

Max-Planck-Institut
für Mathematik
in den Naturwissenschaften
Leipzig

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Preprint no.: 44

2007



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January 25, 2007

PACS numbers: 71.15.-m, 71.10.-w, 71.10.Ca

Abstract

We present a perturbative treatment of Jastrow-type correlation factors which focuses on an accurate description of short-range correlations. Our approach is closely related to coupled cluster perturbation theory with the essential difference that we start from a variational formulation for the energy. Such kind of perturbation theory is especially suited for multiscale bases, like wavelets, which provide sparse representations for Jastrow factors. Envisaged applications in solid state physics are confined many-particle systems like electrons or multi excitons in quantum dots. The resulting Jastrow factors can be further used as trial wavefunctions in quantum Monte Carlo calculations for these systems. First applications to a screened homogeneous Fermi gas model demonstrate that already first order Jastrow factors recover 95% of the correlation energy in variational Monte Carlo calculations over a fairly large range of densities and screening parameters. The corresponding second and third order perturbation energies turned out to be more sensitive to the specific choice of the model parameters. Furthermore, we have compared our first order Jastrow factors with those obtained from Fermi hypernetted chain calculations, where excellent agreement at short and intermediate inter-particle distances has been observed.

1 Introduction

The determination of ground state properties for interacting many-particle systems is a central topic in condensed matter physics and quantum chemistry. It can be either treated by *density functional theory* (DFT), or through a more direct approach based on certain kinds of many-particle wavefunctions. Within DFT, the original many-particle problem is mapped, via the Kohn-Sham equation [1], into an effective one-particle problem and thus considerably reduces the computational effort. Despite the great success of DFT for a large variety of systems, it has the basic drawback that there exists no systematic way to improve the exchange-correlation potential in the Kohn-Sham equation. On the contrary various schemes for a systematic improvement of many-particle wavefunctions exist. We just want to mention *coupled cluster* (CC) theory [2, 3] and the *Fermi hypernetted chain* (FHNC) method [4, 5]. By construction CC theory is aiming towards a direct approximation of the exact wavefunction, while the FHNC method deals with a more restricted class of Jastrow-type wavefunctions which is supposed to be very close to the exact one. A common feature of both methods is the ansatz

$$\Psi(\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N) = \mathcal{F} \Phi(\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N), \quad (1.1)$$

where a correlation operator \mathcal{F} acts on a single Slater determinant Φ . Here and in the following $\mathbf{x}_i := (\mathbf{r}_i, \sigma_i)$ denotes the combined spatial and spin coordinate of a particle. Their distinguished role in condensed matter physics is due to the fact that both methods provide the correct asymptotic behaviour for short- and long-range correlations. The latter, however, inevitably lead to certain types of nonlinearities which make these methods considerably more complicated than standard *many-body perturbation theory* (MBPT). Concerning computational complexity, the CC method appears to be better tractable, however for the price of a rather special ansatz for the correlation operator. This excludes e.g. the Jastrow ansatz

$$\mathcal{F}(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = e^{\sum_i u^{(1)}(\mathbf{r}_i) + \sum_{i < j} u^{(2)}(\mathbf{r}_i, \mathbf{r}_j)} \dots \quad (1.2)$$

where the correlation operator simply consists of a symmetric function of the spatial electron coordinates. In this context we have to mention the generalized CC method of Noga and Kutzelnigg [6] which incorporates a spatial function which depends explicitly on the inter-electron distance. Such kind of term describes the wavefunction near inter-electron cusps and improves convergence with respect to the size of the basis set.

Jastrow factors provide the starting point for the FHNC method and are particularly interesting concerning possible applications in *quantum Monte Carlo* (QMC) calculations [7]. Within *variational Monte Carlo* (VMC), the expectation value of the energy is calculated for Jastrow-type wavefunctions using various variants of the Metropolis algorithm. The *diffusion Monte Carlo* (DMC) method enables an exact solution of Schrödinger's equation within the fixed-node approximation. This method requires Jastrow factors as accurate trial wavefunctions for an efficient reduction of the statistical variance of the energy. Herewith it is the short-range behaviour of the Jastrow factor near the coalescence points of particles that gives the dominant contribution. QMC methods enable a compact representation of the k -particle correlation functions $u^{(k)}$ in terms of rational functions of the inter-particle distances [8, 9]. Because of the computational complexity of the FHNC method, it became common practice to use stochastic approaches for the optimization of these Jastrow factors [8, 9, 10, 11, 12, 13]. Either variance-minimization techniques [8, 9, 10, 11] or methods for a direct minimization of the energy [12, 13] were employed. These optimization techniques yield wavefunctions which typically recover between 70% to 95% of the correlation energy with amazingly small numbers of parameters compared to other many-particle methods of similar accuracy. A potential drawback of stochastic approaches, however, is the multitude of local minima that are encountered during the optimization. The resulting Jastrow factors, therefore, typically correspond to local minima which might depend on the initial guess for the parameters. Such kind of ambiguity has only minor effects concerning the total energy of a system, however, it hampers an unbiased calculation of energy differences.

An alternative representation for Jastrow factors can be obtained from wavelet based multiresolution analysis. This approach takes into account the multiscale character of many-particle systems and provides sparse approximations for correlation functions $u^{(k)}$ in terms of hierarchical tensor product wavelets [14, 15, 16, 17] which can be adapted to the specific length- and energy-scales of the system under consideration. Multiscale representations are also of potential interest for QMC calculations because of the local character of wavelet bases. In a previous paper [16], we have studied the computational complexity of wavelet approximations for the local ansatz of Stollhoff and Fulde [18, 19] using diagrammatic techniques from FHNC theory. Within the present work, we consider a more general approach based on standard perturbation theory. It turns out that our approach is closely related to the *coupled cluster perturbation theory* (CCPT) presented in Ref. [2]. Furthermore we have to mention the huge amount of work devoted to linked-cluster expansions for Jastrow-type wavefunctions in nuclear physics during the late sixties and early seventies of the last century, which finally culminated into the development of FHNC theory (see e.g. Ref. [20] for a comprehensive discussion of this subject). We just want to refer to Talman's approximate variational treatment of Jastrow factors [21, 22] that agrees in lowest order with our approach. Except for homogeneous systems, these methods become very costly from the computational point of view. Therefore, standard perturbation theory turns out to be an interesting alternative at the expense of an accurate treatment of long-range correlations. For our envisaged applications in solid state physics, like electrons or

multi excitons confined to a quantum dot, and quantum chemistry this is perfectly justified. Otherwise it is possible to combine our perturbative approach with a recently developed *random phase approximation* (RPA) for inhomogeneous systems by Gaudoin et al. [23] that provides an accurate description of the long-range behaviour of Jastrow factors.

The paper is organized as follows: In Section 2 we present a perturbation analysis of Jastrow factors, where linear and exponential perturbation expansions for correlation factors are derived in Sections 2.1 and 2.2, respectively. Applications to a screened Fermi gas model with periodic boundary conditions are discussed in Section 3. A qualitative study of perturbative Jastrow factors through comparison with FHNC calculations is presented in Section 3.1. The second and third order correlation energies are compared with results from QMC calculations in Section 3.2. Finally, in Section 4 we make some concluding remarks.

2 Perturbation analysis of Jastrow factors

We consider a general product ansatz for the wavefunction

$$\Psi(\mathbf{x}_1, \dots, \mathbf{x}_N) = \mathcal{F}(\mathbf{x}_1, \dots, \mathbf{x}_N) \Phi(\mathbf{x}_1, \dots, \mathbf{x}_N), \quad (2.1)$$

where the correlation factor \mathcal{F} is a symmetric function of the electron coordinates. The corresponding variational problem consists of minimizing the expectation value of the energy

$$E = \min_{\mathcal{F}} \frac{\int d^3x_1 \dots d^3x_N \mathcal{F} \Phi^* (\mathbf{x}_1, \dots, \mathbf{x}_N) H \mathcal{F} \Phi (\mathbf{x}_1, \dots, \mathbf{x}_N)}{\int d^3x_1 \dots d^3x_N \mathcal{F} \Phi^* (\mathbf{x}_1, \dots, \mathbf{x}_N) \mathcal{F} \Phi (\mathbf{x}_1, \dots, \mathbf{x}_N)}, \quad (2.2)$$

with respect to the correlation factor \mathcal{F} for a fixed reference wavefunction Φ . Two different perturbation scheme for this variational problem are discussed below. The first scheme utilizes a linear representation for the correlation factor, whereas the second scheme is based on the exponential Jastrow-type ansatz (1.2). Both schemes employ symmetric many-particle basis functions of the form

$$\begin{aligned} \mathcal{U}^{(0)}(\mathbf{X}) &= 1, \\ \mathcal{U}_{\alpha}^{(1)}(\mathbf{X}) &= \sum_i u_{\alpha}^{(1)}(\mathbf{x}_i), \\ \mathcal{U}_{\beta}^{(2)}(\mathbf{X}) &= \frac{1}{2} \sum_{i \neq j} u_{\beta}^{(2)}(\mathbf{x}_i, \mathbf{x}_j), \\ &\vdots \end{aligned} \quad (2.3)$$

where $u_{\alpha}^{(k)}(\mathbf{x}_{i_1}, \mathbf{x}_{i_2}, \dots, \mathbf{x}_{i_k})$ denote symmetric k -particle basis functions with indices $\alpha \in \Lambda_k$ taken from conveniently defined index sets. In the following we refer to k as the *degree* of the basis functions. For notational convenience we have introduced the short-hand notation $\mathbf{X} := (\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N)$ to represent the combined vector of all electron coordinates. The computational complexity increases substantially with the degree of the basis functions. Therefore it is essential to truncate the basis at a certain degree. It turns out that both perturbation schemes discussed below provide such truncations in a natural way.

Obviously there are certain redundancies in our many-particle basis. According to definition (2.3), one-particle basis functions can be expressed e.g. in terms of two-particle functions etc., which means that the basis is overcomplete without further restrictions. Such kind of restrictions can be imposed in various ways. For example it is possible to derive a suitable basis from multiresolution analysis. Neglecting spin degrees of freedom, a many-particle basis can be defined via wavelet tensor products

$$\begin{aligned} u_{\alpha}^{(1)}(\mathbf{r}_i) &= \gamma_{\alpha}(\mathbf{r}_i), \\ u_{\alpha, \beta}^{(2)}(\mathbf{r}_i, \mathbf{r}_j) &= \gamma_{\alpha}(\mathbf{r}_i) \gamma_{\beta}(\mathbf{r}_j) + \gamma_{\alpha}(\mathbf{r}_j) \gamma_{\beta}(\mathbf{r}_i), \\ &\vdots \end{aligned} \quad (2.4)$$

where a k -particle basis function corresponds to a k -fold tensor product of 3d-wavelets γ_α . The multivariate wavelets γ_α themselves consist of three-fold mixed tensor products of univariate wavelets and scaling functions, at which each tensor product contains at least one univariate wavelet. For further details and applications, we refer to our previous publication [14, 16]. By definition these 3d-wavelets span so called homogeneous function spaces [24] that do not contain constant functions. Thereby, this ansatz largely avoids possible redundancies within the many-particle basis. Wavelet tensor products enable an adaptive approximation of electron correlations [14], where the size of the k -particle basis increases almost linearly i.e. $O(M \log(M)^{k-1})$ with the number M of 3d-wavelets. Furthermore, tensor product structures considerably simplify the evaluation of matrix elements [16, 25].

Second quantization provides another possibility to impose further restrictions on the many-particle basis (2.3). It has been suggested by Stollhoff and Fulde [18, 19] to take only the normal ordered part of the basis functions (2.3) which removes all contributions with degree $< k$ from $\mathcal{U}_\alpha^{(k)}$. Furthermore, second quantization introduces additional flexibility into the perturbation analysis. This can be used to reduce the computational complexity which is essential for practical applications. In order to illustrate our assertion, we consider an arbitrary two-particle basis function as an operator in second quantization

$$\mathcal{U}_\alpha^{(2)} \equiv \hat{U}_\alpha^{(2)} = \frac{1}{2} \sum_{pqrs} \langle pq | u_\alpha^{(2)} | rs \rangle c_p^\dagger c_q^\dagger c_s c_r, \quad (2.5)$$

where c_p^\dagger, c_s denote creation and annihilation operators for the corresponding orbitals φ_p, φ_s of a single-particle Hamiltonian H_0 . In the following, we denote virtual orbitals by a, b, \dots , occupied orbitals by i, j, \dots and arbitrary orbitals by p, q, \dots . Using standard diagrammatic notation [26] we can express two-particle operators in terms of Goldstone diagrams. These diagrams are depicted in Fig. 2.1, where horizontal dashed lines represent two-particle integrals $\langle pq | u_\alpha^{(2)} | rs \rangle$ and upward and downward pointing solid lines denote “particle” (virtual orbitals) and “hole” states (occupied orbitals), respectively. We want to mention that virtual orbitals $\{\varphi_a\}$ are introduced for purely formal reasons and that for diagrammatic calculations only occupied orbitals $\{\varphi_i\}$ are required. This is due to the fact that the underlying basis (2.3) consists of simple functions which allows us to use the identity

$$\sum_a \varphi_a(\mathbf{x}_1) \varphi_a^*(\mathbf{x}_2) = \delta(\mathbf{r}_1 - \mathbf{r}_2) \delta_{\sigma_1, \sigma_2} - \sum_i \varphi_i(\mathbf{x}_1) \varphi_i^*(\mathbf{x}_2), \quad (2.6)$$

we refer to Ref. [16] for further details. The normal ordered part of the operator (2.5) comprises the diagrams 1 to 10. According to the original suggestion of Stollhoff and Fulde [18] only these diagrams should be taken into account. The remaining diagrams 11 to 18 represent one-particle operators, however, diagrams 11 to 14 are intrinsically nonlocal and are therefore not represented by basis functions of degree < 2 . To keep all nonlocal diagrams preserves commutativity of the many-particle basis, which turns out to be essential for the exponential perturbation scheme. Furthermore it has the advantage that it is directly applicable in QMC calculations where only local functions can be used to represent the Jastrow factor. The new local basis functions, corresponding to diagrams 1 to 14, are given by

$$\tilde{\mathcal{U}}_\alpha^{(2)}(\mathbf{X}) = \mathcal{U}_\alpha^{(2)}(\mathbf{X}) - \sum_i \int d^3x' u_\alpha^{(2)}(\mathbf{x}_i, \mathbf{x}') \rho(\mathbf{x}') - \frac{1}{2} \sum_{i,j} \left(\langle ij | u_\alpha^{(2)} | ij \rangle - \langle ij | u_\alpha^{(2)} | ji \rangle \right) \quad (2.7)$$

where $\rho(\mathbf{x})$ is the spin density of the reference wavefunction Φ . A more radical approach in the spirit of CC theory, is to keep only the 10th diagram, where the corresponding cluster-type operator

$$\hat{U}_\alpha^{(2,c)} := \frac{1}{2} \sum_{abij} \langle ab | u_\alpha^{(2)} | ij \rangle c_a^\dagger c_b^\dagger c_j c_i, \quad (2.8)$$

is of nonlocal character and cannot be immediately applied in QMC calculations. It turns out that cluster operators significantly reduce the computational complexity at higher orders of perturbation theory.

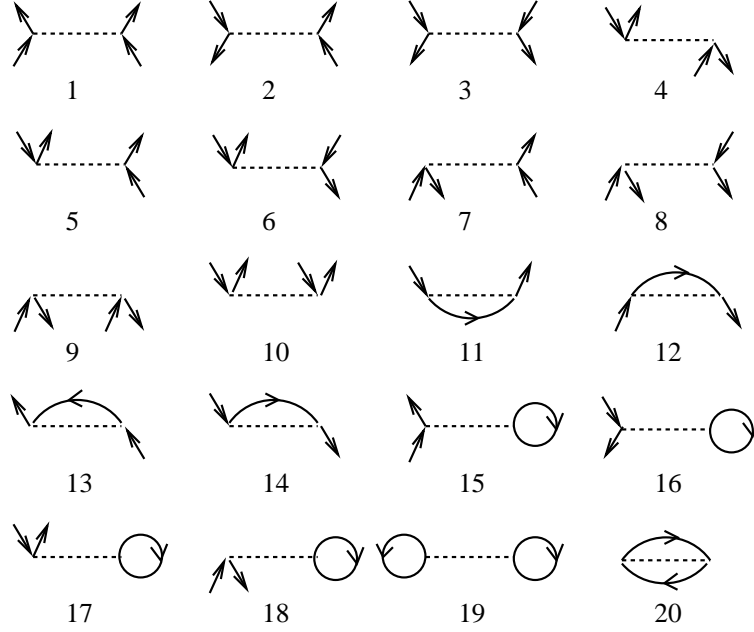


Figure 2.1: Goldstone diagrams representing a symmetric two-particle basis function $\mathcal{U}_\alpha^{(2)}$.

2.1 Linear perturbation expansion of the correlation factor

It is instructive to consider first in second quantization a linear expansion of the correlation factor

$$\hat{\mathcal{F}} = \sum_{k=0}^N \sum_{\alpha \in \Lambda_k} a_{k\alpha} \hat{U}_\alpha^{(k)}, \quad (2.9)$$

where e.g. $\hat{U}_\alpha^{(2)}$ can be represented by any subset of the diagrams in Fig. 2.1. Variation of the Rayleigh-quotient (2.2) with respect to the coefficients $a_{k\alpha}$ leads to a generalized eigenvalue problem

$$\langle \hat{U}_\alpha^{(k)\dagger} \hat{H} \hat{\mathcal{F}} \rangle = E \langle \hat{U}_\alpha^{(k)\dagger} \hat{\mathcal{F}} \rangle, \quad (2.10)$$

where we use here and in the following $\langle \hat{O} \rangle$ to denote $\int d^3x_1 \dots d^3x_N \Phi^*(\mathbf{x}_1, \dots, \mathbf{x}_N) \hat{O} \Phi(\mathbf{x}_1, \dots, \mathbf{x}_N)$. The eigenvalue problem (2.10) determines the correlated wave function $\hat{\mathcal{F}}\Phi$ up to a normalization constant. In order to fix this constant we have chosen the intermediate normalization condition

$$\langle \hat{\mathcal{F}} \rangle = 1, \quad (2.11)$$

which is frequently used in standard MBPT [27].

A basic drawback of the variational treatment of the linear ansatz (2.9) is that there exists no simple size-consistent truncation scheme with respect to the degree of the basis functions (2.3). However, such a scheme exists for a perturbative treatment of Eq. (2.10). According to a standard procedure in perturbation theory, we split up the Hamiltonian

$$\hat{H}(\lambda) = \hat{H}_0 + \lambda \hat{W}, \quad (2.12)$$

into a one-particle operator \hat{H}_0 and a two-particle operator \hat{W} times the coupling constant λ , where the reference wavefunction Φ solves the ground state eigenvalue problem

$$\hat{H}_0 \Phi = E_0 \Phi. \quad (2.13)$$

Furthermore, we assume that there exist power series expansions with respect to the coupling constant λ for the energy

$$E(\lambda) = E_0 + \lambda E_1 + \lambda^2 E_2 + \dots, \quad (2.14)$$

and the correlation factor

$$\hat{\mathcal{F}}(\lambda) = \hat{F}_0 + \lambda \hat{F}_1 + \lambda^2 \hat{F}_2 + \dots. \quad (2.15)$$

It follows immediately from the intermediate normalization condition (2.11) that

$$\hat{F}_0 = 1, \text{ and } \langle \hat{F}_m \rangle = 0 \text{ for } m \geq 1. \quad (2.16)$$

Actually, $\hat{\mathcal{F}}$ depends on λ , according to Eq. (2.9), via its coefficients $a_{k\alpha}(\lambda)$. These coefficients have formal expansions

$$a_{k\alpha}(\lambda) = \sum_m a_{k\alpha}^{(m)} \lambda^m, \quad (2.17)$$

so that the m 'th order correlation factor \hat{F}_m in Eq. 2.15 can be written as

$$\hat{F}_m = \sum_{k=0}^N \sum_{\alpha \in \Lambda_k} a_{k\alpha}^{(m)} \hat{U}_\alpha^{(k)}. \quad (2.18)$$

Inserting Eqs. (2.12), (2.14) into Eq. (2.10) and comparing different powers of λ , we obtain perturbation equations for the correlation factor

$$\langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{F}_m \rangle = -\langle \hat{U}_\alpha^{(k)\dagger} \hat{W} \hat{F}_{m-1} \rangle + \sum_{p=1}^m E_p \langle \hat{U}_\alpha^{(k)\dagger} \hat{F}_{m-p} \rangle, \quad m = 1, 2, \dots \quad (2.19)$$

These equations resemble to Galerkin discretizations of continuous perturbation equations already mentioned by Møller and Plesset [28]. However we want to remind the reader that our test basis $\mathcal{U}_\alpha(\mathbf{X})\Phi$ is generically incomplete in the space of many-particle wavefunctions. To a certain extent our approach resembles to the fixed-node approximation in QMC calculations, because all of our test basis functions in the Galerkin scheme share the nodes of the reference wavefunction Φ . This important aspect should be beared in mind in the following, when we obtain expressions which appear similar to standard perturbation theory. In this respect our approach differs from the Gaussian geminal method described in Refs. [29, 30], where highly accurate solutions of the continuous perturbation equations have been considered following Sinanoğlu's work [31, 32].

The underlying variational formulation for correlation factors (2.2) is obviously size-consistent. Therefore, size-consistency is satisfied for each order of perturbation theory separately. Furthermore, the sequence of perturbation equations (2.19) implies a truncation scheme with respect to the degree of the many-particle basis (2.3), which has been already recognized by Sinanoğlu [32]. For example, a first order correlation factor \hat{F}_1 contains contributions only from one- and two-particle normal ordered or cluster-type operators $\hat{U}_\alpha^{(k)}$.

2.2 Perturbation expansion of the Jastrow ansatz

The linear expansion of the correlation factor is not convenient for further usage in QMC calculations. Although the perturbation energies are size consistent this is not the case any more for a variational treatment of the perturbative wavefunction. It is therefore natural to consider a perturbation theory for the exponential Jastrow ansatz

$$\hat{\mathcal{F}} = e^{\hat{\tau}} \text{ with } \hat{\tau} = \sum_{k=0}^K \sum_{\alpha \in \Lambda_k} a_{k\alpha} \hat{U}_\alpha^{(k)}, \quad (2.20)$$

where we can truncate the expansion with respect to k at any order $K \leq N$ without violating size-consistency for the variational energy (2.2). It is well known that the two-particle terms $\hat{U}_\alpha^{(2)}$ give the dominant contribution to the correlation energy, cf. Ref. [33] for a discussion of higher order terms. The exponential ansatz incorporates already a lot of essential physics and therefore considerably reduces the number of degrees of freedom that have to be taken into account. Variation with respect to the coefficients $a_{k\alpha}$ leads to a nonlinear eigenvalue problem

$$\langle \hat{U}_\alpha^{(k)\dagger} e^{\hat{\tau}^\dagger} \hat{H} e^{\hat{\tau}} \rangle = E \langle \hat{U}_\alpha^{(k)\dagger} e^{\hat{\tau}^\dagger} e^{\hat{\tau}} \rangle, \quad (2.21)$$

where we have assumed that the operators $\hat{U}_\alpha^{(k)}$ commute with each other. This is the case for simple functions as well as for cluster-type operators. According to our previous discussion, cluster-type operators are much more favourable concerning the diagrammatic evaluation of matrix elements. On a first glance, the exponential Jastrow ansatz (2.20) resembles closely to the CC approach [3]. There is however an essential difference inasmuch as the underlying function basis $\mathcal{U}_\alpha^{(k)}$ does not guaranty convergence of the product ansatz (2.1) to the exact wavefunction in the complete basis set limit. Here completeness has to be considered in an appropriate function space to which the $\mathcal{U}_\alpha^{(k)}$ belong. This shortcoming of the Jastrow ansatz manifests itself in the fixed-node error of DMC calculations. For that reason we take the variational energy (2.2) as the starting point for our perturbation analysis instead of the CC energy and projection equations. The latter assume an exact ansatz for the wavefunction and provide the basis for the CCPT presented in Ref. [2].

For the perturbation analysis, we assume again a λ dependent Hamiltonian (2.12) which leads to power series expansions for the energy (2.14) and correlation operator

$$\hat{\tau}(\lambda) = \hat{\tau}_0 + \hat{\tau}_1 \lambda + \hat{\tau}_2 \lambda^2 + \dots + \hat{\tau}_m \lambda^m + \dots \quad (2.22)$$

Analogously to Eq. (2.9), different orders of the correlation operator are represented through expansions in the operator basis

$$\hat{\tau}_m = \sum_{k=0}^K \sum_{\alpha \in \Lambda_k} a_{k\alpha}^{(m)} \hat{U}_\alpha^{(k)}, \quad (2.23)$$

which can be truncated at any order $K \leq N$. In order to derive the perturbation equations for $\hat{\tau}_m$, we expand the exponential ansatz in powers of λ .

$$e^{\hat{\tau}(\lambda)} = \hat{F}_0 + \hat{F}_1 \lambda + \hat{F}_2 \lambda^2 + \dots + \hat{F}_m \lambda^m + \dots, \quad (2.24)$$

where the constant term $\hat{F}_0 = 1$ ($\hat{\tau}_0 = 0$) is fixed by the intermediate normalisation condition (2.11) and the next lowest order terms are given by

$$\begin{aligned} \hat{F}_1 &= \hat{\tau}_1, \\ \hat{F}_2 &= \hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2, \\ \hat{F}_3 &= \hat{\tau}_3 + \hat{\tau}_2 \hat{\tau}_1 + \frac{1}{3!} \hat{\tau}_1^3, \\ \hat{F}_4 &= \hat{\tau}_4 + \hat{\tau}_3 \hat{\tau}_1 + \frac{1}{2} \hat{\tau}_2^2 + \frac{1}{2} \hat{\tau}_2 \hat{\tau}_1^2 + \frac{1}{4!} \hat{\tau}_1^4, \\ &\vdots \end{aligned}$$

Perturbation equations for the Jastrow ansatz can be obtained by inserting Eqs. (2.14), (2.24) into the eigenvalue problem (2.21) and comparing the coefficients for fixed powers of λ . In the following, we refer to this method as *Jastrow perturbation theory* (JPT). The first order equation for the correlation operator $\hat{\tau}_1$ is given by

$$\langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{\tau}_1 \rangle = -\langle \hat{U}_\alpha^{(k)\dagger} (\hat{W} - E_1) \rangle, \quad (2.25)$$

from which we obtain the second order energy (JPT2)

$$E_2 = \langle \hat{W} \hat{\tau}_1 \rangle, \quad (2.26)$$

with $E_1 = \langle \hat{W} \rangle$ and $\langle \hat{\tau}_1 \rangle = \langle \hat{F}_1 \rangle = 0$ as before. For higher orders, we have the following general equation

$$\begin{aligned} \langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{F}_m \rangle &= - \sum_{p=1}^{m-1} \langle \hat{U}_\alpha^{(k)\dagger} \hat{F}_p^\dagger (\hat{H}_0 - E_0) \hat{F}_{m-p} \rangle - \sum_{p=0}^{m-1} \langle \hat{U}_\alpha^{(k)\dagger} \hat{F}_p^\dagger \hat{W} \hat{F}_{m-p-1} \rangle \\ &\quad + \sum_{p=1}^m E_p \sum_{q=0}^{m-p} \langle \hat{U}_\alpha^{(k)\dagger} \hat{F}_q^\dagger \hat{F}_{m-p-q} \rangle. \end{aligned} \quad (2.27)$$

From this, we get the second order equation

$$\begin{aligned} \langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{\tau}_2 \rangle &= - \langle \hat{U}_\alpha^{(k)\dagger} (\hat{W} - E_1) \hat{\tau}_1 \rangle - \frac{1}{2} \langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{\tau}_1^2 \rangle \\ &\quad - \langle \hat{U}_\alpha^{(k)\dagger} \hat{\tau}_1^\dagger (\hat{H}_0 - E_0) \hat{\tau}_1 \rangle - \langle \hat{U}_\alpha^{(k)\dagger} \hat{\tau}_1^\dagger (\hat{W} - E_1) \rangle + E_2 \langle \hat{U}_\alpha^{(k)\dagger} \rangle, \end{aligned} \quad (2.28)$$

and third order energy (JPT3)

$$\begin{aligned} E_3 &= \frac{1}{2} \langle \hat{\tau}_1^\dagger (\hat{W} - E_1) \hat{\tau}_1 \rangle + \langle \hat{\tau}_1^\dagger (\hat{H}_0 - E_0) (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2) \rangle + \langle (\hat{W} - E_1) (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2) \rangle + \text{c.c.} \\ &= \frac{1}{2} \langle \hat{\tau}_1^\dagger (\hat{W} - E_1) \hat{\tau}_1 \rangle + \frac{1}{2} \langle \hat{\tau}_1^\dagger (\hat{H}_0 - E_0) \hat{\tau}_1^2 \rangle + \frac{1}{2} \langle (\hat{W} - E_1) \hat{\tau}_1^2 \rangle + \text{c.c.}, \end{aligned} \quad (2.29)$$

where we have used the relation

$$\langle \hat{\tau}_1^\dagger (\hat{H}_0 - E_0) \hat{\tau}_2 \rangle = - \langle (\hat{W} - E_1) \hat{\tau}_2 \rangle \quad (2.30)$$

which can be easily derived from Eq. (2.25). In accordance with Wigner's $2m+1$ rule [2], the third order energy requires only the first order correction of the wavefunction. Furthermore with the second order wavefunction we obtain an expression for the fourth order energy (JPT4)

$$\begin{aligned} E_4 &= \frac{1}{2} \langle (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2)^\dagger (\hat{H}_0 - E_0) (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2) \rangle + \langle \hat{\tau}_1^\dagger (\hat{W} - E_1) (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2) \rangle - E_2 \frac{1}{2} \langle \hat{\tau}_1^\dagger \hat{\tau}_1 \rangle \\ &\quad + \langle \hat{\tau}_1^\dagger (\hat{H}_0 - E_0) (\hat{\tau}_2 \hat{\tau}_1 + \frac{1}{3!} \hat{\tau}_1^3) \rangle + \langle (\hat{W} - E_1) (\hat{\tau}_2 \hat{\tau}_1 + \frac{1}{3!} \hat{\tau}_1^3) \rangle + \text{c.c.} \end{aligned} \quad (2.31)$$

If a cluster-type basis is used, further simplifications of the second order Eq. (2.28) as well as of third and fourth order energies (2.29), (2.31) can be achieved, provided that the operator basis is closed with respect to multiplication up to degree two i.e.

$$\hat{U}_\alpha^{(1)} \hat{U}_\beta^{(1)} = \sum_\gamma c_\gamma \hat{U}_\gamma^{(2)}. \quad (2.32)$$

Under this assumption, we can make use of the relation

$$\langle \hat{U}_\alpha^{(k_1)\dagger} \hat{U}_\beta^{(k_2)\dagger} \hat{U}_\lambda^{(k_3)\dagger} (\hat{H}_0 - E_0) \hat{\tau}_1 \rangle = - \langle \hat{U}_\alpha^{(k_1)\dagger} \hat{U}_\beta^{(k_2)\dagger} \hat{U}_\lambda^{(k_3)\dagger} (\hat{W} - E_1) \rangle. \quad (2.33)$$

For an operator product $\hat{U}_\alpha^{(k_1)\dagger} \hat{U}_\beta^{(k_2)\dagger} \hat{U}_\lambda^{(k_3)\dagger}$ of degree $k_1 + k_2 + k_3 > 2$ both sides of the relation vanish since it can not be fully contracted by $(\hat{H}_0 - E_0) \hat{\tau}_1$ and \hat{W} respectively. In the remaining cases, the relation (2.33) either directly corresponds to the first order Eq. (2.25), or can be reduced to it by applying the closedness assumption (2.32). Using relation (2.33) for a cluster-type operator basis which satisfies the closedness condition (2.32), the Eqs. (2.28), (2.29), and (2.31) reduce to

$$\langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{\tau}_2 \rangle = - \langle \hat{U}_\alpha^{(k)\dagger} (\hat{W} - E_1) \hat{\tau}_1 \rangle - \frac{1}{2} \langle \hat{U}_\alpha^{(k)\dagger} (\hat{H}_0 - E_0) \hat{\tau}_1^2 \rangle + E_2 \langle \hat{U}_\alpha^{(k)\dagger} \rangle \quad (2.34)$$

$$E_3 = \langle \hat{\tau}_1^\dagger (\hat{W} - E_1) \hat{\tau}_1 \rangle, \quad (2.35)$$

$$E_4 = \frac{1}{2} \langle (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2)^\dagger (\hat{H}_0 - E_0) (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2) \rangle + \langle \hat{\tau}_1^\dagger (\hat{W} - E_1) (\hat{\tau}_2 + \frac{1}{2} \hat{\tau}_1^2) \rangle - \frac{1}{2} E_2 \langle \hat{\tau}_1^\dagger \hat{\tau}_1 \rangle + \text{c.c.} \quad (2.36)$$

We want to close this section with a remark concerning the intimate relation between JPT3 and the local ansatz of Stollhoff and Fulde [18]. In Ref. [16], we have discussed the local ansatz in its simplest variant namely for two-particle normal ordered operators only. Concerning the local ansatz these operators are equivalent to cluster-type operators (2.8). In our present notation, the local ansatz corresponds to the linear equation

$$\langle \hat{U}_\alpha^{(2)\dagger} (\hat{H}_0 + \hat{W}) \hat{\tau} \rangle - (E_0 + E_1) \langle \hat{U}_\alpha^{(2)\dagger} \hat{\tau} \rangle = -\langle \hat{U}_\alpha^{(2)\dagger} \hat{W} \rangle, \quad (2.37)$$

where the correlation energy is given by

$$E_{\text{corr}} = \langle \hat{W} \hat{\tau} \rangle. \quad (2.38)$$

It is easy to see that a perturbative treatment of the local ansatz using Eqs. (2.12), (2.14), and (2.22) reproduces the first and second order Eqs. (2.25) and (2.34) as well as the second and third order energy expressions (2.26) and (2.35).

3 Perturbative Jastrow factors for a homogeneous Fermi gas

In order to test the accuracy of the JPT approach for short-range correlations, we have studied a homogeneous spin unpolarized Fermi gas at various densities. Because of the well known shortcoming of finite order perturbation theory for the homogeneous electron gas, we have chosen, instead of the bare Coulomb potential, a Yukawa potential

$$v(\mathbf{r}) = \frac{e^{-\mu|\mathbf{r}|}}{|\mathbf{r}|}, \quad (3.1)$$

with screening parameter μ . To be consistent with QMC methods, the calculations have been performed for supercell models with periodic boundary conditions [7, 34]. The supercell Hamiltonian is of the form

$$\hat{H} = -\frac{1}{2} \sum_{i=1}^N \Delta_i + \frac{1}{2} \sum_{\mathbf{R}} \sum_{i \neq j} v(\mathbf{r}_i - \mathbf{r}_j + \mathbf{R}), \quad (3.2)$$

where the periodic interaction potential has been taken as a sum over all lattice vectors \mathbf{R} of the supercell. We have considered two different decompositions of the Hamiltonian $\hat{H} = \hat{H}_0 + \hat{W}$, where \hat{H}_0 either corresponds to the bare kinetic energy or to the Fock operator. For homogeneous systems this makes only a minor difference because the zeroth order wavefunction Φ is already fixed by translational symmetry. For small values of the screening parameter ($\mu \leq 1 \text{ bohr}^{-1}$), the Fock operator yields more accurate correlation energies and pair-correlation functions. It turns out, however, that with increasing μ , this difference becomes marginal. In the following, we present results only for the Fock operator.

Because of translational symmetry, the first order Jastrow factor requires only two-particle basis functions. Adapted to periodic boundary conditions and translational symmetry, we have chosen two-particle basis functions of the form

$$u_\alpha^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{\mathbf{R}} g_\alpha(\mathbf{r}_1 - \mathbf{r}_2 + \mathbf{R}) \quad \text{with } g_\alpha(\mathbf{r}) = \exp(-\xi_\alpha \mathbf{r}^2), \quad (3.3)$$

where the exponents of the Gaussian basis set have been taken from an even-tempered sequence $\xi_\alpha = a_0 b^{\alpha-1}$, $\alpha = 1, \dots, n$, with appropriately chosen parameters $a_0 = 0.03$, $b = 3$ and $n = 6$. Expressed in second quantization, this basis can be modified by removing the constant diagrams 19, and 20 from Fig. 2.1 in order to satisfy the intermediate normalization condition $\langle \tau_1 \rangle = 0$. Actually, only diagram 10 in Fig. 2.1 contributes to the first order equation (2.25). It makes no difference with respect to the coefficients $a_{2\alpha}^{(1)}$ and therefore for the JPT2 and JPT3 correlation energies, cf. Eqs. (2.26) and (2.29),

whether local basis functions (2.7), or cluster-type operators (2.8) have been chosen as a basis set. For higher orders in perturbation theory the two basis sets are not equivalent any more.

The matrix elements of the first order Eq. (2.25) require the calculation of the Goldstone diagrams shown in Fig. 3.1, where the symbol $-X$ denotes insertion of the Fock operator. With the first order Jastrow factor in hands, one can also calculate the second and third order contributions to the correlation energy. These contributions are expressed in terms of Goldstone diagrams in Figs. 3.2 and 3.3, where dashed lines represent the first order correlation operator τ_1 and wavy lines represent the interaction potential (3.1). For the third order contribution, we have assumed that $\hat{W} - E_1$ is normal ordered, which is actually the case if H_0 corresponds to the Fock operator. The computational complexity of third order diagrams for wavelet bases has been discussed in detail in Ref. [16].

3.1 Comparison with FHNC Jastrow factors

In order to judge the quality of first-order Jastrow factors, we have performed FHNC//0 calculations [4, 5] for the homogeneous Fermi gas model (3.1). The FHNC//0 calculations represent the thermodynamic limit of an infinitely large supercell. Due to the finite size of the supercell in our JPT calculations, Jastrow factors possess a periodic structure with a small anisotropy due to the lattice sum (3.3) over neighbouring supercells. In Fig. 3.4, we present first order pair-correlation functions along the (100) direction for different supercell size at density $r_s = 2$ and $\mu = 1 \text{ bohr}^{-1}$, together with the corresponding FHNC//0 result. For short-range correlations, a good agreement between first order approximations and the FHNC//0 Jastrow factor can be observed already for rather small supercells. With increasing size of the supercell, first order JPT converges amazingly well towards the FHNC//0 result even at intermediate distances where a good agreement cannot be taken for granted.

Since our focus is on short-range correlations, we have chosen in the following a fixed supercell with $N = 54$ particles. Different values of the screening parameter μ , at fixed density $r_s = 2$, have been considered in Fig. 3.5 a). It turns out that there is a close agreement between first order JPT and FHNC//0 pair-correlation functions for the whole range of parameters. To avoid computational artefacts we have plotted the JPT pair-correlation functions only up to 0.1 bohr because our Gaussian approximation (3.3) does not provide a resolution of the cusp beyond this scale. For comparison, Fig. 3.5 b) shows the behaviour at different densities and fixed $\mu = 1 \text{ bohr}^{-1}$. From this figure we observe a discrepancy between first order JPT and FHNC//0 pair-correlation functions at low densities and intermediate inter-particle distances $\geq 1 \text{ bohr}$. This is because in the low density regime correlations are getting stronger and perturbation theory in general becomes less accurate.

In our previous calculations, we have not distinguished between pairs with parallel and anti-parallel spins. By a slight modification of the formalism, however, it is possible to calculate spin dependent Jastrow factors. The resulting first order pair-correlation functions for $\mu = 1 \text{ bohr}^{-1}$, $r_s = 2$ and $N = 54$ are shown in Fig. 3.6. It is interesting to compare the slopes at short inter-particle distances with Kato's cusp condition [35, 36]. For this, we have used a slightly enlarged basis containing 14 even-tempered Gaussians. The value of the averaged slope in the interval (0.1, 0.2) is 0.23 and 0.46 for parallel and anti-parallel spins, respectively. This compares favourably with Kato's cusp condition which requires spherically averaged derivatives of 0.25 and 0.5 at the inter-particle cusp.

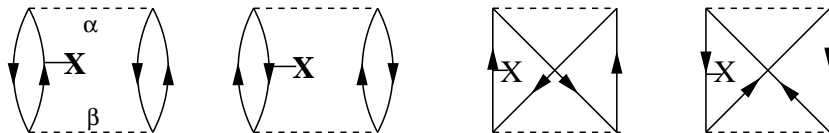


Figure 3.1: Diagrams required for the calculation of matrix elements $\langle \hat{U}_\alpha^{(2)\dagger} (\hat{H}_0 - E_0) \hat{U}_\beta^{(2)} \rangle$.

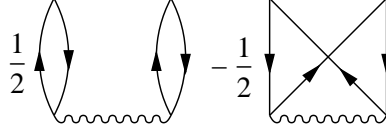


Figure 3.2: Diagrammatic representation of $E_2 = \langle \hat{W} \hat{\tau}_1 \rangle$

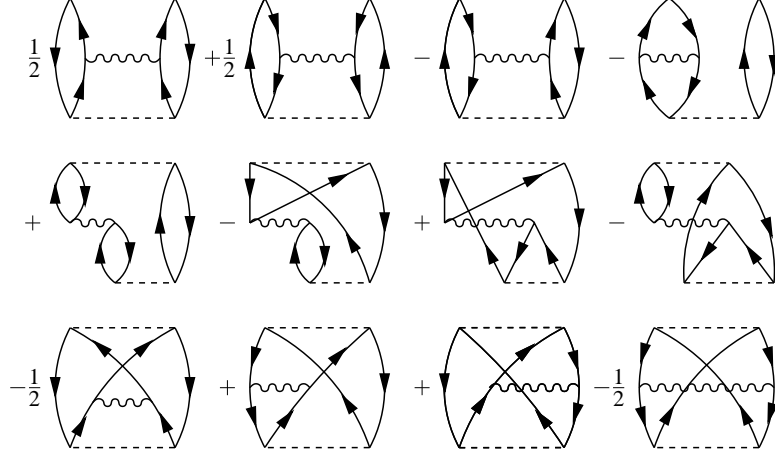


Figure 3.3: Diagrammatic representation of $E_3 = \langle \hat{\tau}_1^\dagger (\hat{W} - E_1) \hat{\tau}_1 \rangle$

3.2 Correlation energies

With first order Jastrow factors at hand, we have calculated JPT2 and JPT3 correlation energies for the homogeneous Fermi gas model. Due to momentum conservation, the diagrams with two external lines in Fig. 2.1 do not contribute to Eq. (2.25). Therefore we can immediately identify correlation operators $\hat{\tau}_1$ with ordinary two-particle correlation functions in Jastrow factors. In order to judge the quality of our results, we have performed VMC and *pure diffusion quantum Monte Carlo* (PDMC) calculations [37, 38] for the Hamiltonian (3.2) using these Jastrow factors as trial wavefunctions. The VMC method directly calculates the energy expectation value (2.2) for a given trial wavefunction and therefore provides a measure for the quality of the Jastrow factors. Despite small fixed node errors, PDMC provides fairly accurate benchmark values for the correlation energy of a homogeneous Fermi gas. Furthermore, we compare with standard *second order Møller Plesset* (MP2) perturbation theory [27], which can be expressed as a sum over momentum vectors \mathbf{k}_p and corresponding eigenvalues ϵ_p of spatial HF orbitals

$$E^{(2)} = \sum_{ijab} \frac{2\langle ij||ab\rangle\langle ab||ij\rangle - \langle ij||ab\rangle\langle ab||ji\rangle}{\epsilon_i + \epsilon_j - \epsilon_a - \epsilon_b}, \quad (3.4)$$

$$\langle ij||ab\rangle = \frac{1}{\Omega_s} \frac{4\pi}{(\mathbf{k}_a - \mathbf{k}_i)^2 + \mu^2} \delta_{\mathbf{k}_i + \mathbf{k}_j, \mathbf{k}_a + \mathbf{k}_b}, \quad (3.5)$$

where Ω_s denotes the volume of the supercell and indices i, j, a, b have the same meaning as in Section 2.

According to our discussion in Section 3.1, we first consider the size dependence of JPT2, MP2, VMC, and PDMC correlation energies. These are shown in Fig. 3.7 for different size of the supercell in the case of a homogeneous Fermi gas at density $r_s = 2$ and $\mu = 1$ bohr⁻¹. The JPT2 and VMC calculations have been performed for spin independent and spin dependent (JPT2*, VMC*) Jastrow factors. It can be seen that the different methods yield almost parallel curves, where the correlations energies turned out to be fairly stable already for a small number of particles ($N \geq 54$). Both JPT2 and JPT2* overestimate the correlation energy by roughly 9% and 14%, respectively. By comparison with the corresponding VMC calculations, we observed that the first order perturbative Jastrow factors are actually significantly better

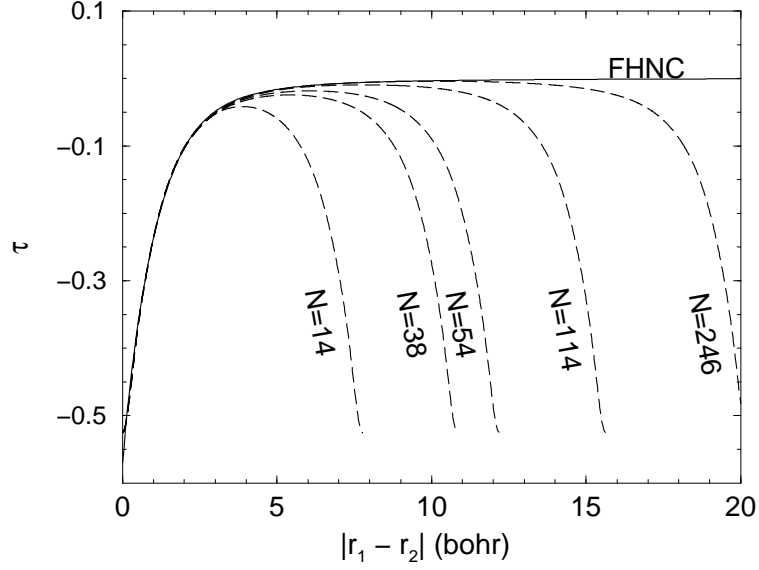


Figure 3.4: First order JPT pair-correlation functions (dashed lines) for a homogeneous Fermi gas at density $r_s = 2$ and screening parameter $\mu = 1 \text{ bohr}^{-1}$ for different size of the supercell. For comparison, the FHNC//0 result for the infinite system (solid line) is shown.

and recover 93% and 96% of the PDMC correlation energy, respectively. These results have to be compared with standard MP2 calculations which overestimate the correlation energy by about 28%.

In order to study the homogeneous Fermi gas model for a larger range of densities and screening parameters, we restricted ourselves to a fixed supercell size with $N = 54$ particles. Since we compare our results with PDMC benchmark calculations for the same supercell size, possible finite-size errors can be ignored. Correlation energies for a homogeneous Fermi gas at density $r_s = 2$ and different screening parameters μ are listed in Table 3.1. It can be seen that JPT2 overestimates whereas JPT3 underestimates the correlation energy. The JPT3 results are in better agreement with VMC calculations than the corresponding JPT2 energies. Except for very small values $\mu < 1 \text{ bohr}^{-1}$, the relative errors of JPT3 and VMC calculations remain almost constant with respect to variations of the screening parameter as it is shown in Fig. 3.8 (a) for spin-dependent Jastrow factors. In contrast to this, the JPT2 relative errors decrease with increasing values for the screening parameter, which has to be expected because short-range correlations become increasingly important. These observations once again suggest that the first-order Jastrow factor is more accurate than the corresponding perturbative correlation energies. A similar behaviour has been observed for a fixed screening parameter $\mu = 1 \text{ bohr}^{-1}$ and different densities which can be seen from Table 3.2 and Fig. 3.8 (b).

It turns out that the second order correlation energies for $r_s = 2$ and $\mu = 0.5, 1, 2, 4 \text{ bohr}^{-1}$ are quite close to those for $\mu = 1 \text{ bohr}^{-1}$ and $r_s = 1, 2, 4, 8$, respectively. Actually they should be exactly the same if \hat{H}_0 contains only the kinetic energy. By rescaling the electron coordinates $\mathbf{r}'_i = \mathbf{r}_i/r_s$ for $i = 1, \dots, N$ it can be seen that r_s serves as a coupling constant in the Schrödinger equation

$$\left[-\frac{1}{2} \sum_{i=1}^N \Delta'_i + r_s \sum_{i<j} \frac{e^{-\mu r_s |\mathbf{r}'_i - \mathbf{r}'_j|}}{|\mathbf{r}'_i - \mathbf{r}'_j|} \right] \Psi = r_s^2 E \Psi = E' \Psi. \quad (3.6)$$

It follows from the perturbation series for the energy

$$E' = E'_0 + r_s E'_1 + r_s^2 E'_2 + \dots \longrightarrow E = E'_0/r_s^2 + E'_1/r_s + E'_2 + \dots \quad (3.7)$$

that the second order correlation energy depends only on the product μr_s .

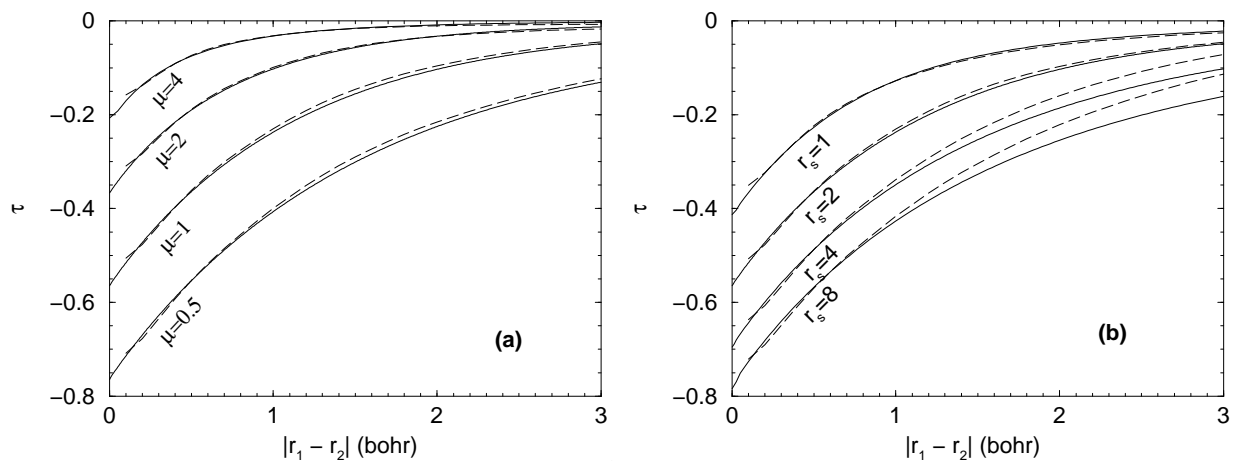


Figure 3.5: Comparison of first order JPT (dashed lines) and FHNC//0 (solid lines) pair-correlation functions for various screening parameters and densities. a) Fixed density $r_s = 2$ and different screening parameters ranging from $\mu = 0.5$ to 4 bohr^{-1} . b) Fixed screening parameter $\mu = 1 \text{ bohr}^{-1}$ and different densities ranging from $r_s = 1$ to 8 . The JPT results have been calculated for a supercell containing $N = 54$ particles.

4 Conclusions

We have presented a perturbation scheme for Jastrow-type correlation functions. From a computational point of view, this scheme is simple enough to be of practical significance for the determination of accurate trial wavefunctions to be used in QMC calculations. Although essentially limited to short-range correlations, JPT can be applied to the model Hamiltonian of the inhomogeneous RPA method in Ref. [23] in order to obtain Jastrow factors for metallic systems, where long-range correlations become important. Our applications to a screened homogeneous Fermi gas suggest that first order Jastrow factors are fairly accurate for short-range correlations and provide significantly better results in VMC calculations than it could be expected on the basis of second and third order correlation energies. It remains to develop an efficient implementation for inhomogeneous systems. For this, we have proposed multiscale bases [16], like wavelets, which provide sparse representations for two-particle correlation functions and a favourable computational complexity concerning the evaluation of Goldstone diagrams.

Table 3.1: Correlation energy per particle (mhartree) of a homogeneous Fermi gas at density $r_s = 2$ and different screening parameters μ . JPT2, JPT3 and VMC results have been obtained from first-order spin independent and spin dependent (*) Jastrow factors. The VMC method corresponds to a direct evaluation of the energy expectation value for first-order wavefunctions, whereas PDMC provides almost exact energies within the fixed-node approximation which do not depend on the Jastrow factor. For comparison MP2 correlation energies have been stated. A supercell containing $N = 54$ particles has been used in all calculations.

μ	JPT2	JPT3	VMC	JPT2*	JPT3*	VMC*	MP2	PDMC
0.5	-21.52	-15.66	-17.69(2)	-22.12	-16.19	-18.12(2)	-24.20	-18.70(3)
1	-8.91	-7.22	-7.63(2)	-9.33	-7.55	-7.94(2)	-10.35	-8.23(3)
2	-2.36	-2.03	-2.08(1)	-2.53	-2.15	-2.20(1)	-2.79	-2.31(2)
4	-0.450	-0.407	-0.415(4)	-0.480	-0.431	-0.435(4)	-0.525	-0.461(6)

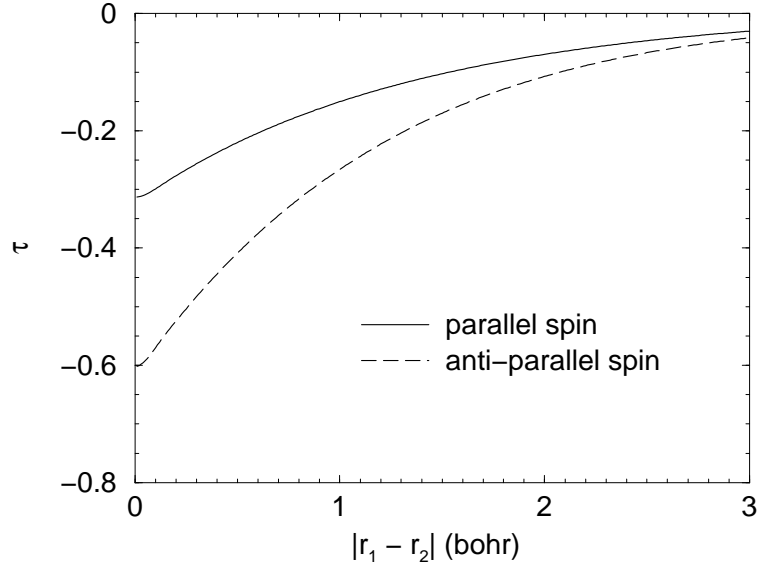


Figure 3.6: First order JPT pair-correlation functions for parallel and anti-parallel spins. The homogeneous Fermi gas has been considered at density $r_s = 2$ and $\mu = 1 \text{ bohr}^{-1}$ for a supercell with $N = 54$ particles.

Table 3.2: Same as Table 3.1 but for fixed screening parameter $\mu = 1 \text{ bohr}^{-1}$ and different densities.

r_s	JPT2	JPT3	VMC	JPT2*	JPT3*	VMC*	MP2	PDMC
1	-22.46	-19.26	-20.03(3)	-23.11	-19.87	-20.61(3)	-25.39	-21.5(1)
2	-8.91	-7.22	-7.63(2)	-9.33	-7.55	-7.94(2)	-10.35	-8.23(3)
4	-2.36	-1.68	-1.84(1)	-2.52	-1.76	-1.93(1)	-2.78	-2.02(1)
8	-0.457	-0.274	-0.322(2)	-0.486	-0.282	-0.335(2)	-0.524	-0.353(2)

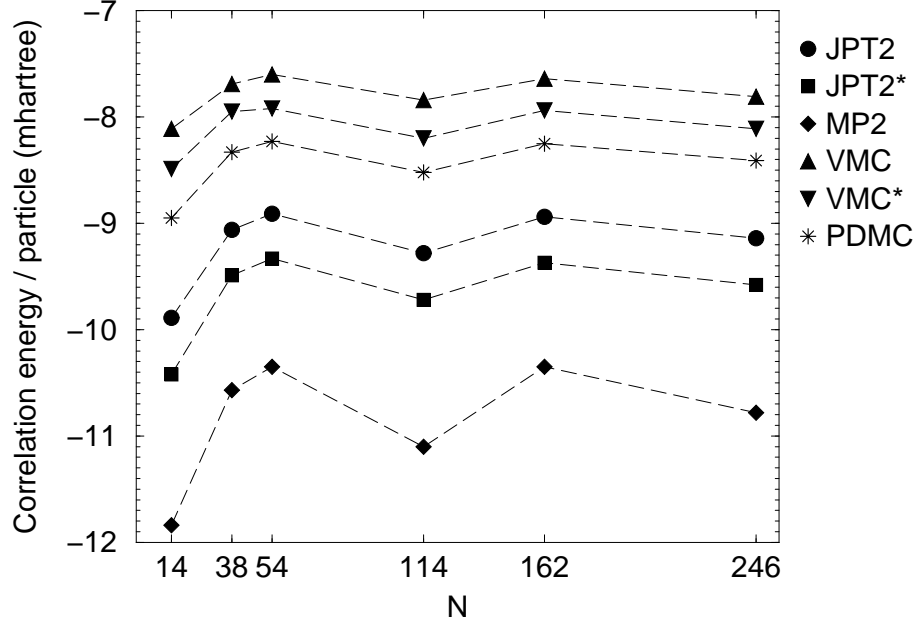


Figure 3.7: Correlation energy per particle (mhartree) of a homogeneous Fermi gas at density $r_s = 2$ and $\mu = 1 \text{ bohr}^{-1}$ for different size of the supercell.

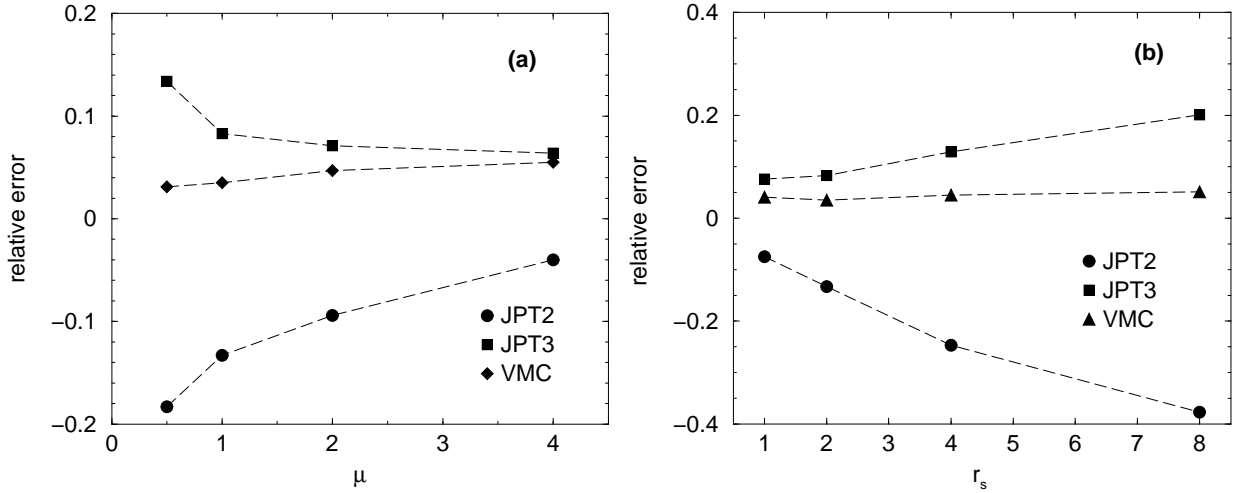


Figure 3.8: Relative errors $(E_{corr} - E_{corr}^{PDMC})/|E_{corr}^{PDMC}|$ of JPT2, JPT3, and VMC correlation energies for different densities and screening parameters. The results are for spin dependent Jastrow factors and supercells with $N = 54$ particles. (a) Fixed density $r_s = 2$. (b) Fixed screening parameter $\mu = 1 \text{ bohr}^{-1}$.

References

- [1] W. Kohn, *Rev. Mod. Phys.* **71**, 1253 (1999).
- [2] T. Helgaker, P. Jørgensen and J. Olsen, *Molecular Electronic-Structure Theory* (Wiley, New York, 1999).
- [3] T. D. Crawford and H. F. Schaefer, in *Reviews in Computational Chemistry*, edited by K. B. Lipkowitz and D. B. Boyd (Wiley-VCH, New York, 2000), Vol. 14, pp. 33–136.
- [4] E. Krotscheck, *Phys. Rev. B* **31**, 4267 (1985).
- [5] E. Krotscheck, W. Kohn and G.-X. Qian, *Phys. Rev. B* **32**, 5693 (1985).
- [6] J. Noga and W. Kutzelnigg, *J. Chem. Phys.* **101**, 7738 (1994).
- [7] W. M. C. Foulkes, L. Mitas, R. J. Needs and G. Rajagopal, *Rev. Mod. Phys.* **73**, 33 (2001).
- [8] C. J. Umrigar, K. G. Wilson, and J. W. Wilkins, *Phys. Rev. Lett.* **60**, 1719 (1988).
- [9] K. E. Schmidt, and J. W. Moskowitz, *J. Chem. Phys.* **93**, 4172 (1990).
- [10] A. J. Williamson, S. D. Kenny, G. Rajagopal, A. J. James, R. J. Needs, L. M. Fraser, W. M. C. Foulkes, and P. Maccullum, *Phys. Rev. B* **53**, 9640 (1996).
- [11] P. R. C. Kent, R. J. Needs and G. Rajagopal, *Phys. Rev. B* **59**, 12344 (1999).
- [12] X. Lin, H. Zhang and A. M. Rappe, *J. Chem. Phys.* **112**, 2650 (2000).
- [13] D. Prendergast, D. Bevan and S. Fahy, *Phys. Rev. B* **66**, 155104 (2002).
- [14] H.-J. Flad, W. Hackbusch, D. Kolb and R. Schneider, *J. Chem. Phys.* **116**, 9641 (2002).
- [15] H. Luo, D. Kolb, H.-J. Flad, W. Hackbusch and T. Koprucki, *J. Chem. Phys.* **117**, 3625 (2002).
- [16] H.-J. Flad, W. Hackbusch, H. Luo and D. Kolb, *Phys. Rev. B* **71**, 125115 (2005).
- [17] H.-J. Flad, W. Hackbusch and R. Schneider, submitted to *M2AN* , (2005).
- [18] G. Stollhoff and P. Fulde, *J. Chem. Phys.* **73**, 4548 (1980).
- [19] P. Fulde, *Electron Correlations in Molecules and Solids*, 3 ed. (Springer, Berlin, 1995).
- [20] J. W. Clark, in *Progress in Nuclear and Particle Physics*, edited by D. H. Wilkinson (Pergamon, Oxford, 1979), Vol. 2, pp. 89–199.
- [21] J. D. Talman, *Phys. Rev. A* **10**, 1333 (1974).
- [22] J. D. Talman, *Phys. Rev. A* **13**, 1200 (1976).
- [23] R. Gaudoin, M. Nekovee, W. M. C. Foulkes, R. J. Needs and G. Rajagopal, *Phys. Rev. B* **63**, 115115 (2001).
- [24] Y. Meyer, *Wavelets and Operators* (Cambridge University Press, Cambridge, 1992).
- [25] W. Hackbusch, *Computing* **67**, 35 (2001).
- [26] I. Lindgren and J. Morrison, *Atomic Many-Body Theory* (Springer, Berlin, 1986).
- [27] A. Szabo and N. S. Ostlund, *Modern Quantum Chemistry* (McGraw-Hill, New York, 1989).

- [28] C. Møller and M. S. Plesset, Phys. Rev. **46**, 618 (1934).
- [29] K. Szalewicz, B. Jeziorski, H. J. Monkhorst and J. G. Zabolitzky, J. Chem. Phys. **78**, 1420 (1983).
- [30] K. Szalewicz, B. Jeziorski, H. J. Monkhorst and J. G. Zabolitzky, J. Chem. Phys. **79**, 5543 (1983).
- [31] O. Sinanoğlu, Phys. Rev. **122**, 493 (1961).
- [32] O. Sinanoğlu, Proc. R. Soc. London, Ser. A **260**, 379 (1961).
- [33] C.-J. Huang, C. J. Umrigar and M. P. Nightingale, J. Chem. Phys. **107**, 3007 (1997).
- [34] D. Ceperley, Phys. Rev. B **18**, 3126 (1978).
- [35] T. Kato, Commun. Pure Appl. Math. **10**, 151 (1957).
- [36] L. M. Fraser, W. M. C. Foulkes, G. Rajagopal, R. J. Needs, S. D. Kenny and A. J. Williamson, Phys. Rev. B **53**, 1814 (1996).
- [37] M. Caffarel and P. Claverie, J. Chem. Phys. **88**, 1088 (1988).
- [38] M. Caffarel and P. Claverie, J. Chem. Phys. **88**, 1100 (1988).