Improved efficiency with variational Monte Carlo using two level sampling

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A two level sampling method is applied to variational Monte Carlo (VMC) that samples the one and two body parts of the wave function separately. The method is demonstrated on a single Li$_2$ molecule in free space and 32 H$_2$ molecules in a periodic box. This simple modification increases the efficiency of a VMC simulation by up to 72%.

Variational Monte Carlo (VMC) [1] is an effective method for computing the ground state properties of atoms, [2] [3] molecules, [4] [5] and solids. [6] [7] [8] [9] Explicitly including the electron correlation into the wave function allows VMC to recover a large percentage of the correlation energy, typically 70-90% (or more for small atoms and molecules). [2] [3] [10]

VMC computes matrix elements with respect to the square of a trial wave function, $\psi$. The most common matrix element is the energy,

$$E = \int dR \frac{\psi^2(R)}{\int dR \psi^2(R)} E_L(R; a)$$

(1)

where $E_L = \frac{1}{\psi} H \psi$. The variational principle states that this energy will be greater than or equal to the true ground state energy for any $\psi$. Typically, the wave function is parameterized and those parameters are varied until a minimum energy [10] (or alternatively, minimum variance [2]) is reached.

The Metropolis method [11] constructs a transition probability, $P(s' \rightarrow s)$, for use in a Markov process. The result of that process is the desired normalized probability distribution. A correct transition probability can be constructed by satisfying detailed balance,

$$\pi(s) P(s \rightarrow s') = \pi(s') P(s' \rightarrow s)$$

(2)

where $\pi(s)$ is the desired probability distribution ($\psi^2(R)$ for VMC). In the generalized Metropolis method, the transition probability is split into two pieces, an a priori sampling distribution, $T(s \rightarrow s')$ and an acceptance probability $A(s \rightarrow s')$.

The original Metropolis method generates a trial move, $R'$, of one particle inside a box of side $\Delta$ centered about the old position, $R$. This leads to a constant $T$ inside the box and zero outside. The trial move is then accepted or rejected with probability

$$A = \min \left[ 1, \frac{\psi^2(R')}{\psi^2(R)} \right]$$

(3)

In electronic problems, a typical wave function is

$$\psi = De^{-U}$$

(4)

where $D$ is the product of spin up and spin down Slater determinants of single particle orbitals and $U$ is the two (or higher) body Jastrow factor. This latter part contains the explicit electron-electron correlation.

A simulation step consists of a trial move of every electron. Each trial move is accepted or rejected according to Eq. (4).

A single electron is moved at a time, which only changes one column of the Slater matrix. By saving and using the inverse of this matrix, computing the new determinant and its derivatives is an $O(N)$ operation. [12]

If the move is accepted, the inverse matrix can be updated with $O(N^2)$ operations. (Note that computing the determinant directly is of order $O(N^3)$.) A smaller acceptance ratio will be faster, since fewer updates need to be performed.

Multilevel sampling has been used extensively in path integral Monte Carlo. [13] In multilevel sampling, the wave function is split into several factors (levels), and an accept/reject step is performed after computing each factor. The entire wave function is computed only if there is an acceptance after every factor. Rejections will make the overall algorithm faster, since not all the factors need to be computed. For VMC it is natural to use two factors - the single body part, $D$, as one factor and the two body part, $e^{-U}$, as the other. The single body part is the cheaper of the two to compute, so computing it first will give the greatest reduction in time.

The two level sampling algorithm for VMC proceeds as follows. A trial move, $R'$, is proposed and accepted at the first stage with probability

$$A_1 = \min \left[ 1, \frac{D^2(R')}{D^2(R)} \right]$$

(5)

If accepted at the first stage, the two body part is computed and the trial move is accepted with probability

$$A_2 = \min \left[ 1, \frac{\exp[-2U(R')] \psi^2(R')}{\exp[-2U(R)] \psi^2(R)} \right]$$

(6)

It can be verified by substitution that these satisfy detailed balance in Eq. (4). After an acceptance at this
second level, the inverse Slater matrices are updated as described previously.

The figure of merit for these simulations is the efficiency

\[ \xi = \frac{1}{\sigma^2 T} \quad (7) \]

where \( T \) is the computer time and \( \sigma \) is the error estimate of the average of some property such as the energy.

Maximum efficiency results from a competition between two effects. The first is that a lower acceptance ratio will yield a lower run time and a larger efficiency. The second is serial correlations. A lower acceptance ratio will give a longer correlation time, hence a larger error and lower efficiency.

In maximizing the efficiency, the obvious parameter to adjust is the step size, \( \Delta \). But the number of steps between computations of \( E_L \) can also be adjusted. The Metropolis algorithm produces correlated state points, so successive samples of \( E_L \) don’t contain much new information. It is advantageous to compute \( E_L \) every few steps rather than every step. In this work the local energy was computed every 5 steps.

The algorithm is demonstrated using a Li$_2$ molecule in free space and a collection of 32 H$_2$ molecules in a periodic box. The wave functions, which are the \( \Psi_{\text{III}} \)'s from Reynolds, et al., use a simple electron-electron and electron-nuclear Jastrow term, and use floating Gaussians for the orbitals. The hydrogen molecules were in box of side 19.344 atomic units (\( r_s = 3.0 \)).

The results for the different sampling methods for the Li$_2$ molecule are given in Tables I and II. The second level acceptance ratio is quite high, indicating the single body part is a good approximation to the whole wave function. The efficiency is also shown in Figure 1.

The results for the different sampling methods for the H$_2$ molecules are given in Tables III and IV. The efficiency is also shown in Figure 2.

Comparing the maximum efficiency for each sampling method, two level sampling is 39% more efficient than the standard sampling for Li$_2$, and 72% more efficient for 32 H$_2$’s.

More complicated schemes using more levels or different splittings of the wave function could be devised. This particular scheme is attractive because it uses quantities readily available in a VMC computation, and requires minimal modification to existing VMC sampling algorithms.

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TABLE I: Timings for Li$_2$ molecule using the standard sampling method. All times in seconds on an SGI Origin 2000.

| $\Delta$ | Acceptance Ratio | Determinant Time | Jastrow Time | Total Time | $\xi$ |
|----------|------------------|------------------|--------------|------------|------|
| 1.0      | 0.610            | 48.3             | 340          | 516        | 1190 |
| 1.5      | 0.491            | 48.1             | 340          | 508        | 1680 |
| 2.0      | 0.407            | 48.2             | 340          | 503        | 1460 |
| 2.5      | 0.349            | 48.2             | 339          | 499        | 1070 |
| 3.0      | 0.307            | 48.2             | 339          | 496        | 800  |

TABLE II: Timings for Li$_2$ molecule using the two level sampling method. All times in seconds on an SGI Origin 2000.

| $\Delta$ | First Level Acc. Ratio | Second Level Acc. Ratio | Total Acc. Ratio | Time | $\xi$ |
|----------|------------------------|-------------------------|------------------|------|------|
| 1.0      | 0.674                  | 0.899                   | 0.606            | 400  | 1580 |
| 1.5      | 0.543                  | 0.894                   | 0.485            | 347  | 2430 |
| 2.0      | 0.447                  | 0.897                   | 0.401            | 304  | 2340 |
| 2.5      | 0.379                  | 0.902                   | 0.342            | 276  | 1910 |
| 3.0      | 0.331                  | 0.906                   | 0.300            | 256  | 1400 |

TABLE III: Timings for 32 H$_2$ molecules in a periodic box using the standard sampling method. All times in seconds on a Sun Ultra 5.

| $\Delta$ | Acceptance Ratio | Determinant Time | Jastrow Time | Total Time | $\xi$ |
|----------|------------------|------------------|--------------|------------|------|
| 2.0      | 0.606            | 167              | 1089         | 2015       | 0.61 |
| 3.0      | 0.455            | 167              | 1085         | 1891       | 1.22 |
| 4.0      | 0.338            | 166              | 1084         | 1794       | 1.23 |
| 5.0      | 0.250            | 166              | 1080         | 1722       | 1.06 |
| 6.0      | 0.185            | 164              | 1080         | 1668       | 1.02 |
| 7.0      | 0.139            | 162              | 1084         | 1629       | 0.76 |

TABLE IV: Timings for 32 H$_2$ molecules in a periodic box using the two level sampling method. All times in seconds on a Sun Ultra 5.

| $\Delta$ | First Level Acc. Ratio | Second Level Acc. Ratio | Total Acc. Ratio | Time | $\xi$ |
|----------|------------------------|-------------------------|------------------|------|------|
| 2.0      | 0.740                  | 0.795                   | 0.589            | 1804 | 0.59 |
| 3.0      | 0.598                  | 0.728                   | 0.436            | 1421 | 1.77 |
| 4.0      | 0.468                  | 0.681                   | 0.319            | 1185 | 2.11 |
| 5.0      | 0.357                  | 0.649                   | 0.232            | 994  | 1.55 |
| 6.0      | 0.370                  | 0.627                   | 0.169            | 849  | 1.87 |
| 7.0      | 0.204                  | 0.609                   | 0.124            | 740  | 1.46 |
FIG. 1: Efficiency of VMC for Li$_2$.

FIG. 2: Efficiency of VMC for 32 H$_2$ molecules.