Available online at www.sciencedirect.com

ScienceDirect

Journal of Computational Physics 217 (2006) 312–339

JOURNAL OF COMPUTATIONAL PHYSICS

www.elsevier.com/locate/jcp

An efficient numerical quadrature for the calculation of the potential energy of wavefunctions expressed in the Daubechies wavelet basis

A.I. Neelov *, S. Goedecker

Institute of Physics, University of Basel, Room 2.05, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

Received 23 May 2005; received in revised form 12 December 2005; accepted 3 January 2006
Available online 10 February 2006

Abstract

An efficient numerical quadrature is proposed for the approximate calculation of the potential energy in the context of pseudo potential electronic structure calculations with Daubechies wavelet and scaling function basis sets. Our quadrature is also applicable in the case of adaptive spatial resolution. Our theoretical error estimates are confirmed by numerical test calculations of the ground state energy and wavefunction of the harmonic oscillator in one dimension with and without adaptive resolution. As a byproduct we derive a filter, which, upon application on the scaling function coefficients of a smooth function, renders the approximate grid values of this function. This also allows for a fast calculation of the charge density from the wavefunction.

Keywords: Orthogonal wavelets; Schroedinger equation; Numerical quadrature; Electronic structure; Daubechies wavelets; Multiresolution; Adaptivity; Real space methods; Potential energy operator

1. Introduction

Gaussians and plane waves are at present the most popular basis sets for density functional electronic structure calculations. Wavelets are a promising new basis set that combines most of the theoretical advantages of these two basis sets. They can form a systematic orthogonal basis set that allows for adaptivity, the basis functions being localized both in real (compact support) and in Fourier space.

The first attempts to use wavelets in the electronic structure calculations appeared more than 10 years ago. The first papers we are aware of used the Mexican hat wavelet [1,2] and the Meyer wavelet [3]. However, these wavelet families were soon abandoned because they do not have compact support. Daubechies [4] wavelets were then investigated in a series of publications [5–8]. This basis is orthogonal and has the property of having the highest number of vanishing moments for the given support width, thus, combining locality and

---

* Corresponding author. Tel.: +44 0 61 267 3708; fax: +41 0 61 267 3784.
E-mail addresses: Alexey.Neelov@unibas.ch (A.I. Neelov), Stefan.Goedecker@unibas.ch (S. Goedecker).

0021-9991/$ - see front matter © 2006 Elsevier Inc. All rights reserved.
doi:10.1016/j.jcp.2006.01.003

approximating power. It is also localized in the momentum space. To use the Daubechies wavelets in the variational Galerkin method for the Schroedinger equation, one has to compute the matrix elements of the kinetic and potential energy operators. The algorithm for the kinetic part is straightforward [9].

The main difficulty is the calculation of the potential energy matrix elements [10–12]. They were computed by expanding the potential in terms of scaling functions, too, and then a convolution was performed with the matrix of products of three scaling functions. We will call this "the triple-product method": for details, see the beginning of Section 4. It requires a lot of computer resources; this motivated alternative approaches. The collocation approach [13] is not well suited for the Daubechies' wavelets since it spoils the favorable convergence rate of variational schemes. Another approach [14] involved designing a quadrature for the product of two scaling functions and a smooth function. It decreased the amount of computations in comparison to the triple-product method, but not sufficiently.

It follows from the above considerations that the Daubechies basis set can only be useful for electronic structure calculations if one has a better algorithm for the calculation of the potential energy than those available at the moment. Such an algorithm will be proposed in the present paper.

Due to the above listed problems with the Daubechies family, interest focused recently on the interpolating Deslarier–Dubuc [15] family [16–19]. Because the scaling functions of this family are interpolating (cardinal), the collocation approximation is much more accurate for them than for the Daubechies family. An even more accurate approximation for the potential energy is based on a relation with the analytically known overlap matrix elements [19].

The major disadvantage of the Deslarier–Dubuc wavelets is that they are not orthogonal. For very large systems, the dominating term in independent particle electronic structure calculations is the orthogonalization of the one-particle orbitals. The prefactor for this dominating cubic term is much smaller if an orthogonal basis set is used compared to a non-orthogonal basis set. Orthogonal wavelets are in addition also interesting candidates for the implementation of linear scaling algorithms [20].

Alpert [21,22] polynomial multiwavelets overcome the above-mentioned disadvantages. The potential energy can be calculated easily and they are orthogonal. It seems that they are the ideal basis set for all electron electronic structure calculations and impressive results have been reported [23–25]. The Chui–Lian [26] family has also been used in the same context [27]. Since multiwavelets can represent discontinuous functions, they are well suited to represent the electron–nucleus cusp in all electron calculations. However, if one uses the Bachelet–Hamann–Schulter [28] or Gaussian [29] pseudopotentials, the wavefunctions and the potential are smooth and this property is not needed. Then one may prefer the Daubechies wavelets due to their simplicity and small support length. For this reason, we explore in this paper the use of Daubechies wavelets for pseudopotential electronic structure calculations, together with our novel quadrature scheme. A topic that is closely related to the problem of integrating the potential energy is the problem of finding quadrature schemes for the product of a scaling function(wavelet) and a smooth function [14,30–33].

The quadrature scheme to be presented in this paper is aimed at applications in independent particle schemes such as density functional theory where one three-dimensional single particle orbital is provided for each electron in the system. For completeness, we will mention that wavelets have also been explored as a basis set for high dimensional many-electron wavefunctions [34–36]. Another approach is to use the so-called Weyl–Heisenberg wavelets that do not have multiresolution properties but are related to the structure of the phase space [37–40]. Although these two approaches are promising, any treatment of correlation entails an important increase of the numerical effort and such approaches will not allow to treat systems with several hundred atoms in the near future.

In the present paper, we apply one-scaling-function quadratures to the numerical calculation of the potential energy matrix element (1) between two smooth functions. For this purpose, we developed an algorithm for the reconstruction of grid values of a smooth function from its scaling function expansion coefficients. This technique might also be used in other contexts such as in speech reconstruction. Our scheme also provides a way to calculate the density from the wavefunction expressed in scaling functions. The extension of our potential energy quadrature onto the case of adaptive spatial resolution is then described.

The paper is organized as follows:

In Section 2 we briefly recall the definition and properties of the Daubechies wavelet family.

In Section 3 we recall the quadrature of [14,30] for the product of a scaling function and a smooth function.

Using that, in Section 4 we construct the quadrature for the potential energy functional, i.e., for the product of the form

$$\langle \text{smooth function} \,|\, \text{potential} \,|\, \text{another smooth function} \rangle. \tag{1}$$

In Section 4.1 we derive the quadrature and estimate its error, which behaves essentially as a square of the error for the wavelet expansion of the smooth functions involved. Then in Sections 4.2 and 4.3 we prove that the quadrature is exact if the potential and one of the functions in (1) are polynomials and another function is a scaling function. This confirms the previous estimation of error and extends it to the case when only one of the functions in (1) is smooth.

In Section 5 we extend our method onto the case of adaptive resolution, repeating the procedure of the previous section. The errors are again estimated. It is shown that now the main source of error is the boundary between regions with different resolution.

In Section 6 we modify our adaptive quadrature so that in the regions with constant resolution it reduces to the non-adaptive one. This reduces the computational burden. The price is that now we need to minimize the Rayleigh–Ritz (RR) functional in the space of smooth functions, i.e., smoothen the cusp of its gradient at the boundary between regions with different resolution.

In Section 7 we present a way to compute the density corresponding to a wavefunction expressed in scaling functions. It uses the approximate wavefunction values derived by the method described in Appendix A. This way is fast, but it reproduces multipole moments of the density only approximately (although with good precision).

Finally, in Section 8 we apply our methods to the calculation of the ground state energy and wavefunction of the harmonic oscillator, both with and without adaptivity. The results are then compared with those obtained with the RR functional in which the potential energy was calculated exactly. We consider both the least asymmetric and extremal phase Daubechies wavelets and argue that the least asymmetric family is preferable.

2. The orthogonal wavelets

In this work we use the Daubechies [4] scaling functions $\phi(x)$ and wavelets $\psi(x)$, in the dilated and shifted form:

$$\phi_i^k(x) \equiv 2^{k/2}\phi(2^k x - i), \quad \psi_i^k(x) = 2^{k/2}\psi(2^k x - i),$$

where i and k are integers. Sometimes we will also employ the intermediate notation

$$\phi_i(x) \equiv \phi(x - i), \quad \phi^k(x) = 2^{k/2}\phi(2^k x).$$

Our conclusions are the same for the least asymmetric and extremal phase [4] Daubechies wavelets, but the least asymmetric ones behave better in the examples we considered. Thus we will use the least asymmetric family for the illustration. The graph of the scaling function of this family of the order 8 is given in Fig. 1.

Since the Daubechies scaling functions and vectors are orthogonal, it is natural to use the bra and ket notation for them. Then, their orthogonality conditions can be written as

$$\langle \phi_i^k | \phi_j^k \rangle = \delta_{ij}; \quad \langle \phi_i^k | \psi_j^{k'} \rangle = 0, \ k' > k; \quad \langle \psi_i^k | \psi_j^{k'} \rangle = \delta_{ij}\delta_{kk'}.$$

One can define the following pair of sequences of spaces:

$$\mathscr{V}_k = \operatorname{span}\{|\phi_i^k\rangle\}, \quad \mathscr{W}_k = \operatorname{span}\{|\psi_i^k\rangle\}, \tag{2}$$

where $\mathscr{V}_k \cap \mathscr{W}_k = \{0\}$. It turns out that for any $k \geqslant 0$ the space of square integrable functions $\mathscr{L}^2$ can be decomposed into the following infinite direct sum:

$$\mathscr{L}^2 = \mathscr{V}_k \oplus \mathscr{W}_k \oplus \mathscr{W}_{k+1} \oplus \cdots \tag{3}$$

In this paper we will need the refinement relations

$$|\phi_i^{k-1}\rangle = \sum_j h_j |\phi_{2i+j}^k\rangle, \quad |\psi_i^{k-1}\rangle = \sum_j g_j |\phi_{2i+j}^k\rangle. \tag{4}$$

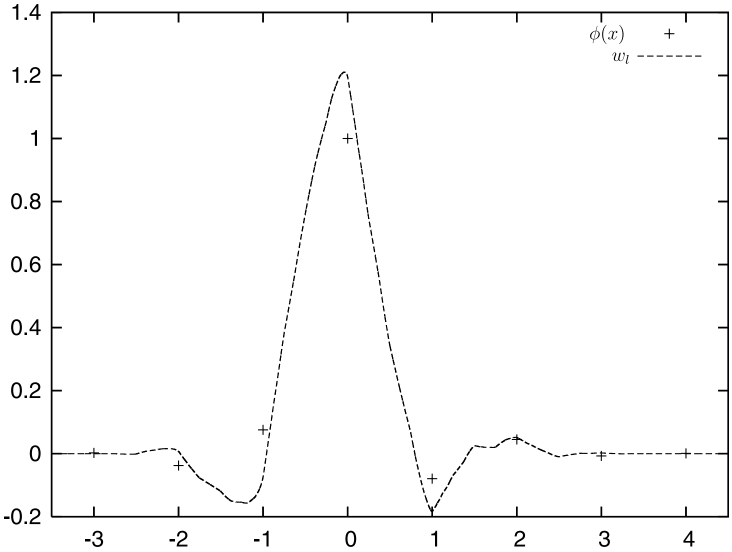

Fig. 1. The least asymmetric Daubechies-8 scaling function and the corresponding quadrature filter. As seen from the graph, the filter values are close to the scaling function values at integer points.

They are just a unitary transformation to a new basis; for the spaces (2) it means that

$$\mathscr{V}^k = \mathscr{V}^{k-1} \oplus \mathscr{W}^{k-1}. \tag{5}$$

The inverse of (4) is called the forward wavelet transformation, but we do not need its explicit form in the present text.

The decomposition (3) can be reformulated in the following way: any square integrable function $f(x)$ can be uniquely expanded as

$$f(x) = \sum_j c_j^k \phi_j^k(x) + \delta f(x), \quad \delta f(x) = \sum_{q=k}^{\infty} \sum_j d_j^q \psi_j^q(x). \tag{6}$$

The Daubechies-2m scaling functions are a basis of degree $m-1$. This means that every polynomial of degree less than m is contained in $\mathscr{V}_k$. Therefore [41], the tail part of the series (6) behaves as

$$\delta f(x) = \mathcal{O}(h^m), \quad h = 2^{-k} \tag{7}$$

with respect to the $\mathscr{L}^2$ norm. The $\mathcal{O}$ notation corresponds to the limit of $k \to \infty$. For the polynomials of degree less than m, $\delta f(x) = 0$.

The methods to be presented can be extended to other wavelet families, not necessarily orthogonal, but in our opinion, the Daubechies family is the optimal choice for the electronic structure calculations.

3. Quadrature for orthogonal wavelets

A wavelet quadrature is determined by the set of coefficients w_l such that for a smooth function $G(x)$, the integral $\int G(x)\phi_r^k(x)\,\mathrm{d}x$ is approximated by $\sqrt{h}\sum_l w_l G_{l+r}^k$, where $G_j^k \equiv G(2^{-k}x)$. The $\sqrt{h}$ factor comes from the normalization of the scaling function. The degree of accuracy of a quadrature formula is M if it yields the exact result for every polynomial of degree less than or equal to M. This is equivalent to the condition [14]

$$\sum_l \omega_l l^s = M_s, \quad M_s \equiv \int y^s \phi(y)\,\mathrm{d}y, \tag{8}$$

where M_s are the scaling function moments.

Table 1
The values of the filters w_l for the Daubechies-2m least asymmetric filters; $m = 3,\ldots,6$

l	Daubechies-6	Daubechies-8	Daubechies-10	Daubechies-12
−5				0.0000754232174770
−4			0.0003712028220936	−0.0011760498174610
−3		0.0026299127476935	−0.0046529756260417	0.0104347966396891
−2	0.0858797754503928	−0.0377927339236569	0.0306436002784248	−0.0340901829704789
−1	1.0472376804223309	0.0755988357512099	−0.1207447752890374	−0.0067678682684262
0	−0.1886782932535312	0.9999560903030736	0.1338108260452157	1.0005931732054807
1	0.0795781221430145	−0.0794124676160406	0.9123169219278740	0.0041859363010669
2	−0.0288721312776034	0.0451427040622791	0.0109419516584456	0.0351468153360141
3	0.0048548465153963	−0.0069875964135745	0.0393078583967683	−0.0096794739531791
4		0.0008652550890159	−0.0022599250999316	0.0015648660417616
5			0.0002653148861886	−0.0003139771845937
6				0.0000265414526497

If the quadrature filter is of degree M and $G(x)$ belongs to $\mathscr{C}^{M+1}$ then [14,30]

$$\int G(x)\phi_r^k(x)\,\mathrm{d}x = \sqrt{h}\sum_l w_l G_{l+r}^k + \mathcal{O}(h^{M+3/2}) = \sqrt{h}\sum_s w_{s-r} G_s^k + \mathcal{O}(h^{M+3/2}), \quad h = 2^{-k}. \tag{9}$$

In this paper we will usually work with the uniform quadrature (of degree M) set forth in [14]

$$w_l = \sum_{r=0}^{M} P_{lr} M_r. \tag{10}$$

It will be supposed that $M = 2m - 1$ for the Daubechies-2m wavelets, and $l = 1 - m,\ldots,m$. In (10), the Lagrange polynomials of degree M are used

$$P_l(y) = \prod_{j=1-m}^{m} \frac{y-j}{l-j} \equiv \sum_{r=0}^{2m-1} P_{lr} y^r, \quad P_{lr} = \frac{1}{r!}\frac{\mathrm{d}^r P_l(y)}{\mathrm{d}y^r}\Big|_{y=0}.$$

The non-zero values of the filters w_l for $m = 3,\ldots,6$ are shown in Table 1.

Fig. 1 also contains the values of w_l for $m = 4$ and $M = 7$, compared with the graph of the corresponding scaling function (least asymmetric Daubechies-8). One sees that the filter values are close to the scaling function values at integer points.

4. The potential energy functional for orthogonal wavelets

An important step in the electronic structure calculation is to find the eigenspectrum of a one-particle Schroedinger equation. Although our approach can be extended to the three-dimensional systems, in this paper we will consider only one-dimensional ones

$$\hat{H}\Psi(x) = E\Psi(x), \quad \hat{H} = -\frac{1}{2}\frac{\partial^2}{\partial x^2} + V(x) \equiv \hat{T} + \hat{U}, \quad \int \mathrm{d}x\, \Psi^2(x) = 1. \tag{11}$$

This equation can be solved by representing a trial wavefunction as a linear combination of finite elements [42]. In the case considered in this paper, these finite elements are the Daubechies scaling functions (wavelets). Thus we select the trial function in the form

$$\Psi_I(x) = \sum_i c_i^k \phi_i^k(x), \tag{12}$$

which can also be represented by the ket vector $|c^k\rangle = \sum_{i=-N}^{N} c_i|\phi_i^k\rangle$ with $N = L/h = 2^{-k}L$. In this paper we use non-periodic boundary conditions, although we could use the periodic ones too. Then if the Rayleigh–Ritz functional is defined as:

$$R(\{c_i^k\}) = \frac{\langle c^k|\hat{H}|c^k\rangle}{\langle c^k|c^k\rangle} = \frac{T(\{c_i^k\}) + U(\{c_i^k\})}{\langle c^k|c^k\rangle}, \tag{13}$$

$$T(\{c_i^k\}) \equiv \langle c^k|\hat{T}|c^k\rangle, \quad U(\{c_i^k\}) \equiv \langle c^k|\hat{U}|c^k\rangle, \tag{14}$$

the variational ground state energy and wavefunction expansion coefficients have the form

$$E_0 = \min R(\{c_i^k\}), \quad |c_0^k\rangle = \arg\min R(\{c_i^k\}). \tag{15}$$

The other wavefunctions are obtained in the same way using the Lagrange multipliers.

Since the Daubechies-$2m$ scaling functions have degree $m-1$ (see Section 2), the ground state energy error behaves [42] as

$$\delta E \equiv E_0 - E_{\text{ex}} = \mathcal{O}(h^{2m-2}), \tag{16}$$

where E_{ex} is the exact ground state energy of (11).

The kinetic energy has the form [43]

$$T(\{c_i^k\}) = -\frac{1}{2h^2}\sum_{i,j} a_{i-j} c_i^k c_j^k,$$

where a_l is the kinetic energy filter.

The potential energy (14) can be written in the conventional form as

$$U = \int \Psi_I(x) V(x) \Psi_I(x)\,\mathrm{d}x.$$

For the calculation of the excited states we have to evaluate the more general potential energy bilinear form

$$U = \int \Phi_I(x) V(x) \Psi_I(x)\,\mathrm{d}x, \tag{17}$$

where the wavefunction $\Phi_I(x)$ has the same form (12)

$$\Phi_I(x) = \sum_i c_i^k \phi_i^k(x). \tag{18}$$

In the electronic structure calculations the exact analytical form of the potential $V(x)$ is not known. Any approximation of the potential should invoke energy errors that alter the Rayleigh–Ritz functional (13) as little as possible; otherwise the minimization with an approximate functional will not converge to the true minimum. We choose the following quantitative criterion for this: the error in the potential that arises from approximation should be small compared to (16). When the grid parameter h is small, there is a convenient way to ensure that: we require that the approximation error behaves as h^{2m}. Note that, e.g., the collocation approximation [13] does not fulfill the latter requirement.

The most natural way to approximate the potential is to expand it in the interpolating scaling functions $\phi_i^{kI}(x)$ [15,43] of degree $2m$

$$V(x) = \sum_i V_i^k \phi_i^{kI}(x) + \delta V(x), \quad \delta V(x) = \mathcal{O}(h^{2m}), \quad \phi_i^{kI}(x) \equiv \phi^I(x/h - i), \quad V_i^k \equiv V(ih), \quad h = 2^{-k}, \tag{19}$$

where the error estimate (19) holds for sufficiently smooth potentials. Substituting the expansions (12), (18), and (19) into (17), we get:

$$U = \sum_{ijl} c_i^k V_j^k s_l^k \int \phi_i^k(x)\phi_j^{kI}(x)\phi_l^k(x)\,\mathrm{d}x + \mathcal{O}(h^{2m}) = \sum_{ijl} c_i^k V_j^k s_l^k I_{i-j,l-j} + \mathcal{O}(h^{2m}), \tag{20}$$

$$I_{rs} \equiv \int \phi_r(x)\phi^I(x)\phi_s(x)\,\mathrm{d}x, \tag{21}$$

where (21) is the triple-product matrix mentioned in Section 1. Unfortunately, the formula (20) requires too many floating point operations (flops) per grid point. In the simplest case when s_i^k coincide with c_i^k, the leading term in that number is $3/2N^2$ in one dimension and $2N^4$ in three dimensions. N is the dimension of the matrix

(21), and eigenvalue decomposition of the matrix (21) is used in the 3D case. For the Daubehies-$2m$ scaling functions, $N = 2m - 2$, so the leading term in m of the number of flops per gridpoint is $6m^2$ in 1 dimension and $32m^4$ in 3 dimensions. This is unacceptable for the realistic values of m of the order of 10.

Therefore we need some approximation that would decrease the amount of computations, but still invoke the error that asymptotically behaves as $\mathcal{O}(h^{2m})$, at most. Such an approximation will be described in the following section.

4.1. The quadrature for the case of smooth wavefunctions

Let us consider two smooth functions $\Phi(x)$, $\Psi(x)$ together with their approximate wavelet expansion $\Phi_I(x)$, $\Psi_I(x)$ (12),(18). The expansion coefficients c_i^k and s_i^k are obviously given by

$$c_i^k = \int \phi_i^k(x)\Phi(x)\,\mathrm{d}x, \quad s_i^k = \int \phi_i^k(x)\Psi(x)\,\mathrm{d}x. \tag{22}$$

Then, according to (6) and (7),

$$\Phi(x) = \Phi_I(x) + \delta\Phi(x), \quad \Psi(x) = \Psi_I(x) + \delta\Psi(x), \tag{23}$$

$$\delta\Phi(x) = \sum_{k'>k}\sum_i d_i^{k'}\psi_i^{k'}(x) = \mathcal{O}(h^m), \quad \delta\Psi(x) = \sum_{k'>k}\sum_i b_i^{k'}\psi_i^{k'}(x) = \mathcal{O}(h^m). \tag{24}$$

From (23) we get

$$\Phi_I(x) = \Phi(x) - \delta\Phi(x), \quad \Psi_I(x) = \Psi(x) - \delta\Psi(x).$$

Plugging this expression into (17), we can get after some easy calculations:

$$U = U_\mathrm{A} + \delta U_1 + \delta U_2, \quad U_\mathrm{A} \equiv \int \Phi_I(x)V(x)\Psi(x)\,\mathrm{d}x, \tag{25}$$

$$\delta U_1 \equiv \int \delta\Phi(x)V(x)\delta\Psi(x)\,\mathrm{d}x, \quad \delta U_2 \equiv -\int \Phi(x)V(x)\delta\Psi(x)\,\mathrm{d}x. \tag{26}$$

In the following section we will prove that

$$\delta U_1 + \delta U_2 = \mathcal{O}(h^{2m}) \tag{27}$$

from the local properties of scaling functions. In this section we will show for the two error terms separately that

$$\delta U_1 = \mathcal{O}(h^{2m}), \quad \delta U_2 = \mathcal{O}(h^{2m}). \tag{28}$$

The first estimate of (28) follows from (24) and the fact that individual wavelet coefficients $d_i^{k'}$, $b_i^{k'}$ from the tail parts behave as $2^{-k'(m+1/2)}$ [30]. If the potential $V(x)$ is bounded then it follows from the Schwarz inequality:

$$\left|\int \delta\Phi(x)V(x)\delta\Psi(x)\,\mathrm{d}x\right| \leqslant \max|V|\int |\delta\Phi(x)||\delta\Psi(x)|\,\mathrm{d}x \leqslant \max|V|\|\delta\Phi\|\|\delta\Psi\|.$$

To prove the second part of (28), let us define the function $H(x) = \Phi(x)V(x)$. It can also be interpolated by scaling functions:

$$H(x) = H_I(x) + \delta H(x), \quad H_I(x) = \sum_i h_i^k\phi_i^k(x), \quad h_i^k = \int \phi_i^k(x)H(x)\,\mathrm{d}x, \quad \delta H(x) = \sum_{k'>k}\sum_i q_i^{k'}\psi_i^{k'}(x) = \mathcal{O}(h^m).$$

Then, $\int H_I(x)\delta\Psi(x)\,\mathrm{d}x = 0$, and

$$\delta U_2 = -\int \delta H(x)\delta\Psi(x)\,\mathrm{d}x = \mathcal{O}(h^{2m}).$$

We are left with the approximation (25). Substituting (18) into it, we get

$$U_{\mathrm{A}} = \sum_i c_i^k \int \phi_i^k(x) G(x)\,\mathrm{d}x, \tag{29}$$

where $G(x) \equiv V(x)\Psi(x)$. The scalar products in (29) can now be determined by applying the scaling function quadrature (9)

$$U_{\mathrm{A}}[\Psi_I] = \sum_i c_i^k \left[\sqrt{h}\sum_s w_{s-i} G_s^k + \mathcal{O}(h^{2m+1/2})\right] = \sum_s p_s G_s^k + \mathcal{O}(h^{2m}), \tag{30}$$

where

$$G_q^k = G(hq) = V(hq)\Psi(hq) = V_q^k \Psi_q^k$$

are the grid values of the function $G(x)$. As seen from (30), the quantity

$$p_s^k \equiv \sqrt{h}\sum_i c_i^k w_{s-i}$$

plays the role of the quadrature for the whole function Φ_I.

Now we need to find the grid values of the smooth function $G(x)$. We suppose that we know the grid values V_q^k of the smooth potential. Then it remains to reconstruct the grid values Ψ_i^k of the unknown wavefunction from the known coefficients s_i^k.

The easiest way to obtain the grid values would be by using (12)

$$\Psi(\mathrm{j}h) \approx \Psi_I(\mathrm{j}h) = \sum_i s_i^k \phi_i^k(\mathrm{j}h). \tag{31}$$

Since the Daubechies scaling functions are not very regular, the value of a scaling function at a real space grid point does not very well represent the behavior of the scaling function in a small interval around this grid point. As a consequence of this and in accordance with (7), the error in (31) behaves like $\mathcal{O}(h^m)$, which is much worse than (27) and we therefore discard this possibility. We will instead introduce some smoothed grid values $\bar{\Psi}_q^k$ which better represent the average behavior of Ψ_I. In Appendix A it will be shown that there exists a filter W_j such that the smoothed grid values can be obtained by a convolution from the scaling function expansion coefficients

$$\bar{\Psi}_q^k \equiv \frac{1}{\sqrt{h}}\sum_t W_{q-t} s_t^k = \frac{1}{\sqrt{h}}\sum_j W_j s_{q-j}^k \tag{32}$$

and that the error behaves as

$$\Psi_q^k = \bar{\Psi}_q^k + \mathcal{O}(h^{2m}).$$

Substituting everything back into (30),we get

$$U_{\mathrm{A}} = \sum_s p_s^k V_s^k \bar{\Psi}_s^k + \mathcal{O}(h^{2m}). \tag{33}$$

Taking together (25), (27) and (33), one gets

$$U = U_{\mathrm{f}} + \mathcal{O}(h^{2m}), \quad U_{\mathrm{f}} \equiv \sum_s p_s^k V_s^k \bar{\Psi}_s^k, \tag{34}$$

where “f” stands for “filters”.

As explained in Appendix A, in the case of the Daubechies scaling functions the quadrature filter w_l and the reconstruction filter W_l are identical. Then (34) assumes the following simple form:

$$U_{\mathrm{f}} \equiv h\sum_s \bar{\Phi}_s^k V_s^k \bar{\Psi}_s^k, \quad \bar{\Phi}_q^k \equiv \frac{1}{\sqrt{h}}\sum_t w_{q-t} c_t^k. \tag{35}$$

This formula can be computed very fast: when $c_i^k = s_i^k$, the number of flops per grid point is just $4m+3$. One can use it in three dimensions too. In that case the quadrature and reconstruction filters are tensor products of the one-dimensional ones. Thus one only needs three convolutions with filters of length $2m$ per grid point for the calculation of the potential energy, thus $12m + 3$ flops. This is clearly better than the $32m^4 + \cdots$ result for the triple-product method.

Formulas (34) and (35) have a global nature; they characterize the approximation error over the whole domain of the wavefunction. In the following two sections we will choose a local point of view instead, and prove that Eqs. (34) and (35) hold for unbounded $V(x)$ too.

4.2. The matrix elements of the potential energy in the case of the Daubechies family

If we substitute the wavefunction expansion (12) and (18) into the energy expression (17), we get

$$U = \sum_{i,j} c_i^k s_j^k U_{ij}, \quad U_{ij} \equiv \int \mathrm{d}x \phi_i^k(x) V(x) \phi_j^k(x). \tag{36}$$

Similarly, the approximate energy (35) in the case of the Daubechies family has the form

$$U_\mathrm{f} = \sum_{i,j} c_i^k s_j^k U_{ij}^\mathrm{f}, \quad U_{ij}^\mathrm{f} \equiv \sum_l V_l^k w_{l-i} w_{l-j}, \tag{37}$$

where w_l is the quadrature filter for the Daubechies family.

It is enough to consider the matrix elements U_{0q}; U_{0q}^f since others can be obtained by shifting the potential. Let us assume that $V(x) = x^t$. Then, after going to the variable $y = x/h$ under the integral one obtains

$$U_{0q} = \int \phi^k(x) x^t \phi_q^k(x)\,\mathrm{d}x = h^t K_{qt}, \quad K_{qt} \equiv \int \mathrm{d}y\, \phi(y) \phi_q(y) y^t. \tag{38}$$

Similarly, one can check that

$$U_{0q}^\mathrm{f} = h^t K_{qt}^\mathrm{f}, \quad K_{qt}^\mathrm{f} \equiv \sum_l w_{l-q} w_l\, l^t. \tag{39}$$

Now, if the potential $V(x)$ is a smooth function, one can expand it into a Taylor series around the origin

$$V(x) = \sum_{p=0}^{2m-1} \frac{V^{(p)}(0)}{p!} x^p + \mathcal{O}(x^{2m}). \tag{40}$$

Our aim is to use wavelets for the electronic structure calculations with pseudopotentials, and the local part of the Gaussian pseudopotentials introduced in [29] is smooth (has infinite number of continuous derivatives). The smoothness of the potential is best exploited if the grid is fine enough. If necessary, one can also increase the accuracy of our approximation without changing the number of basis functions, by going to the doubly dense real space grid. This is discussed in more detail in the very end of Section 5.1. The multiscale nature of wavelets allows one to adapt the grid resolution locally, which will be described in Section 5.

Note however that one needs only the grid values of the potential for the actual calculations (according to Eq. (35)). The Taylor expansion of the potential (40) is presented here only to analyze the errors.

Plugging the expansion (40) into (36) and taking into account (38), we get

$$U_{0q} = \sum_{p=0}^{2m-1} \frac{V^{(p)}(0)}{p!} h^p K_{qp} + \mathcal{O}(h^{2m}). \tag{41}$$

In the same way, plugging (40) into the right equation of (37) and using (39), one obtains

$$U_{0q}^\mathrm{f} = \sum_{p=0}^{2m-1} \frac{V^{(p)}(0)}{p!} h^p K_{qp}^\mathrm{f} + \mathcal{O}(h^{2m}).$$

It is in the coefficients K_{qp} that the approximation differs from the exact calculation. Eq. (39) can be considered as a quadrature approximation of the integral (38).

It would be convenient if the values of K_{qp} and K_{qp}^f coincided. It would explain the smallness of error in (33), since in that case, U_{ij} and U_{ij}^f would differ only by $\mathcal{O}(h^{2m})$. However, for the cases we checked (Daubechies-6 to

18) the values of K turned out to be different. Thus, the smoothness of the potential alone does not explain the smallness of error (34). In the following section, we will see that one needs also the smoothness of the wavefunction to explain that.

Note that the strategy used in [14] was to find an approximation of the matrix (36) such that the corresponding K_q^p be exactly equal to (38), i.e., that the approximation is exact for the polynomial potentials. This is a flexible scheme but it requires more computer resources than ours. The reason is that in the scheme of [14] one has to apply two-index filters at each point. Therefore we have the same problem as with the triple-product formula (20): the number of flops per grid point is $\sim N^2$ in one dimension and $\sim N^4$ in three dimensions, where N is the dimension of the matrix which plays the role of (21).

4.3. The gradient of the potential energy in the case of the Daubechies family

The gradients of the exact and approximate energies (36) and (37) w.r.t. c_i^k are

$$\frac{\partial U}{\partial c_i^k} = \sum_j s_j^k U_{ij}, \quad \frac{\partial U_{\mathrm{f}}}{\partial c_i^k} = \sum_j s_j^k U_{ij}^{\mathrm{f}}, \tag{42}$$

where the matrix elements are discussed in the previous section. In the case of the potential energy functional, with the coefficients c_i^k instead of s_i^k in (36) and (37), there should be an additional factor of 2 in (42).

From (34) one could guess that

$$\frac{\partial U}{\partial c_i^k} = \frac{\partial U_{\mathrm{f}}}{\partial c_i^k} + \mathcal{O}(h^{2m+1/2}), \tag{43}$$

since there are no preferred points in space for the energy expressions (36) and (35), and thus the energy error should be "smeared" smoothly over the grid points. Later in this section we will see that (43) is indeed satisfied.

The condition (43) is very important. It means that if we minimize U_{f} using some numerical method (steepest descent, etc.) then the gradients along which we change the wavefunctions will be very close for the exact and approximated energies, and thus the results of minimization, the two ground state wavefunctions, will also be close.

On the other hand, Eq. (34) that was derived above only for a bounded $V(x)$, follows from (43). What is more, it follows from (43) that (34) is satisfied even if we do not require the function $\Phi(x)$ to be smooth.

At first we will prove (43) for the partial case of $i = 0$, $\Psi(x) = x^l$ and $V(x) = x^t$, from which we then will easily make extension onto the general case. With the above assumptions, acting similarly to the proof of (100) one can check that

$$s_j^k = h^{l+1/2} \sum_{u=0}^{l} C_l^u j^u M_{l-u}, \tag{44}$$

where C_l^u are the binomial coefficients, and M_s are the scaling function moments (8).

Substituting (38) and (44) into (42), one obtains

$$\frac{\partial U}{\partial c_0^k} = h^{l+t+1/2} A_{lt}, \quad A_{lt} \equiv \sum_{u=0}^{l} C_l^u M_{l-u} \sum_j K_{jt} j^u. \tag{45}$$

Similarly, in the case of the approximate energy,

$$\frac{\partial U_{\mathrm{f}}}{\partial c_0^k} = h^{l+t+1/2} A_{lt}^{\mathrm{f}}, \quad A_{lt}^{\mathrm{f}} \equiv \sum_{u=0}^{l} C_l^u M_{l-u} \sum_j K_{jt}^{\mathrm{f}} j^u. \tag{46}$$

The coefficients (45) and (46) satisfy the following equalities:

$$A_{lt} = M_{l+t}, \quad l,t < m; \qquad A_{l0} = M_l, \quad l < 2m; \tag{47}$$

$$A_{lt}^{\mathrm{f}} = M_{l+t}, \quad t + m < 2m; \tag{48}$$

where M_s are the scaling function moments (8). Their analytical proof can be found in Appendix B.

Since we were unable to find an analytical proof of (47) for the remaining relevant values of indices:

$$A_{lt} = M_{l+t}, \quad m < t < 2m, \tag{49}$$

we checked it numerically with Mathematica for the Daubechies wavelets of the orders from 6 to 18. We checked both the extremal phase and least asymmetric Daubechies families. The relative error of (49) can be made arbitrarily small by increasing the precision of wavelet filters. We went down to 10^{-40}.

Thus we see that for the Daubechies wavelets,

$$A_{lt} = A_{lt}^{\mathrm{f}} = M_{l+t}, \quad \frac{\partial U}{\partial c_0^k} = \frac{\partial U_{\mathrm{f}}}{\partial c_0^k} = h^{l+t+1/2} M_{l+t}, \qquad l + t < 2m \tag{50}$$

for arbitrary polynomial $V(x)$ and $\Psi(x)$ with the sum of their degrees less than $2m$. The second equality follows from (45) and (46).

In the remaining part of this section we will infer (43) from (50). Let us consider the exact gradient. It is given by the left formula (42). The matrix element U_{0j} has been calculated in the previous section. Now let us consider the scaling function expansion coefficient s_j^k (22) for a smooth function $\Psi(x)$. Since the latter is smooth, it can be expanded into the Taylor series at the origin

$$\Psi(x) = \sum_{p=0}^{2m-1} \frac{\Psi^{(p)}(0)}{p!} x^p + \mathcal{O}(x^{2m}). \tag{51}$$

Plugging (51) into (22) and using (44), we get

$$s_j^k = \sum_{p=0}^{2m-1} \frac{\Psi^{(p)}(\mathrm{i}h)}{p!} h^{p+1/2} \sum_{t=0}^{p} C_p^t q^{p-t} M_t + \mathcal{O}(h^{2m+1/2}). \tag{52}$$

Substituting (52) and (41) into (42), we get

$$\begin{aligned} \frac{\partial U}{\partial c_0^k} &= \sum_{s=0}^{2m-1} \frac{h^{s+1/2}}{s!} \sum_{p=0}^{s} C_s^p \Psi^{(p)}(0) V^{(s-p)}(0) A_{p,s-p} + \mathcal{O}(h^{2m+1/2}) = \sum_{s=0}^{2m-1} \frac{h^{s+1/2}}{s!} G^{(s)}(0) M_s + \mathcal{O}(h^{2m+1/2}) \\ &= g_0^k + \mathcal{O}(h^{2m+1/2}), \end{aligned} \tag{53}$$

where we have used (50). As before, $G(x) \equiv V(x)\Psi(x)$, and $g_i^k \equiv \int G(x)\phi_i^k(x)\,\mathrm{d}x$. The analog of (53) for an arbitrary gradient component $\frac{\partial U}{\partial c_i^k}$ can be obtained by replacing all the 0 by $\mathrm{i}h$. Then our error estimate for the potential energy of Eq. (27) follows from the formula

$$U = \sum_i c_i^k \frac{\partial U}{\partial c_i^k}. \tag{54}$$

For the gradient of the approximate energy one can exactly repeat the above steps, just using K_{jt}^{f} instead of K_{jt} (since one starts from (39) instead of (41)). The result will be identical to (53); thus, (43) is proved.

5. The case of adaptive resolution

In the actual calculations, most of the wavefunction coefficients for wavelets on fine levels are very small. An advantage of wavelets is that the coefficients smaller than certain threshold can be set to zero [6,7]; this is called dynamical adaptivity. We will use here the so-called static adaptivity when one constrains the fine wavelets to be zero at the some parts of configuration space – e.g., far from the atomic cores. This effectively leads to grid resolution slowly varying in real space. Since our quadrature approximation involves some extra care of the boundaries between regions of different grid resolution (see below), it is more suited for the static adaptivity where one can make sure that these boundaries have simple form.

The simplest two-level example will be considered below.

5.1. The energy error for the adaptive approximation

In this section we will extensively use the bra and ket notation for the wavefunctions (12) and (18)

$$|c^k\rangle \equiv \sum_i c_i^k |\phi_i^k\rangle, \quad |s^k\rangle \equiv \sum_i s_i^k |\phi_i^k\rangle. \tag{55}$$

The ket vectors belong to the space $\mathcal{V}_k$ (2). It is finite-dimensional after we apply boundary conditions, whether periodic or non-periodic.

The energy expressions (36) and (37) then can be written as

$$U = \langle c^k|\hat{U}|s^k\rangle, \quad U_{\mathrm{f}} = \langle c^k|\hat{U}_{\mathrm{f}}^k|s^k\rangle. \tag{56}$$

Note the difference between the exact and approximate operators. $\hat{U}$ is the operator of multiplication by $V(x)$ in a Hilbert space. On the other hand, $\hat{U}_{\mathrm{f}}^k$ is an operator in the finite-dimensional space V_k, taking into account the boundary conditions. Its matrix elements are given by (37).

In the present section we will consider two resolution levels simultaneously. In addition to the resolution level k, considered up to now, we have a more coarse level $k-1$ together with the vectors associated with it

$$|c^{k-1}\rangle \equiv \sum_i c_i^{k-1} |\phi_i^{k-1}\rangle, \quad |d^{k-1}\rangle \equiv \sum_i d_i^{k-1} |\psi_i^{k-1}\rangle. \tag{57}$$

For some time we will discuss only the vectors corresponding to $\Phi_I(x)$; those related to $\Psi_I(x)$ satisfy the same relations with the symbols c,d replaced by s,b where appropriate.

Similarly to (22), the coefficients in (55) and (57) can be expressed in terms of the smooth function $\Phi(x)$:

$$c_i^k = \int \phi_i^k(x)\Phi(x)\,\mathrm{d}x, \quad c_i^{k-1} = \int \phi_i^{k-1}(x)\Phi(x)\,\mathrm{d}x, \quad d_i^{k-1} = \int \psi_i^{k-1}(x)\Phi(x)\,\mathrm{d}x.$$

The bra and ket notation is convenient because in accordance with (5), the forward and backward wavelet transformations can be written as

$$|c^k\rangle = |c^{k-1}\rangle + |d^{k-1}\rangle. \tag{58}$$

The left and right parts of the above equality differ only in the choice of basis functions: $\{\phi_i^k\}$ at the left and $\{\phi_i^{k-1}, \psi_i^{k-1}\}$ at the right.

The adaptive approximation consists of replacing the second relation in (57) by

$$|\tilde{d}^{k-1}\rangle \equiv \sum_{i\in D} d_i^{k-1} |\psi_i^{k-1}\rangle. \tag{59}$$

In this paper we consider the so-called static adaptivity, where one chooses some fixed predetermined region D of space such that outside D the wavelet coefficients d_i^k are known to be small and can therefore be neglected. We will call D the fine region.

Replacing $|d^{k-1}\rangle$ in (58) by (59), one can form the adaptive vector

$$|\tilde{c}^k\rangle \equiv |c^{k-1}\rangle + |\tilde{d}^{k-1}\rangle. \tag{60}$$

In other words, if we forward transform $|c^k\rangle$, then discard the wavelet part outside the domain D according to Eq. (59) and then backward transform the result, then we get $|\tilde{c}^k\rangle$.

The discarded wavelet part can also be written as a vector

$$|\check{d}^{k-1}\rangle \equiv \sum_{i\notin D} d_i^{k-1} |\psi_i^{k-1}\rangle, \tag{61}$$

so that, taking into account (59),

$$|d^{k-1}\rangle = |\tilde{d}^{k-1}\rangle + |\check{d}^{k-1}\rangle. \tag{62}$$

The exact energy expression (56) in the adaptive case assumes the form

$$U^{\mathrm{a}} = \langle \tilde{c}^k|\hat{U}|\tilde{s}^k\rangle, \tag{63}$$

where $\hat{U}$ is the matrix (36), and $|\tilde{s}^k\rangle$ is formed from $|s^k\rangle$ in the same way as $|\tilde{c}^k\rangle$ is from $|c^k\rangle$ (by discarding the wavelet part outside the same region D). For the approximate energy we make a similar ansatz

$$U_{\mathrm{f}}^{\mathrm{a}} = \langle \tilde{c}^k | \hat{U}_{\mathrm{f}}^k | \tilde{s}^k \rangle, \tag{64}$$

where the matrix elements of $\hat{U}_{\mathrm{f}}^k$ are given by (37).

Let us estimate the error of the energy expression (64), i.e., its difference from (63). It follows from (60) and (62) that

$$|\tilde{c}^k\rangle = |c^k\rangle - |\breve{d}^{k-1}\rangle \tag{65}$$

and, similarly, for $|\tilde{s}^k\rangle$:

$$|\tilde{s}^k\rangle = |s^k\rangle - |\breve{b}^{k-1}\rangle. \tag{66}$$

Plugging (65) and (66) into (63) and (64), we get:

$$\begin{aligned} U^{\mathrm{a}} &= \langle c^k|\hat{U}|s^k\rangle - \langle \breve{d}^{k-1}|\hat{U}|s^k\rangle - \langle c^k|\hat{U}|\breve{b}^{k-1}\rangle + \langle \breve{d}^{k-1}|\hat{U}|\breve{b}^{k-1}\rangle,\\ U_{\mathrm{f}}^{\mathrm{a}} &= \langle c^k|\hat{U}_{\mathrm{f}}^k|s^k\rangle - \langle \breve{d}^{k-1}|\hat{U}_{\mathrm{f}}^k|s^k\rangle - \langle c^k|\hat{U}_{\mathrm{f}}^k|\breve{b}^{k-1}\rangle + \langle \breve{d}^{k-1}|\hat{U}_{\mathrm{f}}^k|\breve{b}^{k-1}\rangle. \end{aligned}$$

Therefore, the error of the adaptive energy approximation has the form

$$\begin{aligned} &\delta U^{\mathrm{a}} \equiv U^{\mathrm{a}} - U_{\mathrm{f}}^{\mathrm{a}} = \delta U_1 + \delta U_2 + \delta U_3 + \delta U_4; \qquad \delta U_1 = \langle c^k|\hat{U}|s^k\rangle - \langle c^k|\hat{U}_{\mathrm{f}}^k|s^k\rangle,\\ &\delta U_2 = \langle \breve{d}^{k-1}|\hat{U}_{\mathrm{f}}^k|s^k\rangle - \langle \breve{d}^{k-1}|\hat{U}|s^k\rangle, \quad \delta U_3 = \langle c^k|\hat{U}_{\mathrm{f}}^k|\breve{b}^{k-1}\rangle - \langle c^k|\hat{U}|\breve{b}^{k-1}\rangle,\\ &\delta U_4 = \langle \breve{d}^{k-1}|\hat{U}|\breve{b}^{k-1}\rangle - \langle \breve{d}^{k-1}|\hat{U}_{\mathrm{f}}^k|\breve{b}^{k-1}\rangle. \end{aligned} \tag{67}$$

One can show that all four error terms are small

$$\delta U^{\mathrm{a}} = \mathcal{O}(h^{2m}). \tag{68}$$

First, $\delta U_1 = \mathcal{O}(h^{2m})$ because this is the error term in the non-adaptive case. Then, let us consider the term

$$\delta U_2 = \langle \breve{d}^{k-1}|(\hat{U}_{\mathrm{f}}^k - \hat{U})|s^k\rangle.$$

One can show that it contains the non-adaptive gradients

$$\hat{U}_{\mathrm{f}}^k|s^k\rangle = \hat{U}_{\mathrm{f}}^k \sum_j s_j^k |\phi_j^k\rangle = \sum_{i,j} |\phi_i^k\rangle U_{ij}^{\mathrm{f}} s_j^k = \sum_i |\phi_i^k\rangle \frac{\partial U_{\mathrm{f}}}{\partial c_i^k}$$

and the same for $\hat{U}|s^k\rangle$. Therefore,

$$(\hat{U}_{\mathrm{f}}^k - \hat{U})|s^k\rangle = \sum_i |\phi_i^k\rangle \left(\frac{\partial U_{\mathrm{f}}}{\partial c_i^k} - \frac{\partial U}{\partial c_i^k} \right) = \mathcal{O}(h^{2m})$$

because of (43). Also, the wavelet coefficients asymptotically behave as

$$d_i^{k-1} = \mathcal{O}(h^{m+1/2}), \quad \text{so } |d^{k-1}\rangle = \mathcal{O}(h^m) \tag{69}$$

(see [30]). Therefore, $\delta U_2 = \mathcal{O}(h^{3m})$ and this term can be neglected. Since the potential energy matrices (36) and (37) are Hermitean, the same applies to the third term.

Eq. (69) also determines the asymptotic behavior of the fourth term: $\delta U_4 = \mathcal{O}(h^{2m})$. The fourth error term is formed by wavelet coefficients on the $k-1$ level, so it is the main source of error. However, it vanishes outside D. On the other hand, inside D one can use the non-adaptive error estimate (34). Thus, the main contribution to the energy error comes from the boundary of D where neither of the above arguments applies.

An interesting partial case is when the fine region D is empty. Then, all the wavelets on the finest level k are discarded, so the wavelet vector (59) is zero. Thus Eq. (60) is simplified to $|\tilde{c}^k\rangle = |c^{k-1}\rangle$, which is equivalent to using just the scaling functions on the level $k-1$. However, we still use the quadrature (10) on the level k, and evaluate the wavefunction on the grid with constant $h = 2^{-k}$, which is twice denser than the grid on which the scaling functions are defined. This is useful if we want to determine the potential energy more precisely without raising the number of the degrees of freedom in the variational minimization.

5.2. The matrix elements for the adaptive case

If one substitutes (65) and (66) directly into (63), one obtains:

$$U^{\mathrm{a}} = \langle c^{k-1}|\hat{U}|s^{k-1}\rangle + \langle \tilde{d}^{k-1}|\hat{U}|s^{k-1}\rangle + \langle c^{k-1}|\hat{U}|\tilde{b}^{k-1}\rangle + \langle \tilde{d}^{k-1}|\hat{U}|\tilde{b}^{k-1}\rangle, \tag{70}$$

$$U^{\mathrm{a}}_{\mathrm{f}} = \langle c^{k-1}|\hat{U}^k_{\mathrm{f}}|s^{k-1}\rangle + \langle \tilde{d}^{k-1}|\hat{U}^k_{\mathrm{f}}|s^{k-1}\rangle + \langle c^{k-1}|\hat{U}^k_{\mathrm{f}}|\tilde{b}^{k-1}\rangle + \langle \tilde{d}^{k-1}|\hat{U}^k_{\mathrm{f}}|\tilde{b}^{k-1}\rangle \tag{71}$$

with the elements of the exact matrix having the form:

$$u^{cc}_{ij} \equiv \langle \phi^{k-1}_i|\hat{U}|\phi^{k-1}_j\rangle = \int \phi^{k-1}_i(x)V(x)\phi^{k-1}_j(x)\,\mathrm{d}x, \quad u^{cd}_{ij} \equiv \langle \phi^{k-1}_i|\hat{U}|\psi^{k-1}_j\rangle; \quad u^{dd}_{ij} \equiv \langle \psi^{k-1}_i|\hat{U}|\psi^{k-1}_j\rangle.$$

The approximate matrix elements are a bit more difficult to derive: using the backward transformation and (37), we get

$$u^{cc\,\mathrm{f}}_{ij} \equiv \langle \phi^{k-1}_i|\hat{U}^k_{\mathrm{f}}|\phi^{k-1}_j\rangle = \sum_t V^k_t v_{t-2i} v_{t-2j} = \sum_s V^k_{2i+s} v_s v_{s-2q}, \tag{72}$$

where $q = j - i$ and a new filter is defined

$$v_s \equiv \sum_l h_l w_{s-l}. \tag{73}$$

Suppose that $V(x) = x^t$. Then, we have (38) and an analog of (39)

$$u^{cc}_{0q} = (2h)^t K_{qt}, \quad u^{cc\,\mathrm{f}}_{0q} = (2h)^t k^{\mathrm{f}}_{qt}, \quad k^{\mathrm{f}}_{qt} \equiv \sum_l (s/2)^t v_s v_{s-2q}. \tag{74}$$

For a general potential, we have (41)

$$u^{cc}_{0q} = \sum_{p=0}^{2m-1} \frac{V^{(p)}(0)}{p!}(2h)^p K_{qp} + \mathcal{O}(h^{2m}). \tag{75}$$

For other matrix elements the same formula can be used, just with wavelets instead of scaling functions in the definition (38) of K_{qp}.

If we use a variant of the Taylor series (40)

$$V^k_s = V(sh) = \sum_{p=0}^{2m-1} \frac{V^{(p)}(0)}{p!}(2h)^p (s/2)^p + \mathcal{O}(h^{2m}) \tag{76}$$

and combine it with (74), we will obtain an analog of (75)

$$u^{cc\,\mathrm{f}}_{0q} = \sum_{p=0}^{2m-1} \frac{V^{(p)}(0)}{p!}(2h)^p k^{\mathrm{f}}_{qp} + \mathcal{O}(h^{2m}). \tag{77}$$

The matrix elements containing wavelets have the same form, the only difference being that the filters g_i are used instead of h_i where appropriate.

5.3. The gradient error for the adaptive approximation

The derivatives of (70) and (71) w.r.t. c^{k-1}_i and $\tilde{d}^{k-1}_i$ have the form:

$$\frac{\partial U^{\mathrm{a}}}{\partial c^{k-1}_i} = \sum_j u^{cc}_{ij} s^{k-1}_j + \sum_j u^{cd}_{ij} \tilde{b}^{k-1}_j = \frac{\partial U}{\partial c^{k-1}_i} - \sum_j u^{cd}_{ij} \check{b}^{k-1}_j, \tag{78}$$

$$\frac{\partial U^{\mathrm{a}}}{\partial \tilde{d}^{k-1}_i} = \sum_j u^{dd}_{ij} \tilde{b}^{k-1}_j + \sum_j u^{cd}_{ji} s^{k-1}_j = \frac{\partial U}{\partial \tilde{d}^{k-1}_i} - \sum_j u^{dd}_{ij} \check{b}^{k-1}_j, \tag{79}$$

where $U = \langle c^k|\hat{U}|s^k\rangle$ is the energy with all wavelets kept, i.e., for the non-adaptive case. As in the non-adaptive case, the expressions for the gradient of a quadratic form would differ just by a factor of 2.

For the approximate energy, the gradient expression is similar. The matrix elements for the exact and approximate energy have the form (75) and (77), respectively. Since the filters K^p_q, k^p_q have finite and short length, the gradient expressions (78) and (79) are nearly local. Therefore, one can consider their features depending on the region where c^{k-1}_i is located.

In the fine region D (see (59) and the text after it) the $\check{b}^{k-1}_i$ wavelets (61) are zero. Therefore, Eqs. (78) and (79) reduce to their non-adaptive counterparts. If we apply the backward wavelet transform to them, the result will be (42). Then it follows from (43) that the difference between the gradients of the exact and approximated energy will behave like $\mathcal{O}(h^{2m})$. Also, all the arguments of Section 4.3 apply.

Now there is the region far from D where all the wavelets are discarded. We will also call it "coarse region". The wavelets (59): $\tilde{d}^{k-1}_i, \tilde{b}^{k-1}_i$ are zero there. Thus, in the coarse region (78) assumes the form

$$\frac{\partial U^{\mathrm{a}}}{\partial c^{k-1}_i} = u^{cc}_{ij} s^{k-1}_j. \tag{80}$$

The condition (79) is not applicable in the coarse region, since the $\tilde{d}^{k-1}_i$ wavelets are zero there, and the potential energy does not depend on them.

We will prove below that in the coarse region,

$$\frac{\partial U^{\mathrm{a}}}{\partial c^{k-1}_i} = \frac{\partial U^{\mathrm{a}}_{\mathrm{f}}}{\partial c^{k-1}_i} + \mathcal{O}(h^{2m+1/2}). \tag{81}$$

In the partial case when $D = \emptyset$ discussed in the end of Section 5.1, the above equation plays the role of (43).

In the general case there is also the border region between the fine and coarse ones. The error estimates (43) and (81) were derived under the assumptions that either the $\tilde{d}^k_i$ or $\check{d}^k_i$ wavelets are zero, correspondingly. Since in the border region neither of these assumptions is guaranteed, the estimates (43) and (81) may no longer hold there (and the numerical tests imply that they do not hold indeed). Thus in the border region, one has just (78) and (79), with all terms being non-zero. From the right equations of (43) and (81) one can conclude that the error at the border is bounded by the magnitude of $\check{b}^{k-1}_j$ coefficients which behave according to (69). The other, non-adaptive-like part of error behaves like $\mathcal{O}(h^{2m})$, because of (43). Therefore,

$$\frac{\partial U}{\partial c^{k-1}_i} = \frac{\partial U_{\mathrm{f}}}{\partial c^{k-1}_i} + \mathcal{O}(h^{m+1/2}), \quad \frac{\partial U}{\partial \tilde{d}^{k-1}_i} = \frac{\partial U_{\mathrm{f}}}{\partial \tilde{d}^{k-1}_i} + \mathcal{O}(h^{m+1/2})$$

in the border region, so it is the main source of error. The small energy error (68) is consistent with the above because the rapidly oscillating part of the gradient at the boundary region has small smooth component and gets drastically diminished when multiplied by a smooth function, according to (54).

In the remaining part of this section we will prove (81) in the coarse region. The proof is completely analogous to that of (43), so we will not go into much detail. First, let $V(x) = x^t$ and $\Psi(x) = x^l$. Then, it follows from (50) that

$$\frac{\partial U^{\mathrm{a}}}{\partial c^{k-1}_0} = (2h)^{t+l+1/2} M_{l+t}.$$

Let us prove that the same is true for $U^{\mathrm{a}}_{\mathrm{f}}$. Substituting the potential energy matrix from (74) and (52) for the level $k-1$ into (80) we obtain the following analog of (46)

$$\frac{\partial U^{\mathrm{a}}_{\mathrm{f}}}{\partial c^{k-1}_0} = (2h)^{t+l+1/2} a^{\mathrm{f}}_{lt}, \quad a^{\mathrm{f}}_{p,u} \equiv \sum_{t=0}^{p} C^t_p M_t \sum_q k^{\mathrm{f}}_{qu} q^{p-t}.$$

Similarly to (50), one can show that

$$a^{\mathrm{f}}_{lt} = M_{l+t}, \quad \frac{\partial U^{\mathrm{a}}_{\mathrm{f}}}{\partial c^k_0} = \frac{\partial U^{\mathrm{a}}}{\partial c^k_0} = (2h)^{l+t+1/2} M_{l+t}, \qquad l+t < 2m. \tag{82}$$

For the values $l < m$, Eq. (82) can be proved along the lines of Appendix B. Since we were unable to find an analytical proof of (47) for the remaining values of l, we checked it numerically with Mathematica for the Daubechies wavelets of the orders from 6 to 18. We checked both the extremal phase and least asymmetric families. Same as with (49), the relative error of (82) can be made arbitrarily small by increasing the precision of wavelet filters. We went down to 10^{-40}.

Combining (82) with the Taylor expansions (76) and (51) for the level $k-1$, we get an analog of (53):

$$\begin{aligned}\frac{\partial U_{\mathrm{f}}^{\mathrm{a}}}{\partial c_0^{k-1}} &= \sum_{s=0}^{2m-1} \frac{(2h)^{s+1/2}}{s!} \sum_{p=0}^{s} C_s^p \Psi^{(p)}(0) V^{(s-p)}(0) a_{p,s-p}^{\mathrm{f}} + \mathcal{O}(h^{2m+1/2}) \\ &= \sum_{s=0}^{2m-1} \frac{(2h)^{s+1/2}}{s!} G^{(s)}(0) M_s + \mathcal{O}(h^{2m+1/2}) = g_0^{k-1} + \mathcal{O}(h^{2m+1/2}) = \frac{\partial U^{\mathrm{a}}}{\partial c_0^{k-1}} + \mathcal{O}(h^{2m+1/2}). \qquad (83)\end{aligned}$$

Eq. (81) is thus proved.

6. Reducing the computational cost in the coarse region

6.1. The adaptive quadrature for the product of a scaling function and a smooth function

The approximation derived above is not numerically efficient in the coarse region. Namely, the approximate energy matrix elements are defined by (56) there. However, in the non-adaptive approximation for the level $k-1$, they would be given by (72), which requires roughly twice smaller number of calculations because the filter ω_l is shorter than v_l. The asymptotic behavior of the energy error of these two approximations is the same: $\mathcal{O}(h^{2m})$. Therefore, it would be desirable to use (72) in the coarse region.

This situation is analogous to applying the quadrature (10) on the level k to a two-scale function $G(x)$ that has "fine region" where one should use the quadrature (10) on the level k and "coarse region" where one should use it on the level $k-1$.

Thus we have an adaptive quadrature. Recall that the adaptive approximation considered in the previous sections consists of setting the wavelet coefficients in the coarse region to zero. In addition to that, the adaptive quadrature is defined on a coarser grid in the coarse region.

The question is – what quadrature should we use at the boundary of the coarse and fine regions? Our recipe is to use the fine quadrature, along with backward transformation, with the filter (73)

$$\int G(x)\phi_r^{k-1}(x)\,\mathrm{d}x = \sqrt{h}\sum_l v_l G_{l+2r}^k + \mathcal{O}(h^{M+3/2}) = \sqrt{h}\sum_s v_{s-2r} G_s^k + \mathcal{O}(h^{M+3/2})$$

and go to the coarse quadrature only at some distance from the boundary.

An alternative (and maybe more rigorous) recipe [14] is to use a non-uniform quadrature at the boundary. However,

- The non-uniform quadrature is difficult to program since the points in space near the boundary should be dealt with individually.
- Our error estimates for the potential energy quadrature hold only for the uniform case and it is not trivial to extend them onto the case of non-uniform quadrature of [14].

6.2. The energy gradient in the coarse region

At present, the approximate energy gradient in the coarse region is given by

$$\frac{\partial U^{\mathrm{a}}}{\partial c_i^{k-1}} = \sum_j u_{ij}^{cc\,\mathrm{f}} s_j^{k-1},$$

where the matrix elements are given by (72). In general, the gradient is given by (78) and (79).

The energy gradient on the $k-1$ level in the non-adaptive scheme is given by Eq. (42) with the matrix elements (37) (for the level $k-1$)

$$\frac{\partial U_{\mathrm{f}}}{\partial c_i^{k-1}} = \sum_j s_j^{k-1} U_{ij}^{\mathrm{f}k-1}. \tag{84}$$

One could define a "quasigradient" vector v_i^{k-1} with the following components:

$$v_i^{k-1} = \begin{bmatrix} \frac{\partial U_{\mathrm{f}}^{\mathrm{a}}}{\partial c_i^{k-1}} = \sum_j u_{ij}^{cc\mathrm{f}} s_j^{k-1} + \sum_j u_{ij}^{cd\mathrm{f}} \tilde{b}_j^{k-1}, & i \in D', \\ \frac{\partial U_{\mathrm{f}}}{\partial c_i^{k-1}} = \sum_j s_j^{k-1} U_{ij}^{\mathrm{f}k-1}, & i \notin D', \end{bmatrix} \tag{85}$$

where the set D' is D plus the points that are no more than a distance a (empirically, $a = 3mh$ is enough) away from D. The word "quasigradient" means that the vector (85) is assembled from gradients (78) and (84), but is not necessarily equal to the gradient of any function at all.

We can use now use this vector instead of the gradient in the iterative minimization algorithms.

The quasigradient vector (85) is a good approximation of the adaptive gradient (78). For the points $i \in D'$ their components coincide. For the points $i \notin D'$, they coincide if $V(x)$ and $\Psi(x)$ are polynomials with the sum of their degrees smaller than $2m$, because in that case both the quasigradient and the adaptive gradient coincide with the exact gradient (see (50) and (82)).

For the general potentials and wavefunctions, the gradient (78) assumes the form (83). On the other hand, the quasigradient will have the form (53) for the level $k-1$, which is identical to (83). Therefore, the quasigradient components are asymptotically close to those of the adaptive gradient (78) and its approximate version

$$\frac{\partial U_{\mathrm{f}}^{\mathrm{a}}}{\partial c_i^k} - v_i^k = \mathcal{O}(h^{2m+1/2}), \quad \frac{\partial U^{\mathrm{a}}}{\partial c_i^k} - v_i^k = \mathcal{O}(h^{2m+1/2}). \tag{86}$$

Now let us consider the energy. Eq. (64) can be rewritten in the form similar to (54)

$$U_{\mathrm{f}}^{\mathrm{a}} = \sum_i c_i^{k-1} \frac{\partial U_{\mathrm{f}}}{\partial c_i^{k-1}} + \sum_i \tilde{d}_i^{k-1} \frac{\partial U_{\mathrm{f}}}{\partial \tilde{d}_i^{k-1}}.$$

One can define a likewise energy expression based on (85)

$$U_{\mathrm{e}}^{\mathrm{a}} = \sum_i c_i^{k-1} v_i^{k-1} + \sum_i \tilde{d}_i^{k-1} \frac{\partial U_{\mathrm{f}}}{\partial \tilde{d}_i^{k-1}}, \tag{87}$$

where "e" stands fort for "efficient". Then it follows from (86) that

$$U_{\mathrm{e}}^{\mathrm{a}} = U_{\mathrm{f}}^{\mathrm{a}} + \mathcal{O}(h^{2m}), \quad U_{\mathrm{e}}^{\mathrm{a}} = U^{\mathrm{a}} + \mathcal{O}(h^{2m}).$$

Thus, the energy expression (87) is also a good approximation of (64) and (63).

Note that the gradient of (87) does not coincide with the quasigradient (85). They are different only at the boundary of D': the former behaves badly there, while the latter is smooth. This means, in particular, that simply minimizing (87) would result in a wavefunction that is not smooth at the boundary of D', but oscillates there rapidly.

The quasigradient can be seen as a projection of the gradient of (87) onto the space of functions that are smooth at the boundary of D'. One can then assume that we minimize the energy (87) in the space of such functions. This remedies the above problem of singularity at the boundary.

Alternatively, one can consider the quasigradient as an approximation of the gradient (78) and (87) as an approximation of (64). We have shown above that the error of such approximation is asymptotically small for smooth wavefunctions and potentials.

7. Charge density and products of functions

In the density functional theory one needs to express the charge density

$$\rho(x) = (\Psi_I(x))^2$$

as a linear combination of scaling functions. In the Hartree-Fock method, one uses products of functions of the form (12)

$$\Phi_I(x)\Psi_I(x) = \sum_{i,j} c_i^k s_j^k \phi_i^k(x)\phi_j^k(x) \tag{88}$$

and it would also be useful to have that product expanded in terms of scaling functions or wavelets, not in terms of scaling function products

$$\Phi_I(x)\Psi_I(x) = \sum_j f_j^k \phi_j^k(x) + \delta F(x),$$

where $\delta F(x)$ is small. In general, one can expand the product in scaling functions of the order different from m (that for $\Phi_I(x),\Psi_I(x)$), or even from some other wavelet family.

The obvious way to obtain the coefficients f_j^k is to expand the left part in scaling functions according to (6). The coefficients can then be obtained by convolution with the matrix of triple products, similarly to (20). However, then we would have the same problem as with (20): too many flops per grid point, $\sim N^2$ in one dimension and $\sim N^4$ in three dimensions, where N is the dimension of the triple-product matrix.

Fortunately, we can suggest an alternative way to get the density.

7.1. Using the average grid values of $\Phi(x)$ and $\Psi(x)$

Let us define the following coefficients:

$$F_i^k \equiv \bar{\Phi}_i^k \bar{\Psi}_i^k = \Phi(\mathrm{i}h)\Psi(\mathrm{i}h) + \mathcal{O}(h^{2m}),$$

where the average values are defined in (32).

Then one can approximate the product (88) with the function

$$F_\delta(x) = h\sum F_i^k \delta(x - \mathrm{i}h). \tag{89}$$

One can easily show that this function reproduces the multipole moments of the product (88)

$$\int F_\delta(x)x^t\,\mathrm{d}x = h\sum_j \Phi(\mathrm{j}h)\Psi(\mathrm{j}h)\mathrm{j}^t = \int \Phi_I(x)x^t\Psi_I(x)\,\mathrm{d}x + \mathcal{O}(h^{2m}) \tag{90}$$

for all integer $t \geqslant 0$. The second equality is a partial case of (34).

What is more, if $\Psi(x)$ is a polynomial of the degree l and $l + t < 2m$, then (90) is satisfied *exactly*. This is a consequence of (50).

However, the function (89) is not smooth, in contrast to the product (88) that it should approximate. To remedy this, one can go from (89) to

$$F_I(x) = \sum F_i^k \phi_i^{Ik}(x), \quad \phi_i^{Ik}(x) \equiv \phi^I(x/h - \mathrm{i}),$$

where $\phi^I(y)$ is the interpolating scaling function of the order L. Its first L moments are the same as those of a delta function [9,44]. Therefore, $F_I(x)$ satisfies (90) too (for $t < L$).

On the other hand, if $L \geqslant m$, then

$$F_I(x) = \Phi(x)\Psi(x) + \mathcal{O}(h^L) = \Phi_I(x)\Psi_I(x) + \mathcal{O}(h^m),$$

so the approximated density is close to (88) at any point.

This approximation is much faster than that the triple-product calculation. However, the moment conservation (90) is not exact. To make at least the total charge of a single electron equal to one in this scheme, one has to scale the coefficients F_i^k accordingly. One can also make a forward wavelet transformation to the scaling function coefficients on some coarser level and scale only them; this is enough to get the correct total charge.

8. Application to the harmonic oscillator

8.1. The non-adaptive case

In this section we will test the above approximations by finding the ground state of a unit mass and frequency harmonic oscillator in one dimension. We use non-periodic boundary conditions where the wavefunction was set to zero outside the interval $[-16{:}16]$. The variational ground state is obtained from Eq. (15), with the definitions (13) and (11) and $V(x) = x^2/2$. Since for the oscillator potential, (19) is exact, we can use the triple-product method (20) without invoking additional errors.

Our approximation for the potential energy has the form (35)

$$U_{\mathrm{f}}(\{c_i\}) = h\sum_s V_s^k\left(\bar{\Psi}_s^k\right)^2, \quad \bar{\Psi}_q^k = \frac{1}{\sqrt{h}}\sum_t W_{q-t}c_t^k.$$

Accordingly one can define the approximate RR functional

$$R_{\mathrm{f}}(\{c_i^k\}) = \frac{T(\{c_i^k\}) + U_{\mathrm{f}}(\{c_i^k\})}{\langle c^k|c^k\rangle}, \quad E_0^{\mathrm{f}} = \min R_{\mathrm{f}}(\{c_i^k\}), \quad |c_{\mathrm{f}}^k\rangle = \arg\min R_{\mathrm{f}}(\{c_i^k\}). \tag{91}$$

We performed minimization of (13) in the exact and approximate cases for least asymmetric Daubechies-$2m$ with $3 \leqslant m \leqslant 8$. We used the steepest descent method with diagonal preconditioning and gradient feedback. The results for Daubechies-8 and Daubechies-16 are shown in Figs. 2–5. On the x-axis we have the inverse grid interval: $h^{-1} = 2^k$.

In Figs. 2 and 3, on the y-axis we have the deviation of the variational ground state energy E_0 (15) from the exact result (which is 0.5 for the unit oscillator) and the deviation of the approximate ground state energy E_0^{f} (91) from 0.5.

Also shown is the difference of the variational and approximate ground state energies (91) and (15)

$$\delta E_{\mathrm{app}} \equiv E_0^{\mathrm{f}} - E_0,$$

which we call the approximation error. The graphs are on a double logarithmic scale, and they have a distinct linear part. The slope of the linear part is consistent with (16). For the lower orders, the approximation

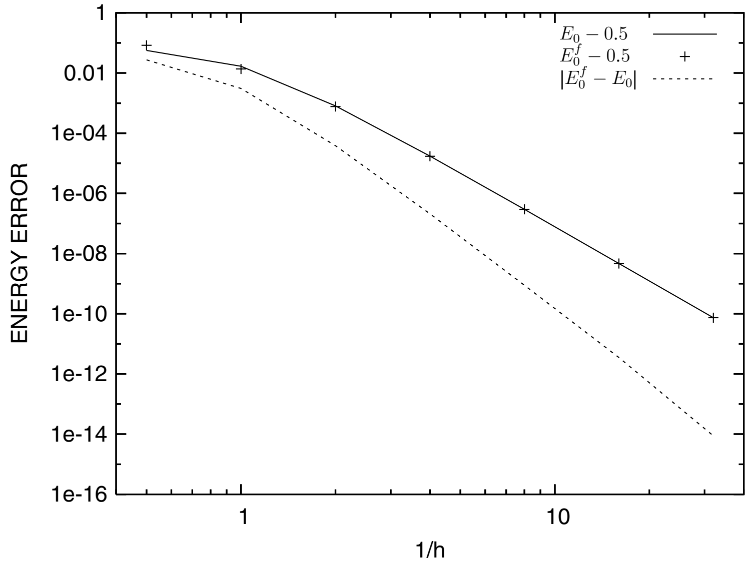

Fig. 2. The energy errors for the least asymmetric Daubechies-8 scaling functions. The slope of the variational error is 6 and that of the approximation error is 8.

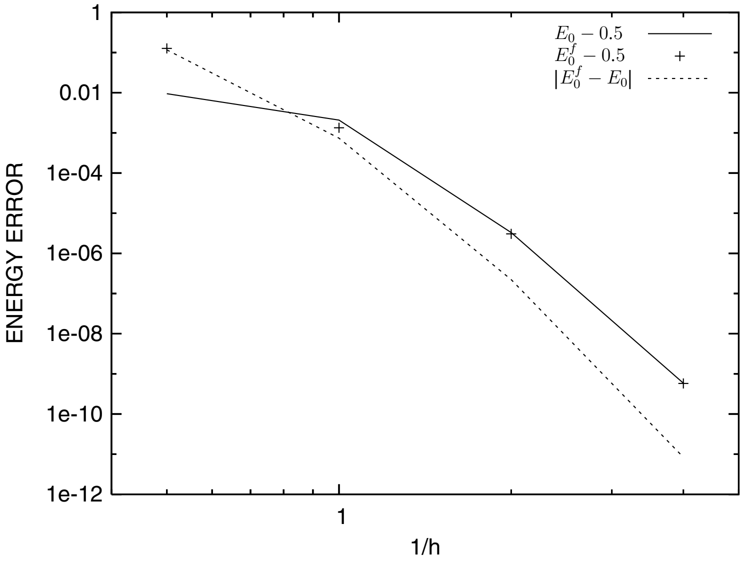

Fig. 3. The energy errors for the least asymmetric Daubechies-16 scaling functions. The slope of variational error reaches 14 but that of the approximation error reaches only 14.7.

error is one or two orders of magnitude smaller than (16). Its slope in the linear region is equal to $2m$, suggesting that

$$\delta E_{\mathrm{app}} = \mathcal{O}(h^{2m}). \tag{92}$$

This behavior is similar to (34). However, note that here we compare the energies for different (although close) quantum states $|c_0^k\rangle$ and $|c_{\mathrm{f}}^k\rangle$ defined in Eqs. (15) and (91), while in the Section 3 we computed the exact and approximate energy for the same state.

The approximation error grows with m faster than the variational error, so that for Daubechies-16 they become of the same magnitude, and have the same slope $2m - 2$.

In Figs. 4 and 5, on the y-axis we have the following quantities:

$$\delta c^k = \sqrt{\sum_i (c_{i0}^k - c_{i\mathrm{g}}^k)^2}, \quad \delta c_{\mathrm{app}}^k = \sqrt{\sum_i (c_{i0}^k - c_{i\mathrm{f}}^k)^2}. \tag{93}$$

We will call the first quantity the variational error and the second one the approximation error. The coefficients

$$c_{i\mathrm{g}}^k \equiv C \int \exp(-x^2/2)\phi_i^k(x)\,\mathrm{d}x$$

correspond to the scaling function expansion of the exact ground state of the unit oscillator in the space $\mathcal{V}_k$ (2). The factor C is chosen such that $\sum_i (c_{i\mathrm{g}}^k)^2 = 1$.

We could not find the evaluation of the quantities (93) neither in [42] nor in the previous papers describing the application of wavelets to the Schroedinger equation [6,14]. The quantity of interest in [42] was something else: the norm of the total difference between the exact ground state of (11) (in this case, a Gaussian) and $\sum_i c_i^k \phi_i^k(x)$:

$$\|\pi^{-1/4}\exp(-x^2/2) - \sum_i c_i^k \phi_i^k(x)\| = \mathcal{O}(h^m). \tag{94}$$

The above quantity includes the fine wavelet part (7) of the Gaussian that also behaves as $\mathcal{O}(h^m)$.

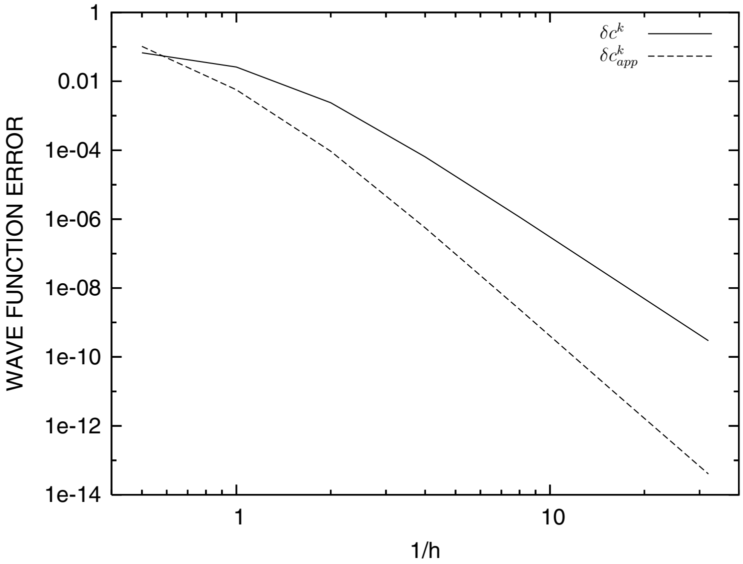

Fig. 4. The wavefunction errors for the least asymmetric Daubechies-8 scaling functions. The slope of the variational error is 6 and that of the approximation error is 8.

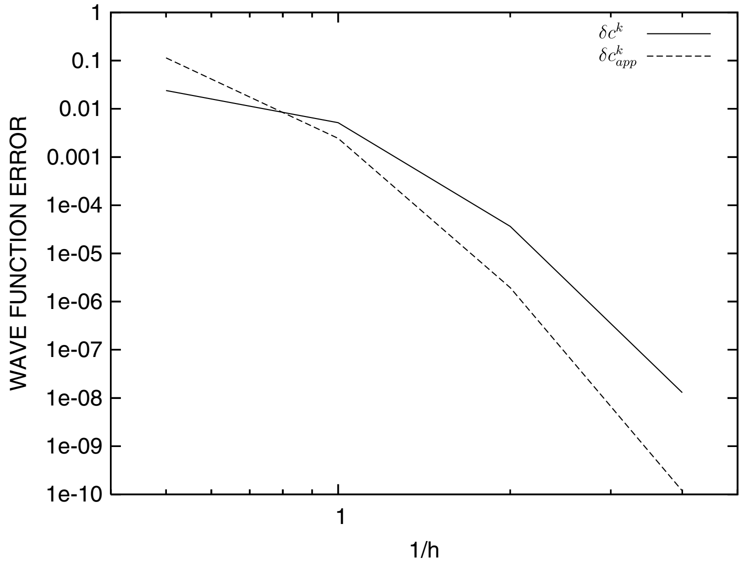

Fig. 5. The wavefunction errors for the least asymmetric Daubechies-16 scaling functions. The slope of the variational error reaches only 11.5 and that of the approximation error reaches 14.

For lower values of m, the slope of the graphs of (93) in the linear region is $2m - 2$ and $2m$ correspondingly. The approximation error in the linear region is again one or two orders of magnitude smaller than the variational error. This suggests the asymptotic behavior

$$\delta c^k = \mathcal{O}(h^{2m-2}), \quad \delta c^k_{\text{app}} = \mathcal{O}(h^{2m}). \tag{95}$$

It follows from the first estimate (95) that the wavefunction error (94) is dominated by the fine wavelet part. The second estimate of (95) means that the minima of the exact and approximate RR functionals are very close both to each other and to the projection of the exact solution onto the space of the scaling functions at resolution level k. This explains in part why the energy error (92) behaves similarly to (34).

For higher values of m the error reaches the machine precision range before the slopes reach their asymptotic values. Still, the approximation error decays faster than the variational one, and the slopes differ by 2, approximately.

For the lower values of m there is no difference between the behavior of the least asymmetric and extremal phase Daubechies scaling functions. However, for $m > 10$ the approximation error for the extremal phase Daubechies approaches the variational error. The convergence of the steepest descent iterations becomes very bad. For the symmetric family nothing of that happens. Thus for the bigger values of m our method is not applicable for the extremal phase family. This is the reason why we prefer the least asymmetric family in general.

In a practical electronic structure calculation the exact minimization is too costly numerically, but one still needs a criterion of accuracy of our approximation. We can use for that purpose the gradient of the exact RR functional (13) at the minimum of the approximate one (91).

8.2. The adaptive case

We will impose adaptivity in the following way: the minimum of the RR functional (13) will be sought in the class of coefficients $\{c_i^{k-1}, \tilde{d}_i^{k-1}\}$ such that the wavelet coefficients are zero outside the interval $[-\kappa,\kappa]$: $\tilde{d}_i^{k-1} = 0$ for $|2^{1-k}i| \geqslant \kappa$. We choose $\kappa = 1.5$ as an illustration. The resulting energy and wavefunction are:

$$E_0 = \min R(\{c_i^{k-1}, \tilde{d}_i^{k-1}\}) = \min R(\{\tilde{c}_i^k\}), \quad |\tilde{c}_0^k\rangle = \arg\min R(\{\tilde{c}_i^k\}),$$

where the coefficients $\tilde{c}_i^k$ are the backward transformation of $\{c_i^{k-1}, \tilde{d}_i^{k-1}\}$.

Their approximate counterparts are

$$E_\mathrm{f} = \min R_\mathrm{f}(\{c_i^{k-1}, \tilde{d}_i^{k-1}\}) = \min R_\mathrm{f}(\{\tilde{c}_i^k\}), \quad |\tilde{c}_\mathrm{f}^k\rangle = \arg\min R_\mathrm{f}(\{\tilde{c}_i^k\}),$$

where in the approximate RR functional the potential energy part is treated according to the Sections 5 and 6.

The graphs of the energy and wavefunction error for the (least asymmetric) Daubechies-8 and Daubechies-16 are shown in Figs. 6–9. The main differences from the non-adaptive case are:

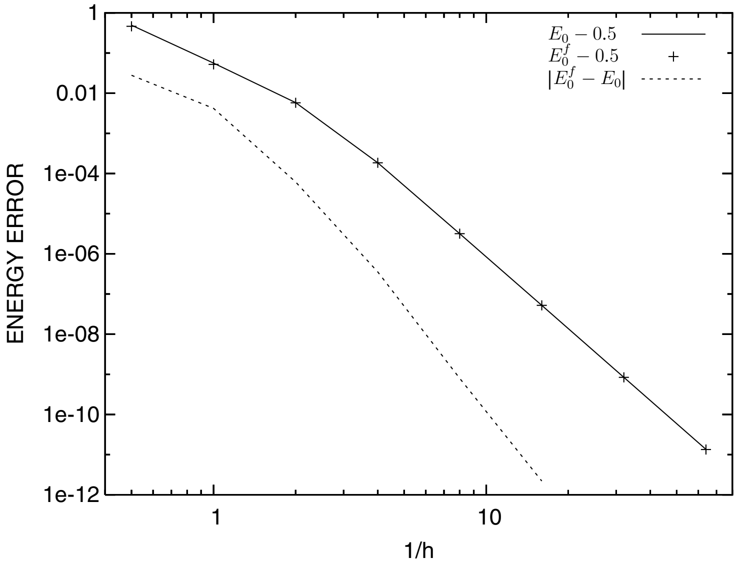

Fig. 6. The energy errors for the least asymmetric Daubechies-8 scaling functions with adaptivity. The slope of the variational error is 6 and that of the approximation error is 8. The approximation error is always smaller than the variational one.

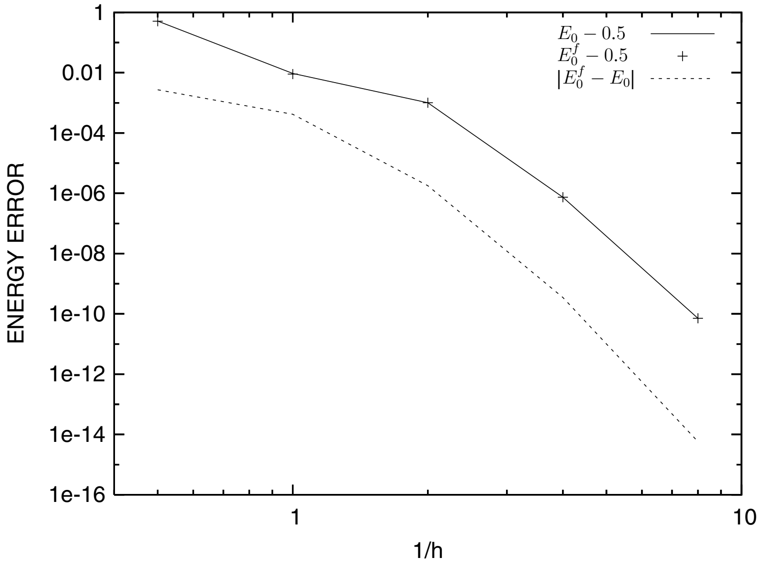

Fig. 7. The energy errors for the least asymmetric Daubechies-16 scaling functions with adaptivity. The slope of the variational error reaches 13.3 and that of the approximation error reaches 16. The approximation error is always smaller than the variational one.

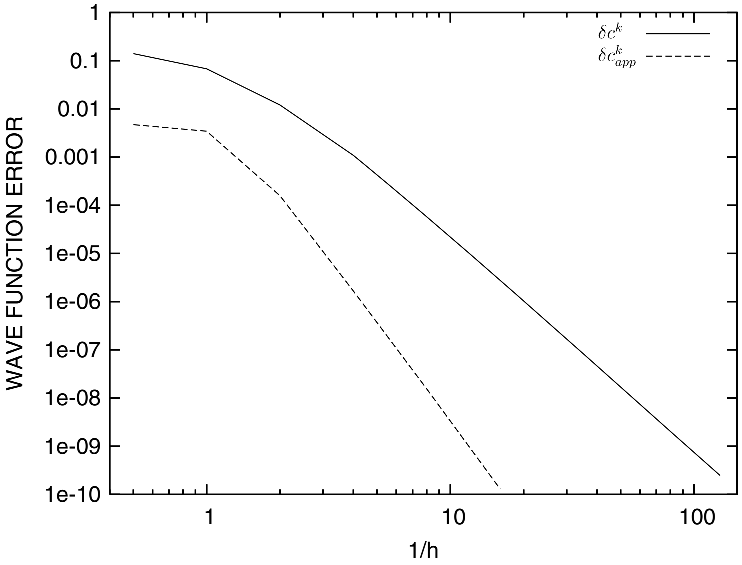

Fig. 8. The wavefunction errors for the least asymmetric Daubechies-8 scaling functions with adaptivity. The slope of the variational error is 4.5 and that of the approximation error is 7. The approximation error is always smaller than the variational one.

- The slope of the variational wavefunction error is now approximately $m + 1/2$. The asymptotical slope of the approximation error exceeds that of the variational error by 2, roughly. Both wavefunction errors are localized at the boundary.
- The approximation error both for the energy and wavefunction for small wavelet orders is an order of magnitude smaller than the variational error, now for the bigger values of h too.
- The behavior of the approximation error is improved for the small k, compared to the non-adaptive case.

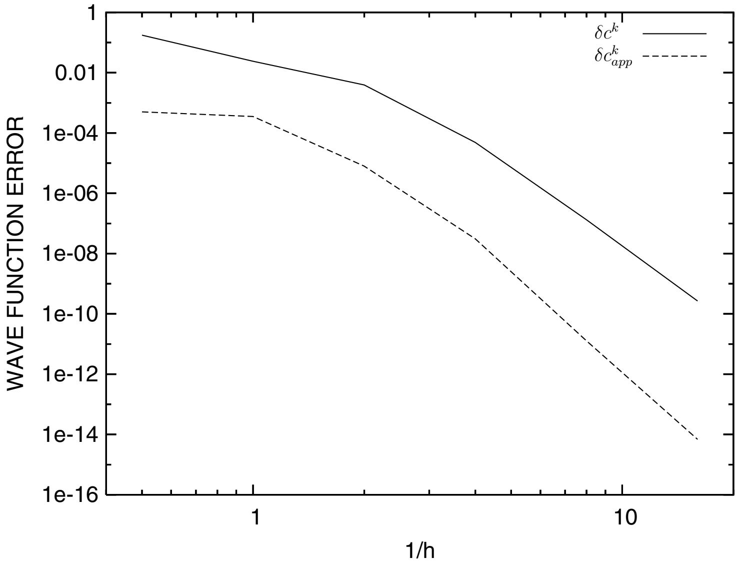

Fig. 9. The wavefunction errors for the least asymmetric Daubechies-16 scaling functions with adaptivity. The slope of the variational error reaches 9 and that of the approximation error reaches 11. The approximation error is always smaller than the variational one.

9. Conclusion

In the present work we propose a quadrature for the evaluation of the potential energy functional when the wavefunction is a wavelet approximation of some smooth function. We used the Daubechies family but the results can be extended onto others. With our algorithm, the potential energy can be calculated using only one-dimensional convolutions and filters, in contrast to the existing methods. The resulting potential energy differs only insignificantly from the exact value. The algorithm is extended onto the case of varying spatial resolution (adaptivity). As a numerical test we calculated the ground state energy and wavefunction of the harmonic oscillator in 1 dimension for the least asymmetric and extremal phase Daubechies wavelets with orders from 6 to 16. We performed the minimization of the RR functional with the potential energy calculated by our method and compared the resulting energy and wavefunction with those obtained from the fully variational minimization. In the case of the least asymmetric Daubechies family the approximate energy and wavefunction are close to the variational values. However, for the extremal phase family our method is reliable only for wavelet orders less than 10.

Our method also allows a fast calculation of a charge density for a wavefunction expressed in the wavelet basis. As a byproduct, we derived a filter for reconstruction of the grid values of a function from its Daubechies-$2m$ scaling function expansion. This reconstruction is exact for polynomials up to order $2m$ and the length of the filter is just $2m$.

The method can readily be generalized to 3 dimensions and it is already being used for three-dimensional electronic structure calculations in the framework of the BIGDFT project [45] which is a subject of future publication.

Acknowledgment

The authors are grateful to R. Schneider for useful discussions.

Appendix A. Reconstruction of grid values of wavefunction

The aim of this section is the reconstruction of the value of a smooth function at the grid points from the scaling function expansion coefficients. For this purpose we need to find the finite (and shortest possible) length filters W_l such that

$$\bar{\Psi}_r^k \equiv \frac{1}{\sqrt{h}} \sum_s W_{r-s} c_s^k = \frac{1}{\sqrt{h}} \sum_s W_q c_{r-q}^k = \Psi(hr) + \mathcal{O}(h^{2m}), \tag{96}$$

where c_i^k are given by (22) and $h = 2^{-k}$.

One can start by finding filters that satisfy (96) exactly for $\Psi(x) = x^l$, $l < 2m$ and $k = 0$

$$\sum_s W_{r-s} c_s[x^l] = r^l, \tag{97}$$

where the coefficients $c_s[x^l]$ are a generalization of the moments (8)

$$c_s[x^l] \equiv \int x^l \phi_s(x)\, \mathrm{d}x.$$

Eq. (97) means that, though with the usual interpolation (31) using Daubechies-$2m$ scaling functions one can exactly reproduce polynomials of degree not more than $m-1$, the desired filter W_l should allow us to reproduce polynomials of degrees up to $2m-1$. Thus for such high-order polynomials, (96) would be satisfied exactly, so the average value $\bar{\Psi}_r^k$ would coincide with value $\Psi_r(rh)$ of the polynomial, but *not* with the value of the scaling function expansion (31)

$$\Psi_I(rh) = \sum_i \phi_i^k(rh) \int \phi_i^k(y) \Psi(y)\, \mathrm{d}y.$$

It is enough to prove (97) at the origin only

$$\delta_l = \sum_s W_{-s} c_s[x^l], \tag{98}$$

since then it would follow that

$$\sum_s W_{r-s} c_s[x^l] = \sum_q W_{-q} c_{q+r}[x^l] = \sum_q W_{-q} c_q[(x+r)^l] = \sum_q W_{-q} \sum_{u=0}^{l} C_l^u r^{l-u} c_q[x^u] = \sum_{u=0}^{l} C_l^u r^{l-u} \delta_u = r^l, \tag{99}$$

where we changed the indices: $q = s - r$. To prove (98), we will use the following auxiliary formula:

$$c_s[x^l] \equiv \int x^l \phi_s(x)\, \mathrm{d}x = \int x^l \phi(x-s)\, \mathrm{d}x = \int (y+s)^l \phi(y)\, \mathrm{d}y = \int \sum_{u=0}^{l} C_l^u y^{l-u} s^u \phi(y)\, \mathrm{d}y = \sum_{u=0}^{l} C_l^u s^u M_{l-u}, \tag{100}$$

where C_l^u are the binomial coefficients, and M_s are the scaling function moments (8).

Substituting (100) into the right part of (98), we get

$$\sum_s W_{-s} c_s[x^l] = \sum_s W_{-s} \sum_{u=0}^{l} C_l^u s^u M_{l-u} = \sum_{u=0}^{l} C_l^u M_{l-u} \gamma_u (-1)^u = \delta_l, \tag{101}$$

$$\gamma_u \equiv \sum_s W_s s^u. \tag{102}$$

The filter W_s can thus be found by solving the systems of linear equations (101) and (102) for γ_u and then for W_s.

Now we are ready to prove (96). It is enough to do it only at the origin

$$\bar{\Psi}_0^k = \frac{1}{\sqrt{h}} \sum_s W_{-s} c_s^k = \Psi(0) + \mathcal{O}(h^{2m}). \tag{103}$$

To prove (103) one should use a version of (98) at the level k

$$\delta_l = \frac{1}{\sqrt{h}} \sum_s W_{-s} c_s^k[x^l] \tag{104}$$

that can be proved in the same way as (98) itself.

Then it remains to expand $\Psi(x)$ in the Taylor series at 0

$$\Psi(x) = \sum_{p=0}^{2m-1} \frac{\Psi^{(p)}(0)}{p!} x^p + \mathcal{O}(h^{2m})$$

and plug it into (103)

$$\frac{1}{\sqrt{h}} \sum_s W_{-s} c_s^k [\Psi(x)] = \frac{1}{\sqrt{h}} \sum_s W_{-s} c_s^k \left[\sum_{p=0}^{2m-1} \frac{\Psi^{(p)}(0)}{p!} x^p + \mathcal{O}(h^{2m}) \right]$$
$$= \frac{1}{\sqrt{h}} \sum_s W_{-s} \sum_{p=0}^{2m-1} \frac{\Psi^{(p)}(0)}{p!} c_s^k [x^p] + \mathcal{O}(h^{2m}) = \Psi(0) + \mathcal{O}(h^{2m}),$$

the last equality following from (104). Eq. (103) is thus proved.

Suppose we work with Daubechies-$2m$ scaling functions. Then, Eq. (101) is satisfied if

$$\gamma_u = M_u, \quad u = 0, \ldots, 2m - 1 \tag{105}$$

because of the equality ((B2) from [14])

$$\sum_{s=0}^{p} C_p^s M_{p-s} M_s (-1)^s = M_p^I = \delta_p, \quad p = 0, \ldots, 2m - 1, \tag{106}$$

where M_p^I are the moments of the lazy-m scaling function. The last equality is a property of the interpolating scaling functions [9,44]. Eq. (106) is satisfied both for the extremal phase and least asymmetric Daubechies wavelets.

The shortest filter with the moments (105) is (10). Thus, (34) reduces to (35). Note however that (106) is no longer satisfied for $p \geqslant 2m$ because the higher moments of an interpolating scaling function are not zero. Therefore one cannot construct a filter of degree p higher than $2m - 1$ that would also satisfy (101) for the powers of x up to the pth. Thus, one cannot improve (35) so that its error scale as $\mathcal{O}(h^s)$ with $s > 2m$.

The reconstruction scheme presented above can be generalized to find the values of $\Psi(x)$ at arbitrary points and also to find its derivatives of arbitrary order.

Appendix B. The analytical derivation of the A_{lt}, A_{lt}^{f} coefficients

In this appendix, we will present the proofs of Eqs. (47) and (48).

At first let us prove (47). Suppose, as in Section 4.3, that $V(x) = x^t$ and $\Psi(x) = x^l$. Then, combining the left equalities of (42) and (38), we get, for $l < m$,

$$\frac{\partial U}{\partial c_0^k} = \sum_j s_j^k U_{0j} = \sum_j s_j^k \int \phi^k(x) x^t \phi_j^k(x)\,\mathrm{d}x = \int \phi^k(x) x^{t+l}\,\mathrm{d}x = h^{t+l+1/2} M_{t+l}, \tag{107}$$

where we used the fact that $\sum_j s_j^k[x^l]\phi_j^k(x) = x^l$ for $l < m$. From (107) and (45), we thus get the first equality of (47). On the other hand, if $t = 0$ then (38) reduces to $U_{0j} = \delta_j$ and thus (42) has the form

$$\frac{\partial U}{\partial c_0^k} = s_0^k[x^l] = h^{l+1/2} M_l, \tag{108}$$

where the last equality is a partial case of (44). Combining (108) and (45) we get the second equality of (47)

Now let us turn to Eq. (48). Plugging (39)

$$U_{0q}^{\mathrm{f}} = h^t \sum_r r^t w_r w_{r-q}$$

into the second equality of (42), we get

$$\frac{\partial U_{\rm f}}{\partial c_0^k} = \sum_j s_j^k U_{0j}^{\rm f} = h^t \sum_r r^t w_r \sum_q w_{r-q} s_q^k [x^l] = h^t \sum_r r^t w_r \sqrt{h} (hr)^l = h^{t+l+1/2} \sum_r w_r r^{t+l} = h^{t+l+1/2} M_{t+l}, \tag{109}$$

where we have used (99) and (105). Now, Eq. (48) follows from (46) and (109).

References

[1] K. Cho, T.A. Arias, J.D. Joannopoulos, P.K. Lam, Wavelets in electronic structure calculations, Phys. Rev. Lett. 71 (1993) 1808.

[2] S. Han, K. Cho, J. Ihm, Wavelets in all-electron density-functional calculations, Phys. Rev. B 60 (1999) 1437.

[3] K. Yamaguchi, T. Mukoyama, Calculations of discrete and continuum wave functions for atoms using wavelets, Nucl. Instrum. Meth. Phys. Res. B 124 (1997) 361.

[4] I. Daubechies, Ten Lectures on Wavelets, SIAM, Philadelphia, 1992.

[5] S. Wei, M.Y. Chou, Wavelets in self-consistent electronic structure calculations, Phys. Rev. Lett. 76 (1996) 2650.

[6] B.R. Johnson, J.P. Modisette, P.J. Nordlander, J.L. Kinsey, Wavelet bases in eigenvalue problems in quantum mechanics, Chem. Phys. Lett. 250 (1996) 485.

[7] C.J. Tymczak, X.-Q. Wang, Orthonormal wavelet bases for quantum molecular dynamics, Phys. Rev. Lett. 78 (1997) 3654.

[8] P. Fischer, M. Defranceschi, Numerical solution of the Schroedinger equation in a wavelet basis for hydrogen-like atoms, SIAM J. Numer. Anal. 35 (1996) 1.

[9] G. Beylkin, On the representation of operators in bases of compactly supported wavelets, SIAM J. Numer. Anal. 29 (1992) 1716.

[10] A. Latto, H.L. Resnikoff, E. Tenenbaum, The evaluation of connection coefficients of compactly supported wavelets, Aware Technical Report AD910708, Aware Inc., Cambridge, MA, 1991.

[11] W. Dahmen, C.A. Micchelli, Using the refinement equation for evaluating integrals of wavelets, SIAM J. Numer. Anal. 30 (1993) 507.

[12] V. Perrier, M.V. Wickerhauser, Multiplication of short wavelet series using connection coefficients, in: Ka-Sing Lau (Ed.), Advances in Wavelets, Springer-Verlag, Singapore, 1999, pp. 77–101.

[13] O.V. Vasilyev, S. Paolucci, A dynamically adaptive multilevel wavelet collocation method for solving partial differential equations in a finite domain, J. Comput. Phys. 125 (1996) 498.

[14] B.R. Johnson, J.P. Modisette, P.J. Nordlander, J.L. Kinsey, Quadrature integration for orthogonal wavelet systems, J. Chem. Phys. 110 (1999) 8309.

[15] G. Deslauriers, S. Dubuc, Symmetric iterative interpolation process, Constr. Approx. 5 (1989) 49 (ISSN 0176-4276, published by Springer-Verlag, NY).

[16] T.A. Arias, Multiresolution analysis of electronic structure: semicardinal and wavelet bases, Rev. Mod. Phys. 71 (1999) 267.

[17] T.D. Engeness, T.A. Arias, Multiresolution analysis for efficient, high precision all-electron density-functional calculations, Phys. Rev. B 65 (2002) 165106.

[18] I.P. Daykov, T.A. Arias, Torkel D. Engeness, Robust ab initio calculation of condensed matter: transparent convergence through semicardinal multiresolution analysis, Phys. Rev. Lett. 90 (2003) 216402.

[19] K.S. Thygesen, M.V. Bollinger, K.W. Jacobsen, Conductance calculations with a wavelet basis set, Phys. Rev. B 67 (2002) 115404.

[20] S. Goedecker, Linear scaling electronic structure methods, Rev. Mod. Phys. 71 (1999) 1085.

[21] B. Alpert, A class of bases in L^2 for the sparse representation of integral operators, SIAM J. Math. Anal. 24 (1993) 246.

[22] B. Alpert, G. Beylkin, D. Gines, L. Vozovoi, Adaptive solution of partial differential equations in multiwavelet bases, J. Comput. Phys. 182 (2002) 149.

[23] R.J. Harrison, G.I. Fann, T. Yanai, Z. Gan, G. Beylkin, Multiresolution quantum chemistry: basic theory and initial applications, J. Chem. Phys. 121 (2004) 11587.

[24] T. Yanai, G.I. Fann, Z. Gan, R.J. Harrison, G. Beylkin, Multiresolution quantum chemistry in multiwavelet bases: HartreeFock exchange, J. Chem. Phys. 121 (2004) 6680.

[25] T. Yanai, G.I. Fann, Z. Gan, R.J. Harrison, G. Beylkin, Multiresolution quantum chemistry in multiwavelet bases: analytic derivatives for HartreeFock and density functional theory, J. Chem. Phys. 121 (2004) 2866.

[26] C.K. Chui, J.-L. Lian, A study of orthonormal multi-wavelets, Appl. Numer. Math. 20 (1996) 273.

[27] S.D. Clow, B.R. Johnson, Wavelet-basis calculation of Wannier functions, Phys. Rev. B 68 (2003) 235107.

[28] G.B. Bachelet, D.R. Hamann, M. Schulter, Phys. Rev. B 26 (1992) 4199.

[29] S. Goedecker, M. Teter, J. Hutter, Separable dual-space Gaussian pseudopotentials, Phys. Rev. B 54 (1996) 1703.

[30] W. Sweldens, R. Piessens, Quadrature formulae and asymptotic error expansions for wavelet approximations of smooth functions, SIAM J. Numer. Anal. 31 (1994) 1240.

[31] W. Sweldens, R. Piessens, Asymptotic error expansion of wavelet approximations of smooth functions II, Numer. Math. 68 (1994) 377.

[32] W. Sweldens, Construction and Applications of Wavelets in Numerical Analysis, PhD thesis, Department of Computer Science, Katholieke Universiteit Leuven, Belgium, 1994.

[33] B.R. Johnson, Multiwavelet moments and projection prefilters, IEEE Trans. Signal Process. 11 (2000) 3100.

[34] H.-J. Flad, W. Hackbusch, D. Kolb, R. Schneider, Wavelet approximation of correlated wave functions. I. Basics, J. Chem. Phys. 116 (2002) 9641.

[35] H. Luo, D. Kolb, H.-J. Flad, W. Hackbusch, T. Koprucki, Wavelet approximation of correlated wave functions. II. Hyperbolic wavelets and adaptive approximation schemes, J. Chem. Phys. 117 (2002) 3625.

[36] H.-J. Flad, W. Hackbusch, H. Luo, D. Kolb, Diagrammatic multiresolution analysis for electron correlations, Phys. Rev. B 71 (2005) 125115.

[37] I. Daubechies, A. Grossman, Y.J. Meyer, Painless non-orthogonal expansions, J. Math. Phys. 27 (1986) 12711283.

[38] B. Poirier, Using wavelets to extend quantum dynamics calculations to ten or more degrees of freedom, J. Theor. Comput. Chem. 2 (2003) 65.

[39] B. Poirier, A. Salam, Quantum dynamics calculations using symmetrized, orthogonal Weyl–Heisenberg wavelets with a phase space truncation scheme. II. Construction and optimization, J. Chem. Phys. 121 (2004) 1690.

[40] B. Poirier, A. Salam, Quantum dynamics calculations using symmetrized, orthogonal Weyl–Heisenberg wavelets with a phase space truncation scheme. III. Representations and calculations, J. Chem. Phys. 121 (2004) 1704.

[41] M. Unser, Vanishing moments and the approximation power of wavelet expansions, in: Proceedings of the 1996 IEEE International Conference on Image Processing (ICIP'96), vol. I, Lausanne, Switzerland, September 16–19, 1996, pp. 629–632.

[42] G. Strang, G.J. Fix, An Analysis of the Finite Element Method, Wellesley-Cambridge Press, 1988.

[43] S. Goedecker, Wavelets and their Application, Presses polytechniques et universitaires romandes, Lausanne, 1998.

[44] J. Tian, R.O. Wells Jr., Vanishing moments and wavelet approximation, Computer Mathematics Laboratory Report CML9501, Rice University, 1995.

[45] Available from: <http://www-drfmc.cea.fr/sp2m/L_Sim/BigDFT/index.en.html>.