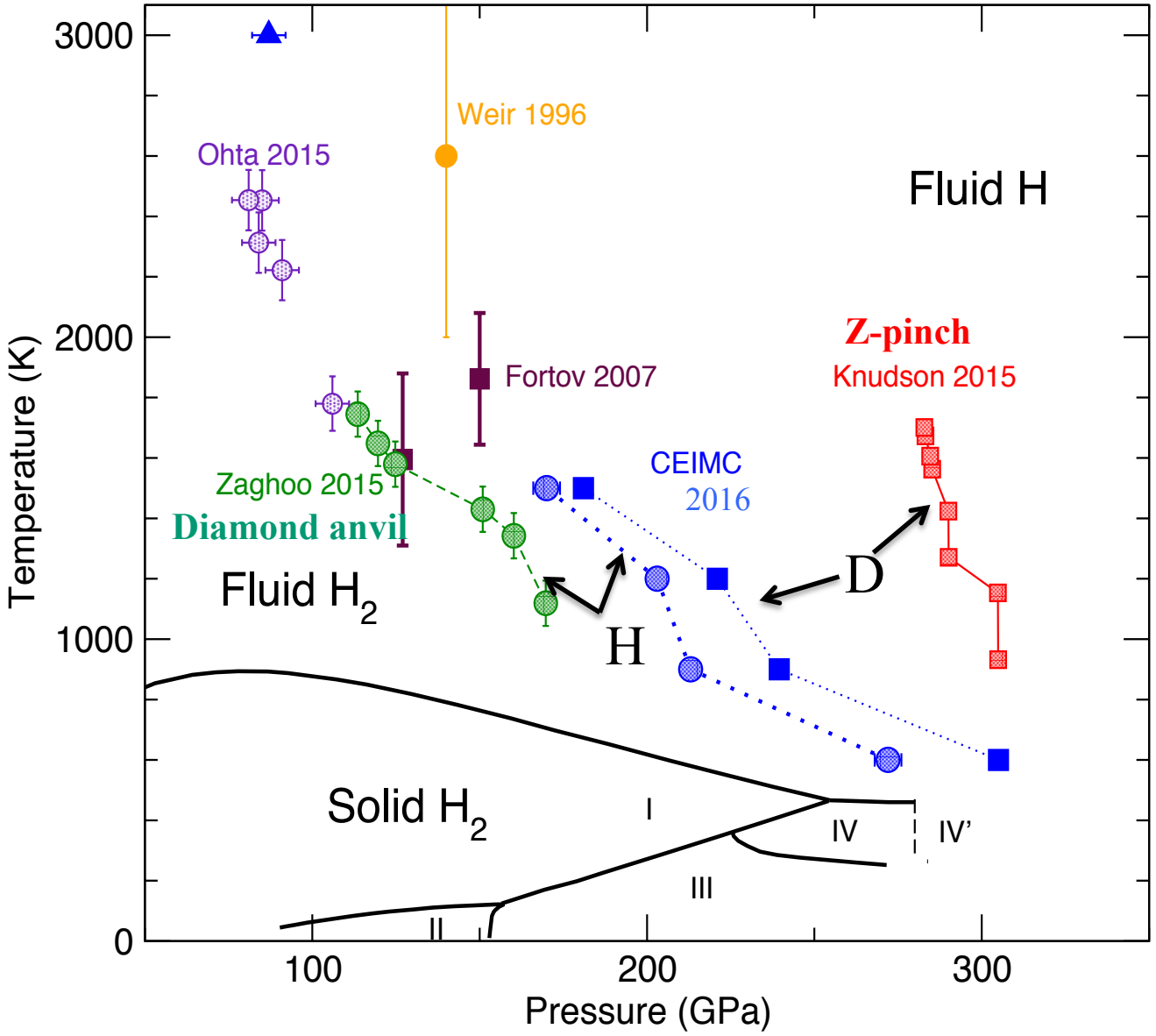
How to simulate a liquid with QMC

1. Do Path Integrals for the ions at T>0.
2. Let electrons be at zero temperature, a reasonable approximation for $T<<E_F$.
3. Use Metropolis MC to accept/reject moves based on QMC computation of electronic energy



The "noise" coming from electronic energy can be treated without approximation using the penalty method.

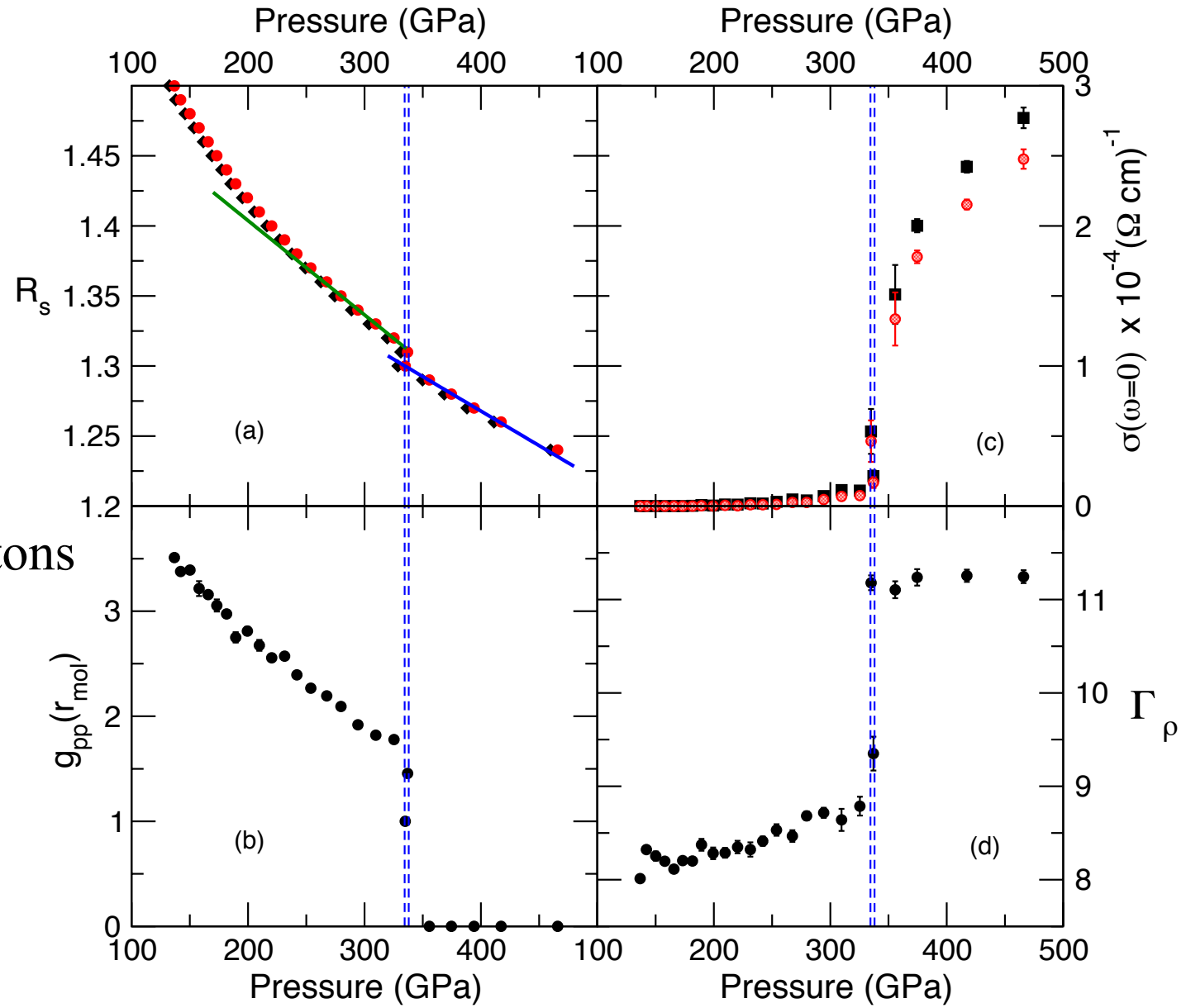
Experimental results differ by a factor 2!!
QMC is in the middle.



Properties in transition region

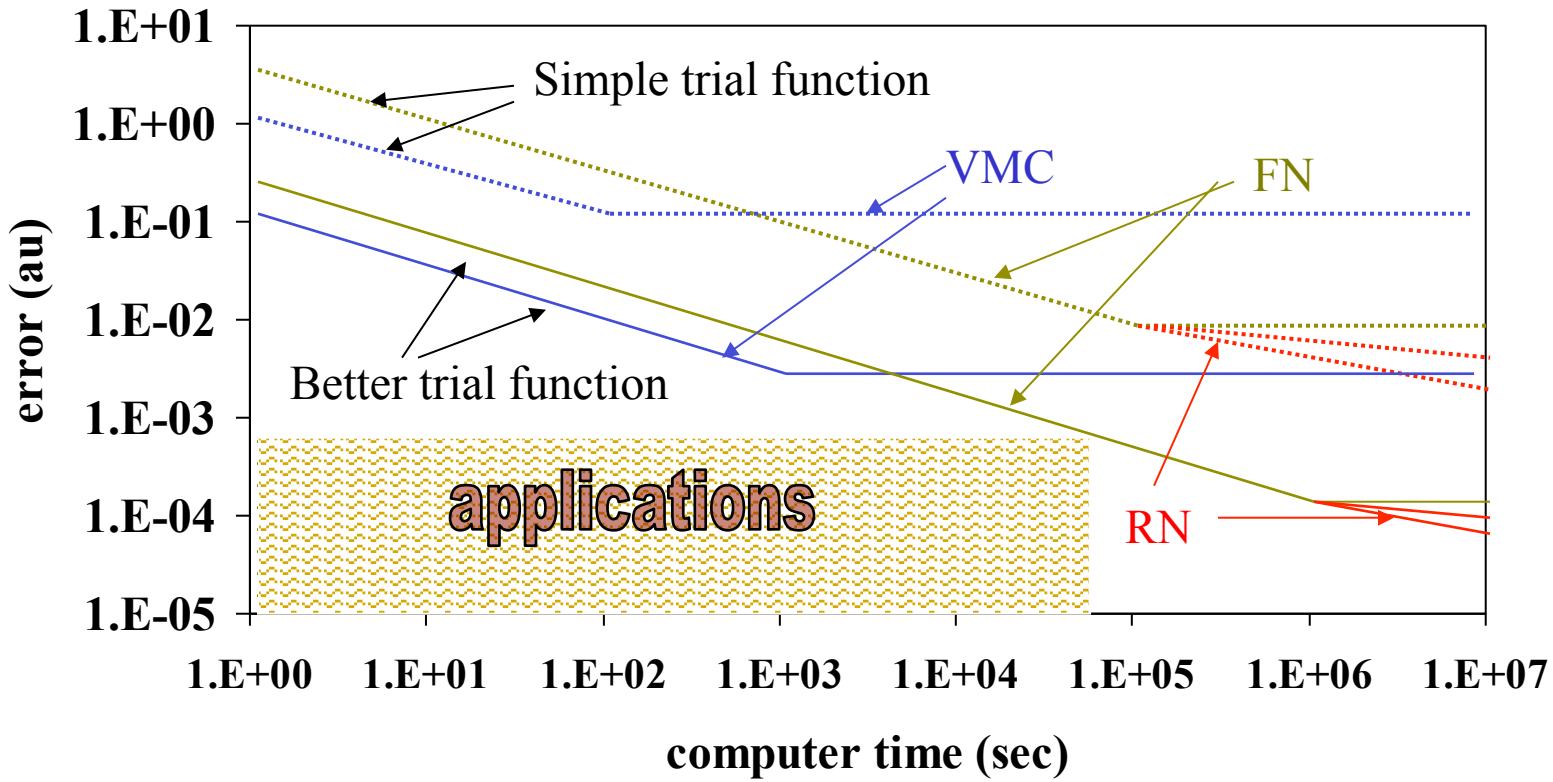
T=600K

Classical protons



Summary of T=0 methods:

Variational(VMC), Fixed-node(FN), Released-node(RN)



Summary of projector methods

- Fixed-node is a super-variational method
- DMC dynamics is determined by Hamiltonian
- Zero-variance principle allows very accurate calculation of ground state energy if trial function is good.
- Excellent application for parallel computers.

- Projector methods need a trial wavefunction for accuracy. They are essentially methods that perturb from the trial function to the exact function. (Note: if you don't use a trial function, you are perturbing from the ideal gas)
- Difficulty calculating properties other than energy. We must use "extrapolated estimators" or "forward walking".

$$f(R,\infty) = \phi_0(R)\psi_T(R) \quad \text{not} \quad |\phi_0(R)|^2$$

- Bad for phase transitions esp. at finite temperature

Potential energy

- Write potential as integral over structure function:

$$V = \int d^3k \frac{4\pi}{k^2} S(k) \qquad S(k) = \langle \rho_{-k} \rho_k \rangle = 1 + (N-1) \left\langle e^{i(r_1 - r_2)k} \right\rangle$$

- Error comes from 2 effects.
  - Approximating integral by sum
  - Finite size effects in S(k) at a given k.

$$S_{HF}(k) = 1 - \frac{2}{N} \sum_{q,q'} \delta_{q-q'+k}$$

- Within HF we get exact S(k) with TABC.
- Discretization errors come only from non-analytic points of S(k).
  - the absence of the k=0 term in sum. We can put it in by hand since we know the limit S(k) at small k (plasmon regime)
  - Remaining size effects are smaller, coming from the non-analytic behavior of S(k) at $2k_F$.

$$S(k) = \frac{k^2}{2m\omega_p} \Rightarrow \Delta E = \frac{\omega_p}{N}$$