Abstract Within the Correlated Gaussian Method the parameters of the Gaussian basis functions are often chosen stochastically using pseudo-random sequences. We show that alternative low-discrepancy sequences, also known as quasi-random sequences, provide bases of better quality.

1 Introduction

The Correlated Gaussian Method is a popular variational method for solving few-body problems in quantum mechanics [1,2]. The wave-function of a quantum few-body system is represented as a linear combination of correlated Gaussians. This reduces the corresponding Schrodinger equation to a generalised matrix eigenvalue problem in the (non-orthogonal) basis of correlated Gaussians. One of the advantages of the Correlated Gaussian Method is that the involved matrix elements are often analytic [3] which significantly simplifies the calculations.

In realistic calculations the dimension of the parameter space of the Gaussian basis is typically too large to be sampled deterministically. Therefore the basis is often constructed stochastically: the parameters of the Gaussians in the basis are chosen randomly using a pseudo-random number generator [1,2].

However, the pseudo-random sequences have the property of high discrepancy or, in other words, low uniformity [4]. Pseudo-random sequences tend to produce relatively large sub-areas of the parameter space that are not sampled at all, and other sub-areas that are sampled excessively. This can lead to bases of lower quality.

On the contrary, the low-discrepancy sequences (also called quasi-random sequences) are designed with the specific purpose to reduce discrepancy and to sample uniformly all sub-areas. Quasi-random sequences might thus potentially produce bases of higher quality.

We compare the quality of Gaussian bases constructed stochastically using pseudo- and quasi-random (Van der Corput) sequences by calculating the ground state energies of two Coulombic three-body systems. We show that quasi-random sequences consistently outperform pseudo-random sequences by providing a lower variational estimate of the ground-state energy.

2 Pseudo-random and Quasi-random Sequences

Pseudo-random sequences are computer generated sequences of numbers which satisfy the statistical criteria for a truly random sequence, that is, the absence of correlations of any type. The consequence is that pseudo-random sequences have high discrepancy or, equivalently, low uniformity. Pseudo-random sequences produce...
Fig. 1 The first 1000 points of a two-dimensional unit-square pseudo-random sequence (left) and quasi-random base-2/3 Van der Corput sequence (right). The pseudo-random sequence exhibits high discrepancy: there exist relatively large areas, exemplified by a circle at the upper left corner, with no points. The quasi-random sequence distributes points with lower discrepancy, that is, more uniformly relatively large non-sampled areas of the size on the order of $1/\sqrt{N}$, where $N$ is the length of the sequence. On the contrary, quasi-random sequences are highly correlated sequences built with the specific purpose of reducing the discrepancy down to approximately $1/N$ [4].

Van der Corput sequence is one simple low-discrepancy sequence constructed by reversing the base-$b$ representation of the sequence of natural numbers [5]. If the $n$-th natural number in base-$b$ representation is given as

$$n = \sum_{k=0}^{L-1} d_k(n)b^k,$$

where $d_k(n)$ are the digits, then the $n$-th number $q_n$ of the van der Corput sequence is given as

$$q_n = \sum_{k=0}^{L-1} d_k(n)b^{-k-1}.$$

The numbers in the van der Corput sequence are uniformly distributed over the unit interval with the discrepancy of $O(\log N/N)$. Figure 1 illustrates the reduced discrepancy of the Van der Corput sequence compared to a pseudo-random sequence.

3 Examples

We use two non-relativistic Coulombic three-body systems to compare the relative performance of pseudo- and quasi-random sequences: the positronium anion, $\text{Ps}^-$, and the $t\mu^{-}$ anion. We calculate the ground-state energies of these systems using correlated Gaussian bases constructed stochastically with pseudo-random and quasi-random sequences.

The Hamiltonian of a system of three bodies with massed $m_i$, charges $q_i$, and cartesian coordinates $r_i$, $i = 1\ldots3$, is given as

$$H = -\sum_{i=1}^{3} \frac{1}{2m_i} \partial^2\partial_{r_i^2}^2 + \sum_{i<j=1}^{3} \frac{q_i q_j}{|r_i - r_j|},$$

where atomic units are used, that is, the unit of charge is the charge of the positron, $e$, the unit of mass is the electron mass, $m_e$, the unit of length is the Bohr radius, $a = \hbar^2/(m_e e^2)$, and the unit of energy is Hartree, $\hbar = m_e e^4/\hbar^2$.

For the positronium anion the masses of the three constituents are $m_i = \{1, 1, 1\}$ and the charges $q_i = \{+1, -1, -1\}$. For the $t\mu^{-}$ molecule we use $m_i = \{5496.918, 3670.481, 206.7686\}$ and $q_i = \{+1, +1, -1\}$ [1].

The correlated Gaussians are parameterised in the form

$$\exp\left(-\sum_{i<j=1}^{3} \left(\frac{r_i - r_j}{b_{ij}}\right)^2\right),$$

where $b_{ij}$ are the correlation lengths. The Hamiltonian of three electronic states with $\{1, -1, -1\}$ and $\{+1, +1, -1\}$ charges is

$$H = -\sum_{i=1}^{3} \frac{1}{2m_i} \partial^2\partial_{r_i^2}^2 + \sum_{i<j=1}^{3} \frac{q_i q_j}{|r_i - r_j|} + \sum_{i=1}^{3} \frac{1}{2m_i} \partial^2\partial_{r_i^2}^2 + \sum_{i<j=1}^{3} \frac{q_i q_j}{|r_i - r_j|}.$$
Fig. 2 The ground state energy $E$ of the $Ps^-$ (left) and $td\mu$ (right) systems as function of the sampling scale factor $b_0$ from Eq. (5). The energies are calculated using stochastically chosen correlated Gaussian bases constructed with pseudo- or quasi-random sequences. The exact energies are $E_x = -0.26200507$ for $Ps^-$ and $E_x = -111.36444$ for $td\mu$ [1].

Fig. 3 The ground state energy $E$ of the $Ps^-$ (left) and $td\mu$ (right) systems as function of the size $n$ of the correlated Gaussian bases chosen stochastically with pseudo- or quasi-random sequences. The exact energies are $E_x = -0.26200507$ for $Ps^-$ and $E_x = -111.36444$ for $td\mu$ [1].

where $b_{ij}$ are the range parameters of the Gaussians. The range parameters are chosen stochastically following the usual uniform sampling strategy [1],

$$b_{ij} = ub_0,$$

where the number $u \in ]0, 1]$ is taken from a pseudo- or a quasi-random sequence, and where $b_0$ is the appropriate scale factor on the order of the size of the system.

For pseudo-random calculations we used the sequence produced by the `rand()` function from the Linux C library seeded to 13. For the quasi-random calculations we used three Van der Corput sequences with bases 2, 3, and 5.

The scale factor $b_0$ must be fine-tuned for a particular calculation. An example of fine-tuning is shown on Fig. 2 where the ground state energies of the two systems are plotted as function of the scale factor for the two sequences. The figure shows that (i) the minima at the optimal values are relatively broad and need not to be calculated exactly, and (ii) the quasi-random sequence outperforms the pseudo-random sequence for both systems.

We now take the optimum values of the scale factor from Fig. 2 and calculate the ground state energies of the two systems for bases with different sizes. Although the optimum scale factors depend slightly on the size of the basis, the dependence is not strong and can be neglected for the variations of the sizes that we use.

Figure 3 compares the ground-state energies of these systems calculated using correlated Gaussian bases constructed stochastically with pseudo- or quasi-random sequences. For all basis sizes the quasi-random sequence provides lower variational estimate of the ground-state energy than the pseudo-random sequence.
4 Conclusion

In the Correlated Gaussian method the variational basis is often constructed stochastically using pseudo-random sequences. Pseudo-random sequences have the intrinsic property of high discrepancy: they produce relatively large non-sampled areas. On the contrary, the low-discrepancy (quasi-random) sequences, like the Van der Corput sequence, are specifically designed to have low discrepancy.

We have compared the relative performance of the two types of sequences by calculating the ground-state energies of two Coulombic three-body systems using stochastic correlated Gaussian bases of various sizes built with both pseudo- and quasi-random sequences. For all bases sizes the quasi-random sequence outperforms the pseudo-random sequence by providing a lower variational estimate of the energy.

References

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