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Representation of the Kato electron-electron cusp condition by wavelet-based density-operator expansions

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Since Kato proved his singularity condition for Coulomb potentials in 1957, there has been interest in the creation of wave functions that meet the prescriptions of the cusp conditions, necessary for high-precision quantum-mechanical calculations. It is well known, that wave-function expansions based on Slater determinants of one-electron functions are poorly convergent with respect to satisfying the electron-electron cusp condition. In this contribution we show that with the wavelet expansion of density operators even the local form of the electron-electron cusp condition is easily representable by Slater determinants of one-electron wavelet functions with a proper asymptotics of the expansion coefficients, which is explicitly calculated for Haar wavelets.

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I. INTRODUCTION

Bingel has published his cusp conditions [1] for density matrices based on Kato's work [2] on singularities of the *N*-electron wave function in the presence of Coulomb-like potentials. Since then it has turned out that for high-precision numerical calculations it is essential to satisfy the above requirements. However, while the nuclear cusp condition is relatively easy to fulfill, the electron-electron cusp is extremely hard to represent by Slater determinants consisting of one-electron functions. As a compromise, explicit $(\mathbf{r}_i - \mathbf{r}_j)$ dependence can be introduced to the trial *N*-electron wave function, as in, e.g., Ref. [3].

Wavelets [4] are often used in many fields of science, especially in data compression and storage. It is also applied to decomposition and analysis of wave functions in Ref. [5]. Wavelet-based multiresolution analysis is helpful in density functional theory, as well. We have introduced previously an expansion method of density operators based on Slater determinants of wavelet one-electron functions [6]. It was shown that for real physical systems both arbitrary fine and arbitrary rough details of the wave function and the density operators are negligible.

In this publication we prove that this expansion offers a natural way for representing the electron-electron cusp condition. Since wavelets are quite well localized and can be transformed into each other by application of elementary dilatations and translations, a simple equation for the cusp condition can be obtained from Bingel's formula. Using the specific set of Haar wavelets we solve this equation for spin-singlet states.

II. WAVELET EXPANSIONS

Wavelets' theory involves representing general functions in terms of simpler, fixed building blocks at different scales and positions [4].

The most powerful method of introducing wavelets is multiresolution analysis. For details, application and mathematical discussion see, e.g., Refs. [7–9]. In the following discussion notation $\mathbb{R}$ is used for the set of real numbers, whereas $\mathbb{Z}$ denotes the set of all integers.

The main idea is to introduce subspaces $\{V_m \subset L^2(\mathbb{R}) |\ m=0,1,2,\dots\}$ of square integrable functions with the following properties. Subspace V_0 is spanned by an orthonormal basis set $\{s_0(r-bl) | r\in\mathbb{R},\ l\in\mathbb{Z}\}$ obtained by equidistant shifts b of a properly defined *scaling function* s_0. Expanding an arbitrary $\varphi(r)$ by the above set of basis functions leads, of course, only to a rough, coarse-grained approximation of $\varphi(r)$. Consequently, a refinement of the basis set is necessary, which is achieved by a suitable dilatation a of the scaling function as $s_1(r)=\sqrt{a}s_0(ar)$ and by shrinking the grid distance to $a^{-1}b$, giving the subspace V_1 spanned by $\{s_1(r-a^{-1}bl) | r\in\mathbb{R}, l\in\mathbb{Z}\}$. As this step should be a refinement, a necessary condition for the subspaces is $V_0 \subset V_1$, i.e., the scaling function $s_0(r)$ at resolution level 0 should be expressed as a linear combination of the basis functions $s_{1l}(r)=\sqrt{a}s_0(ar-bl)$ at level 1.

Increasing the resolution additional subspaces $V_0\subset V_1 \subset V_2 \subset \cdots \subset V_m$ can be defined in a similar way, with an orthonormal basis of scaling functions $\{s_{ml}(r) = \sqrt{a^m} s_0(a^m r - bl) | l\in\mathbb{Z}\}$ at each level m. It can be shown that in the fine resolution limit, as $m\to\infty$, the set V_m is dense in the Hilbert space $L^2(\mathbb{R})$, if $s_0(r)$ and parameters a and b are chosen in a consistent way.

An orthonormal set of *wavelet* basis functions is defined in the orthogonal complement W_m of subspace V_{m-1} in V_m,

$$V_m = V_{m-1} \oplus W_m . \tag{1}$$

The subspace W_m carries "new details" in V_m relative to V_{m-1}. Any element of the orthonormal basis set $\{w_{ml}(r) = \sqrt{a^{m-1}} w(a^{m-1}r - bl) | l\in\mathbb{Z}\}$ of each "detail space" W_m, $(m=1,2,\dots)$ is related to a universal function $w(r)$ called *mother wavelet* by dilatation and equidistant shifts. According to this construction,

$$\bigoplus_{n=1}^{m} W_n \oplus V_0 = V_m \quad \text{and} \quad \bigoplus_{n=1}^{\infty} W_n \oplus V_0 = L^2(\mathbb{R}). \tag{2}$$

The collection of functions $\{s_{0l}(r),w_{nl}(r)|l\in\mathbb{Z};n=1,2,\ldots\}$ is an orthonormal basis in $L^2(\mathbb{R})$. At level m of resolution the space V_m approximates $L^2(\mathbb{R})$ with two equivalent orthonormal basis sets defined by $\{s_{ml}(r)|l\in\mathbb{Z}\}$ and $\{s_{0l}(r),w_{nl}(r)|l\in\mathbb{Z};n=1,2,\ldots,m\}$, respectively. There exists a unitary transformation between the two sets, which will be discussed later. The advantage of using wavelet basis sets is up to the fact that increasing the level of resolution from $m-1$ to m all previous basis functions remain unaffected, just completed by the basis set of W_m.

Wavelets have been found for many applications with various properties and in higher dimensions, too. There exist differentiable as well as completely discontinuous (fractal-like) wavelets. As an illustration we discuss here the properties of the well-known Haar wavelets with the dilatation parameter $a=2$. The Haar scaling functions are defined as

$$s_{ml}(r)=\frac{1}{\sqrt{b}}\times\begin{cases}\sqrt{2}^{m} & \text{if } r\in 2^{-m}b[l,l+1),\\ 0 & \text{else}\end{cases} \tag{3}$$

and the mth level $(m=1,2,\ldots)$ Haar wavelets are

$$w_{ml}(r)=\frac{1}{\sqrt{b}}\times\begin{cases}\sqrt{2}^{m-1} & \text{if } r\in 2^{-m+1}b\left[l,l+\frac{1}{2}\right),\\ -\sqrt{2}^{m-1} & \text{if } r\in 2^{-m+1}b\left[l+\frac{1}{2},l+1\right),\\ 0 & \text{else.}\end{cases} \tag{4}$$

If for convenience we introduce the notation $w_{0l}(r)=s_{0l}(r)$, the wavelet basis set of V_m is simply $\{w_{nl}(r)|l\in\mathbb{Z};n=0,1,2,\ldots,m\}$.

In formulating the electron-electron cusp condition spin plays an indispensable role, therefore we need to extend the basis set with spin variables. It is straightforward to introduce two-component wavelet spinors by the definition

$$w_{mls}(r,\sigma)=w_{ml}(r)\,\delta_{s\sigma} \tag{5}$$

with the possible values $\{\uparrow,\downarrow\}$ of the spin index s and spin variable σ. From the physical point of view the state w_{mls} describes an electron with spin s, localized in a confined region of order $a^{-m+1}b$ at the position $a^{-m+1}bl$. In the following considerations we will introduce the shorthand notations $\mu=(m,l,s)$, $\boldsymbol{m}=(m,l)$, and $\mathbf{x}=(r,\sigma)$.

III. DECOMPOSITION OF WAVE FUNCTIONS

As we have mentioned above, multidimensional wavelets have also been found in the literature [10–12], thus considering $\mathbf{x}=(\mathbf{r},\sigma)$ where $\mathbf{r}\in\mathbb{R}^3$ the following discussions are valid for 3D systems, as well.

Every N-particle wave function Ψ can be expanded as a linear combination of N-particle Slater determinants

$$\Psi(\mathbf{x}_1,\ldots,\mathbf{x}_N)=\sum_{\boldsymbol{\mu}} c_{\boldsymbol{\mu}}\chi_{\boldsymbol{\mu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N). \tag{6}$$

Determinants $\chi_{\boldsymbol{\mu}}$ are built from one-particle basis functions of a complete orthonormal system

$$\chi_{\boldsymbol{\mu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N)=\frac{1}{\sqrt{N!}}\begin{vmatrix} w_{\mu_1}(\mathbf{x}_1) & \ldots & w_{\mu_1}(\mathbf{x}_N)\\ \vdots & \ddots & \vdots\\ w_{\mu_N}(\mathbf{x}_1) & \ldots & w_{\mu_N}(\mathbf{x}_N)\end{vmatrix}, \tag{7}$$

where multi-index $\boldsymbol{\mu}$ denotes the set of indices $\boldsymbol{\mu}=(\mu_1,\ldots,\mu_N)$.

As any permutation of the indices $\mu_1,\ldots,\mu_N$ leads to the same N-electron function $\chi_{\boldsymbol{\mu}}$ up to a possible factor of (-1), there appear $N!$ equivalent terms in the summation (6). The complex expansion coefficients $c_{\mu_1,\mu_2,\mu_3,\ldots,\mu_N}$ are defined by taking into account the sign determined by the parity of the permutation necessary to transform the sequence $\mu_1,\mu_2,\mu_3,\ldots,\mu_N$ to a series of ordered indices, leading to

$$c_{\mu_1,\mu_2,\mu_3,\ldots,\mu_N}=-c_{\mu_2,\mu_1,\mu_3,\ldots,\mu_N}. \tag{8}$$

If for any pair of indices $\mu_i=\mu_j$, by definition the coefficient $c_{\mu_1,\mu_2,\mu_3,\ldots,\mu_N}=0$.

IV. DECOMPOSITION OF DENSITY MATRICES

A pure-state N-particle density matrix arises from the wave function of the system in the following way (see, e.g., Ref. [13]):

$$\gamma_N(\mathbf{x}_1,\ldots,\mathbf{x}_N|\mathbf{x}_1',\ldots,\mathbf{x}_N') =\Psi(\mathbf{x}_1,\ldots,\mathbf{x}_N)\Psi^*(\mathbf{x}_1',\ldots,\mathbf{x}_N'). \tag{9}$$

It is clear from definition (9) that γ_N is square integrable, hermitic, and satisfies the Pauli-principle.

Substituting Ψ by its wavelet decomposition (6), after straightforward algebraic manipulations, we arrive at the expansion

$$\begin{aligned}\gamma_N(\mathbf{x}_1,\ldots,\mathbf{x}_N|\mathbf{x}_1',\ldots,\mathbf{x}_N') &=\sum_{\boldsymbol{\mu},\boldsymbol{\nu}} a_{\boldsymbol{\mu\nu}}A_{\boldsymbol{\mu\nu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N|\mathbf{x}_1',\ldots,\mathbf{x}_N')\\ &\quad+\sum_{\boldsymbol{\mu},\boldsymbol{\nu}} b_{\boldsymbol{\mu\nu}}B_{\boldsymbol{\mu\nu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N|\mathbf{x}_1',\ldots,\mathbf{x}_N'),\end{aligned} \tag{10}$$

where we have introduced

$$\begin{aligned}A_{\boldsymbol{\mu\nu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N|\mathbf{x}_1',\ldots,\mathbf{x}_N') &=\chi_{\boldsymbol{\mu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N)\chi_{\boldsymbol{\nu}}^*(\mathbf{x}_1',\ldots,\mathbf{x}_N')\\ &\quad+\chi_{\boldsymbol{\nu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N)\chi_{\boldsymbol{\mu}}^*(\mathbf{x}_1',\ldots,\mathbf{x}_N'),\end{aligned} \tag{11}$$

$$B_{\boldsymbol{\mu\nu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N|\mathbf{x}'_1,\ldots,\mathbf{x}'_N)$$
$$=i[\chi_{\boldsymbol{\mu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N)\chi^*_{\boldsymbol{\nu}}(\mathbf{x}'_1,\ldots,\mathbf{x}'_N)$$
$$-\chi_{\boldsymbol{\nu}}(\mathbf{x}_1,\ldots,\mathbf{x}_N)\chi^*_{\boldsymbol{\mu}}(\mathbf{x}'_1,\ldots,\mathbf{x}'_N)] \qquad (12)$$

as a set of basis functions for γ_N. As it easily follows from definitions (11) and (12) these basis functions meet the symmetry properties of γ_N, i.e., they are hermitic $A_{\boldsymbol{\mu\nu}}=A^\dagger_{\boldsymbol{\mu\nu}}$, $B_{\boldsymbol{\mu\nu}}=B^\dagger_{\boldsymbol{\mu\nu}}$, satisfy the Pauli principle, and additionally $A_{\boldsymbol{\mu\nu}}=A_{\boldsymbol{\nu\mu}}$, $B_{\boldsymbol{\mu\nu}}=-B_{\boldsymbol{\nu\mu}}$. All $a_{\boldsymbol{\mu\nu}}$ and $b_{\boldsymbol{\mu\nu}}$ are real numbers satisfying the properties

$$a_{\boldsymbol{\mu\nu}}=a_{\boldsymbol{\nu\mu}}, \quad a_{\boldsymbol{\mu\mu}}\geqslant 0; \quad b_{\boldsymbol{\mu\nu}}=-b_{\boldsymbol{\nu\mu}}, \quad b_{\boldsymbol{\mu\mu}}=0. \qquad (13)$$

Reduced density matrices are originated from the N-particle density matrix by a consecutive reduction of degrees of freedom [14]. One step of the process is defined by the partial trace operation

$$\gamma_{N-1}(\mathbf{x}_1,\ldots,\mathbf{x}_{N-1}|\mathbf{x}'_1,\ldots,\mathbf{x}'_{N-1})$$
$$=\int \gamma_N(\mathbf{x}_1,\ldots,\mathbf{x}_{N-1},\mathbf{x}_N|\mathbf{x}'_1,\ldots,\mathbf{x}'_{N-1},\mathbf{x}_N)d\mathbf{x}_N, \qquad (14)$$

where $\int\gamma_{N-1}(\mathbf{x}_1,\ldots,\mathbf{x}_{N-1}|\mathbf{x}_1,\ldots,\mathbf{x}_{N-1})d\mathbf{x}_1\cdots d\mathbf{x}_{N-1}=1$ was applied as a normalization condition. Step (14) is also called *contraction*. As the partial trace operation is linear, contraction is carried out on individual basis functions $A_{\boldsymbol{\mu\nu}}$ and $B_{\boldsymbol{\mu\nu}}$ of Eq. (10). Straightforward calculations show that contraction transforms the subspace spanned by the N-particle functions $A_{\boldsymbol{\mu\nu}}$ into the subspace generated by the $(N-1)$-particle basis functions $A_{\boldsymbol{\mu\nu}}$. A similar statement holds for the contraction of the subspace of $B_{\boldsymbol{\mu\nu}}$ functions as well. The consequence of this fact is that any reduced (ensemble) density matrix can be expanded in the form (10) with real expansion coefficients, that meet the properties of equations (13).

V. THE BINGEL FORMULA

The electron-electron cusp condition reflects a singularity of the wave function, and consequently, the spin-traced two-particle reduced density matrix

$$\gamma_2^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}'_1,\mathbf{r}'_2)=\sum_{\sigma_1,\sigma_2}\gamma_2(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2|\mathbf{r}'_1,\sigma_1,\mathbf{r}'_2,\sigma_2) \qquad (15)$$

of the N-electron system. The symmetry properties of γ_2^s follow from that of γ_2

$$\gamma_2^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}'_1,\mathbf{r}'_2)=\gamma_2^s(\mathbf{r}_2,\mathbf{r}_1|\mathbf{r}'_2,\mathbf{r}'_1)=[\gamma_2^s(\mathbf{r}'_1,\mathbf{r}'_2|\mathbf{r}_1,\mathbf{r}_2)]^*. \qquad (16)$$

Note, however, that generally

$$\gamma_2^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}'_1,\mathbf{r}'_2)\neq-\gamma_2^s(\mathbf{r}_2,\mathbf{r}_1|\mathbf{r}'_1,\mathbf{r}'_2) \qquad (17)$$

except for the case if all electrons carry the same spin (ferromagneticlike states with total spin $S=N/2$). Introducing the new variables

$$\mathbf{R}=\frac{\mathbf{r}_1+\mathbf{r}_2}{2}, \quad \mathbf{R}'=\frac{\mathbf{r}'_1+\mathbf{r}'_2}{2},$$
$$\mathbf{u}=\mathbf{r}_1-\mathbf{r}_2, \quad \mathbf{u}'=\mathbf{r}'_1-\mathbf{r}'_2, \qquad (18)$$

Bingel has shown [1] for an N-electron system with total spin $S<N/2$ that Kato's condition [2] implies if both $\mathbf{u}$ and $\mathbf{u}'$ are small enough

$$\gamma_2^s\left(\mathbf{R}+\frac{\mathbf{u}}{2},\mathbf{R}-\frac{\mathbf{u}}{2}\middle|\mathbf{R}'+\frac{\mathbf{u}'}{2},\mathbf{R}'-\frac{\mathbf{u}'}{2}\right)$$
$$\approx\gamma_2^s(\mathbf{R},\mathbf{R}|\mathbf{R}',\mathbf{R}')\left(1+\frac{|\mathbf{u}|}{2}+\frac{|\mathbf{u}'|}{2}\right)+\mathbf{u}\mathbf{d}^*+\mathbf{u}'\mathbf{d}+\cdots, \qquad (19)$$

where $\mathbf{d}$ is not determined by the Coulombic singularity. According to Eq. (17), $\gamma_2^s(\mathbf{R},\mathbf{R}|\mathbf{R}',\mathbf{R}')\neq 0$ in the general case (a statement that would not hold for spinless particles). Due to the presence of the absolute value of vectors $\mathbf{u}$ and $\mathbf{u}'$ in equation (19) a discontinuity appears in the derivative of γ_2 that can be conveniently expressed in a compact form by the spherical average of the diagonal element of γ_2^s

$$\overline{\gamma}(\mathbf{R},u)=\frac{1}{4\pi}\int\gamma_2^s\left(\mathbf{R}+\frac{\mathbf{u}}{2},\mathbf{R}-\frac{\mathbf{u}}{2}\middle|\mathbf{R}+\frac{\mathbf{u}}{2},\mathbf{R}-\frac{\mathbf{u}}{2}\right)$$
$$\times\sin\theta_{\mathbf{u}}d\theta_{\mathbf{u}}d\phi_{\mathbf{u}}, \qquad (20)$$

where $u=|\mathbf{u}|$ and $\theta_{\mathbf{u}},\phi_{\mathbf{u}}$ are the polar angles of the vector $\mathbf{u}$. As in the spherical average $\mathbf{u}$ vanishes, we get from Eq. (19) the linear approximation

$$\overline{\gamma}(\mathbf{R},u)\approx\gamma_2^s(\mathbf{R},\mathbf{R}|\mathbf{R},\mathbf{R})(1+u). \qquad (21)$$

This requirement on the two-electron density matrix is local in the sense that the above equality must hold at any particular point $\mathbf{R}$ of the space. Numerical checks [15,16] of many-electron wave functions usually test the global fulfilment of a space averaged form of Eq. (21). In this paper, however, we give an explicit construction of the two-particle density matrix that satisfies the much detailed local form (21) of the cusp condition.

Expression (21) shows in a natural way that the electron-electron cusp condition is a typical appearance of multiplicative superstructures found by us previously [17] for the electron density [i.e., $\gamma_1^s(\mathbf{R}|\mathbf{R})$] in extended (near-mesoscopic) molecular and solid-state systems. The particular feature of these densities is the different shape characteristics of the states shown at different length scales, i.e., changing the resolution of observations results in a variation of the overall behavior of the density. Here, the overall behavior of γ_2^s is determined by the values $\gamma_2^s(\mathbf{R},\mathbf{R}|\mathbf{R},\mathbf{R})$ at the center-of-mass $\mathbf{R}$ of the electron pairs, whereas at resolutions where the separation u of the pairs is recognizable the factor $(1+u)$ modifies the value of γ_2^s.

VI. CUSP CONDITION FOR DENSITY-MATRIX EXPANSIONS

Observing that spin tracing (15) is a linear operation, it can be executed individually on each two-electron basis functions $A_{\boldsymbol{\mu\nu}}$ and $B_{\boldsymbol{\mu\nu}}$ of the expansion (10). Consequently,

$$\gamma_2^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')=\sum_{\boldsymbol{\mu},\boldsymbol{\nu}}[a_{\boldsymbol{\mu\nu}}A_{\boldsymbol{\mu\nu}}^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2') + b_{\boldsymbol{\mu\nu}}B_{\boldsymbol{\mu\nu}}^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')] \qquad (22)$$

with $\boldsymbol{\mu}=(\mu_1,\mu_2)$ and $\boldsymbol{\nu}=(\nu_1,\nu_2)$. In the summation all indices run independently, without restrictions, for all possible values, resulting $(2!)^2$ equivalent terms. The relations between the expansion coefficients are defined similarly to Eq. (8) as

$$a_{\mu_1,\mu_2;\nu_1,\nu_2}=-a_{\mu_2,\mu_1;\nu_1,\nu_2}=-a_{\mu_1,\mu_2;\nu_2,\nu_1}=a_{\mu_2,\mu_1;\nu_2,\nu_1},$$
$$b_{\mu_1,\mu_2;\nu_1,\nu_2}=-b_{\mu_2,\mu_1;\nu_1,\nu_2}=-b_{\mu_1,\mu_2;\nu_2,\nu_1}=b_{\mu_2,\mu_1;\nu_2,\nu_1},$$
$$a_{\mu,\mu;\nu_1,\nu_2}=a_{\mu_1,\mu_2;\nu,\nu}=b_{\mu,\mu;\nu_1,\nu_2}=b_{\mu_1,\mu_2;\nu,\nu}=0. \qquad (23)$$

The spin-traced density-matrix basis functions $A_{\boldsymbol{\mu\nu}}^s$ and $B_{\boldsymbol{\mu\nu}}^s$ are formed in analogy with Eq. (15). Using definitions (11), (7) and the two-component spinor basis set $w_{\boldsymbol{m}s}(\mathbf{r},\sigma)=w_{\boldsymbol{m}}(\mathbf{r})\delta_{s\sigma}$ spin tracing results in

$$A_{\boldsymbol{\mu\nu}}^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')=\vartheta^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\delta_{s_1t_1}\delta_{s_2t_2} - \vartheta^A_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\delta_{s_2t_1}\delta_{s_1t_2} \qquad (24)$$

with $\boldsymbol{\mu}=(\mu_1,\mu_2)=(\boldsymbol{m}_1s_1,\boldsymbol{m}_2s_2)$ and $\boldsymbol{\nu}=(\nu_1,\nu_2)=(\boldsymbol{n}_1t_1,\boldsymbol{n}_2t_2)$, respectively. The functions ϑ^A were introduced as

$$\begin{aligned}\vartheta^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')=\tfrac{1}{2}[&w_{\boldsymbol{m}_1}(\mathbf{r}_1)w_{\boldsymbol{m}_2}(\mathbf{r}_2)w^*_{\boldsymbol{n}_1}(\mathbf{r}_1')w^*_{\boldsymbol{n}_2}(\mathbf{r}_2')\\&+w_{\boldsymbol{m}_2}(\mathbf{r}_1)w_{\boldsymbol{m}_1}(\mathbf{r}_2)w^*_{\boldsymbol{n}_2}(\mathbf{r}_1')w^*_{\boldsymbol{n}_1}(\mathbf{r}_2')\\&+w_{\boldsymbol{n}_1}(\mathbf{r}_1)w_{\boldsymbol{n}_2}(\mathbf{r}_2)w^*_{\boldsymbol{m}_1}(\mathbf{r}_1')w^*_{\boldsymbol{m}_2}(\mathbf{r}_2')\\&+w_{\boldsymbol{n}_2}(\mathbf{r}_1)w_{\boldsymbol{n}_1}(\mathbf{r}_2)w^*_{\boldsymbol{m}_2}(\mathbf{r}_1')\\&\times w^*_{\boldsymbol{m}_1}(\mathbf{r}_2')].\end{aligned} \qquad (25)$$

Similar considerations lead to

$$B_{\boldsymbol{\mu\nu}}^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')=\vartheta^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\delta_{s_1t_1}\delta_{s_2t_2} - \vartheta^B_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\delta_{s_2t_1}\delta_{s_1t_2}, \qquad (26)$$

where

$$\begin{aligned}\vartheta^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')=\frac{i}{2}[&w_{\boldsymbol{m}_1}(\mathbf{r}_1)w_{\boldsymbol{m}_2}(\mathbf{r}_2)w^*_{\boldsymbol{n}_1}(\mathbf{r}_1')w^*_{\boldsymbol{n}_2}(\mathbf{r}_2')\\&+w_{\boldsymbol{m}_2}(\mathbf{r}_1)w_{\boldsymbol{m}_1}(\mathbf{r}_2)w^*_{\boldsymbol{n}_2}(\mathbf{r}_1')w^*_{\boldsymbol{n}_1}(\mathbf{r}_2')\\&-w_{\boldsymbol{n}_1}(\mathbf{r}_1)w_{\boldsymbol{n}_2}(\mathbf{r}_2)w^*_{\boldsymbol{m}_1}(\mathbf{r}_1')w^*_{\boldsymbol{m}_2}(\mathbf{r}_2')\\&-w_{\boldsymbol{n}_2}(\mathbf{r}_1)w_{\boldsymbol{n}_1}(\mathbf{r}_2)w^*_{\boldsymbol{m}_2}(\mathbf{r}_1')\\&\times w^*_{\boldsymbol{m}_1}(\mathbf{r}_2')].\end{aligned} \qquad (27)$$

All functions $A_{\boldsymbol{\mu\nu}}^s$, $B_{\boldsymbol{\mu\nu}}^s$, $\vartheta^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}$, and $\vartheta^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}$ meet the symmetry properties (16) and (17) of γ_2^s. Furthermore, we have the index symmetries

$$\begin{aligned}\vartheta^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')&=\vartheta^A_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_2\boldsymbol{n}_1}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\\&=\vartheta^A_{\boldsymbol{n}_1\boldsymbol{n}_2\boldsymbol{m}_1\boldsymbol{m}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2'),\\\vartheta^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')&=\vartheta^B_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_2\boldsymbol{n}_1}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\\&=-\vartheta^B_{\boldsymbol{n}_1\boldsymbol{n}_2\boldsymbol{m}_1\boldsymbol{m}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2'),\end{aligned} \qquad (28)$$

but

$$\vartheta^{A,B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')\neq-\vartheta^{A,B}_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2'). \qquad (29)$$

Let us substitute expressions (25) and (27) into equation (22). Since summation indices run independently, exchanging of $\boldsymbol{m}_1$ for $\boldsymbol{m}_2$, as well as s_1 for s_2 in some terms is possible, resulting

$$\gamma_2^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')=\sum_{\substack{\boldsymbol{m}_1,\boldsymbol{m}_2\\\boldsymbol{n}_1,\boldsymbol{n}_2}}[g^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\vartheta^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2') + g^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\vartheta^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')], \qquad (30)$$

where

$$g^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}=2\sum_{s_1s_2}a_{\boldsymbol{m}_1s_1,\boldsymbol{m}_2s_2;\boldsymbol{n}_1s_1,\boldsymbol{n}_2s_2},$$
$$g^B_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}=2\sum_{s_1s_2}b_{\boldsymbol{m}_1s_1,\boldsymbol{m}_2s_2;\boldsymbol{n}_1s_1,\boldsymbol{n}_2s_2}. \qquad (31)$$

Here we have used the symmetry properties (23). Expression (30) indicates that ϑ^A and ϑ^B serve as natural basis functions for expanding spin-traced density matrices. The expansion coefficients g^A and g^B show the index symmetries

$$g^A_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}=g^A_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_2\boldsymbol{n}_1}=g^A_{\boldsymbol{n}_1\boldsymbol{n}_2\boldsymbol{m}_1\boldsymbol{m}_2},$$

$$g^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}=g^{B}_{\boldsymbol{m}_2\boldsymbol{m}_1\boldsymbol{n}_2\boldsymbol{n}_1}=-g^{B}_{\boldsymbol{n}_1\boldsymbol{n}_2\boldsymbol{m}_1\boldsymbol{m}_2} \tag{32}$$

as a consequence of Eqs. (13) and (23).

We define the spherical average $\overline{\vartheta}^{A}(\mathbf{R},u)$ and $\overline{\vartheta}^{B}(\mathbf{R},u)$ of the diagonal elements of ϑ^{A} and ϑ^{B}, similarly to step (20). Thus, the cusp condition (21) turns into

$$\begin{aligned}\sum_{\substack{\boldsymbol{m}_1\boldsymbol{m}_2\\ \boldsymbol{n}_1\boldsymbol{n}_2}} &[g^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\overline{\vartheta}^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{R},u)+g^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\overline{\vartheta}^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{R},u)]\\ &\approx(1+u)\sum_{\substack{\boldsymbol{m}_1\boldsymbol{m}_2\\ \boldsymbol{n}_1\boldsymbol{n}_2}}[g^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\overline{\vartheta}^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{R},0)\\ &\qquad+g^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\overline{\vartheta}^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{R},0)].\end{aligned} \tag{33}$$

Frequently, real valued basis functions are used. In these cases, according to definition (12), the diagonal elements

$$\begin{aligned}&B_{\boldsymbol{\mu}\boldsymbol{\nu}}(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2|\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2)\\ &\quad=2\ \mathrm{Im}\{\chi^{*}_{\boldsymbol{\mu}}(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2)\chi_{\boldsymbol{\nu}}(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2)\}=0,\end{aligned} \tag{34}$$

thus $\overline{\vartheta}^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(\mathbf{R},u)=0$.

VII. EXAMPLE: HAAR WAVELET EXPANSION IN ONE-DIMENSIONAL SYSTEMS

The spherical average of any function $f(\mathbf{u})$ for a one-dimensional vector $\mathbf{u}$ turns simply into

$$\overline{f}(u)=\frac{f(\mathbf{u})+f(-\mathbf{u})}{2}=\frac{f(u)+f(-u)}{2}. \tag{35}$$

The symmetry properties (16) of ϑ^{A} and ϑ^{B} involve

$$\overline{\vartheta}^{A,B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(R,u)=\vartheta^{A,B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\left(R+\frac{u}{2},R-\frac{u}{2}\middle|R+\frac{u}{2},R-\frac{u}{2}\right), \tag{36}$$

consequently, the cusp condition (33) can be rewritten using the basis functions $\vartheta^{A,B}$ directly.

Multiresolution analysis naturally offers an infinitely dense grid, thus it is sufficient to satisfy the cusp condition (33) in the grid points only. Since the grid points are equivalent, we choose $R=0$. Using the complete system of real Haar wavelets, only expressions $\overline{\vartheta}^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(0,u)=\vartheta^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(v,-v|v,-v)$ with $v=|u|/2$ need to be evaluated, and all $\overline{\vartheta}^{B}=0$ based on the note at the end of the previous section. The fundamental problem is finding appropriate expansion coefficients $g^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}$ for which the form

$$\begin{aligned}&\sum_{\substack{\boldsymbol{m}_1\boldsymbol{m}_2\\ \boldsymbol{n}_1\boldsymbol{n}_2}} g^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\vartheta^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(v,-v|v,-v)\\ &\quad\approx(1+2v)\sum_{\substack{\boldsymbol{m}_1\boldsymbol{m}_2\\ \boldsymbol{n}_1\boldsymbol{n}_2}} g^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}\vartheta^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}(0,0|0,0)\end{aligned} \tag{37}$$

of the cusp condition is satisfied. As a first step we will solve this task at a given resolution level m by using the scaling function basis set (3). At this level of approximation

$$|u|\approx\sum_{l} h_{ml}s_{ml}(u) \tag{38}$$

with

$$h_{ml}=\int|u|s^{*}_{ml}(u)du=(2^{-m}b)^{3/2}|l+\tfrac{1}{2}|. \tag{39}$$

Clearly, $(2^{-m}b)|l+\frac{1}{2}|$ is the distance of the center of basis function s_{ml} from the origin, or in other words, the average value of $|u|$ in the domain of s_{ml}, and $(2^{-m}b)^{-1/2}$ is the value of the same basis function.

For $v\in(2^{-m}bl,2^{-m}b(l+1))$

$$\begin{aligned}s_{ml'}(v)&=(2^{-m}b)^{-1/2}\delta_{l,l'},\\ s_{ml'}(-v)&=(2^{-m}b)^{-1/2}\delta_{-l-1,l'}.\end{aligned} \tag{40}$$

Substituting these values into Eq. (25) the left-hand side of equation (37)

$$\begin{aligned}&(2^{-m}b)^{-2}(g^{A,m}_{l,-l-1,l,-l-1}+g^{A,m}_{-l-1,l,-l-1,l})\\ &\quad=2(2^{-m}b)^{-2}g^{A,m}_{l,-l-1,l,-l-1}.\end{aligned} \tag{41}$$

Here we introduced the shorthand notation $g^{A,m}_{l_1l_2k_1k_2}=g^{A}_{ml_1,ml_2;mk_1,mk_2}$ and used the symmetry property (32). Similar considerations and application of Eq. (39) on the right-hand side of Eq. (37) lead to the condition

$$g^{A,m}_{l,-l-1;l,-l-1}\approx[1+2|l+\tfrac{1}{2}|\times2^{-m}b]g^{A,m}_{0000}. \tag{42}$$

As we see, the spherical average of the cusp condition (21) does not provide information on coefficients $g^{A,m}_{l_1l_2k_1k_2}$ with arbitrary index combination.

However, similarly as above but with more tedious calculations, we can have a solution of the universal form of Bingel's condition (19) for $R=(2^{-m}b)L$, $u/2=v\in(2^{-m}bl,2^{-m}b(l+1))$ and $R'=(2^{-m}b)K$, $u'/2=v'\in(2^{-m}bk,2^{-m}b(k+1))$. Evaluating the level m basis functions by Eq. (40) and substituting to definitions (25) and (27), as well as using the index symmetries (32) we arrive at

$$\begin{aligned}g^{A,B,m}_{L+l,L-l-1;K+k,K-k-1}&\approx[1+(|l+\tfrac{1}{2}|+|k+\tfrac{1}{2}|)\\ &\quad\times2^{-m}b]g^{A,B,m}_{LLKK}.\end{aligned} \tag{43}$$

This approximation is valid independently both for A and B type coefficients, as the separation of Eq. (19) to real and imaginary parts corresponds to a separation of the cusp condition to independent ϑ^A and ϑ^B expressions. Although, Eq. (43) holds for odd l_1-l_2 and k_1-k_2 index differences, a generalization for arbitrary index values is straightforward in the form

$$g^{A,B,m}_{l_1 l_2 k_1 k_2} \approx \left[1+\tfrac{1}{2}\left(|l_1-l_2|+|k_1-k_2|\right)\times 2^{-m} b\right] g^{A,B,m}_{LLKK}, \tag{44}$$

where $L=[(l_1+l_2+1)/2]$ and $K=[(k_1+k_2+1)/2]$ with $[x]$ meaning the integer part of x. The obtained formula shows that due to Bingel's condition the singular part of the density matrix is completely determined by the elements $g^{A,B,m}_{LLKK}$.

Expression (44) describes the singular behavior of the expansion coefficients g^A and g^B, but according to Eq. (19) some additional terms depending "smoothly" on (l_1-l_2) and (k_1-k_2) do also appear in a linear approximation. The cusp condition itself does not provide information on the values of $g^{A,m}_{LLKK}$ and $g^{B,m}_{LLKK}$, except that they must depend "smoothly" on the indices L and K. Generally, these parameters can be used for energy minimization, with restrictions only on the diagonal elements, as $g^{A,m}_{LLLL}$ is strictly connected to the electron density and $g^{B,m}_{LLLL}=0$ by Eq. (32).

The electron-electron cusp is a consequence of the Coulomb interaction between them, which can be approximately described as a simple two-electron problem in a background of an almost constant external potential. Therefore, the domain of validity of Eq. (44) is expected to be in the order of $2a_0$, where a_0 is the Bohr radius and the factor 2 appears due to the reduced mass of two electrons. The normalization condition on γ_2, however, prescribes [6] a fast (probably exponential) long-range decay of the coefficients $g^{A,m}$ and $g^{B,m}$. A numerical study on the average behavior of the two-particle correlation [16] confirms these expectations, with an approximate linear dependence in the range of $|u|<a_0$.

VIII. TRANSITION BETWEEN COARSE AND FINE REPRESENTATIONS

As increasing the resolution level m to $m+1$ should be a refinement of the representation, the scaling functions s_{ml} is necessarily a linear combination of $s_{(m+1)l'}$, known as *refinement equation* [4] for $m=0$

$$s_{0l}(r)=\sum_{l'} 2^{-1/2} p_{l'-2l}\, s_{1l'}(r), \tag{45}$$

where the set of expansion coefficients p_k defines the scaling function itself. In the case of Haar functions, e.g., $p_0=p_1=1$ and $p_k=0$ otherwise. It can also be shown that for the refining wavelets

$$w_{1l}(r)=\sum_{l'} 2^{-1/2} q_{l'-2l}\, s_{1l'}(r), \tag{46}$$

with $q_k=(-1)^k p^*_{-k+1}$, which gives for Haar wavelets $q_0=1$, $q_1=-1$ and zero in all other cases.

The fine→coarse transition (45) and (46) is a unitary transformation of the basis functions, which has the inverse in the coarse→fine direction

$$\begin{aligned} s_{1l'}(r) &= \sum_{l} 2^{-1/2}\left[p^*_{l'-2l}\, s_{0l}(r) + q^*_{l'-2l}\, w_{1l}(r)\right] \\ &= \sum_{l} \sum_{m=0,1} U^{ml}_{l'}\, w_{ml}(r), \end{aligned} \tag{47}$$

with

$$U^{0l}_{l'} = 2^{-1/2} p^*_{l'-2l} \quad \text{and} \quad U^{1l}_{l'} = 2^{-1/2} q^*_{l'-2l}. \tag{48}$$

Note, that we have applied here our previous convention $w_{0l}=s_{0l}$.

In order to represent γ_2^s at resolution level $m=1$ we can equivalently use in Eqs. (25) and (27) the basis $\{s_{1l'}\}$, resulting in basis functions $\vartheta^{A,B,1}_{l'_1 l'_2 k'_1 k'_2}$ or the set $\{s_{0l}, w_{1l}\}$, leading to $\vartheta^{A,B}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}$ with $\boldsymbol{m}=(m,l)$. The corresponding equivalent expansions are

$$\begin{aligned} \gamma_2^s &= \sum_{\substack{l'_1, l'_2 \\ k'_1, k'_2}} \left[g^{A,1}_{l'_1 l'_2 k'_1 k'_2} \vartheta^{A,1}_{l'_1 l'_2 k'_1 k'_2} + g^{B,1}_{l'_1 l'_2 k'_1 k'_2} \vartheta^{B,1}_{l'_1 l'_2 k'_1 k'_2}\right] \\ &= \sum_{\substack{\boldsymbol{m}_1, \boldsymbol{m}_2 \\ \boldsymbol{n}_1, \boldsymbol{n}_2}} \left[g^{A}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2} \vartheta^{A}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2} + g^{B}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2} \vartheta^{B}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}\right]. \end{aligned} \tag{49}$$

Using transformation (47), the lengthy but simple algebra leads to

$$\begin{aligned} \vartheta^{A,1}_{l'_1 l'_2 k'_1 k'_2} = \sum_{\substack{\boldsymbol{m}_1, \boldsymbol{m}_2 \\ \boldsymbol{n}_1, \boldsymbol{n}_2}} &\left[X^{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}_{l'_1 l'_2 k'_1 k'_2} \vartheta^{A}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}\right. \\ &\left. + Y^{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}_{l'_1 l'_2 k'_1 k'_2} \vartheta^{B}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}\right], \end{aligned} \tag{50}$$

$$\begin{aligned} \vartheta^{B,1}_{l'_1 l'_2 k'_1 k'_2} = \sum_{\substack{\boldsymbol{m}_1, \boldsymbol{m}_2 \\ \boldsymbol{n}_1, \boldsymbol{n}_2}} &\left[X^{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}_{l'_1 l'_2 k'_1 k'_2} \vartheta^{B}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}\right. \\ &\left. - Y^{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}_{l'_1 l'_2 k'_1 k'_2} \vartheta^{A}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}\right], \end{aligned} \tag{51}$$

where the real transformation coefficients X and Y are defined by

$$U^{\boldsymbol{m}_1}_{l'_1} U^{\boldsymbol{m}_2}_{l'_2} (U^{\boldsymbol{n}_1}_{k'_1})^* (U^{\boldsymbol{n}_2}_{k'_2})^* = X^{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}_{l'_1 l'_2 k'_1 k'_2} + i\, Y^{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}_{l'_1 l'_2 k'_1 k'_2}. \tag{52}$$

The coarse-level expansion coefficients $g^{A,B}_{\boldsymbol{m}_1 \boldsymbol{m}_2 \boldsymbol{n}_1 \boldsymbol{n}_2}$ can be identified by substituting Eqs. (50) and (51) into Eq. (49)

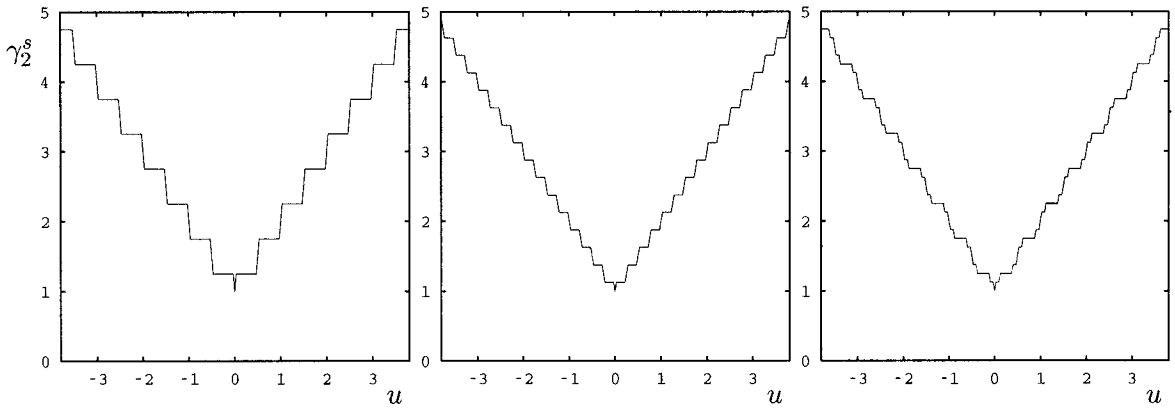

FIG. 1. Second-order density matrix at different placements of electron pairs. The center of pairs is characterized by the values (from left to right): $R=R'=0$ (both at grid points); $R=0$, $R'=0.5(2^{-m}b)$ (the second pair is halfway between two grid points); and $R=0$, $R'=1.3\,b$, which is never a grid point at any resolution level m. All calculations were carried out at level $m=2$. Atomic units were used.

$$g^{A}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}=\sum_{\substack{l_1',l_2'\\k_1',k_2'}}[X^{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}_{l_1'l_2'k_1'k_2'}g^{A,1}_{l_1'l_2'k_1'k_2'}-Y^{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}_{l_1'l_2'k_1'k_2'}g^{B,1}_{l_1'l_2'k_1'k_2'}] \tag{53}$$

$$g^{B}_{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}=\sum_{\substack{l_1',l_2'\\k_1',k_2'}}[X^{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}_{l_1'l_2'k_1'k_2'}g^{B,1}_{l_1'l_2'k_1'k_2'}+Y^{\boldsymbol{m}_1\boldsymbol{m}_2\boldsymbol{n}_1\boldsymbol{n}_2}_{l_1'l_2'k_1'k_2'}g^{A,1}_{l_1'l_2'k_1'k_2'}]. \tag{54}$$

In the case of the real-valued Haar basis functions all $Y=0$ and

$$g^{A,B}_{0l_1,0l_2;0k_1,0k_2}=\frac{1}{4}\sum_{\substack{\lambda_1=0,1\\\lambda_2=0,1}}\sum_{\substack{\kappa_1=0,1\\\kappa_2=0,1}}g^{A,B,1}_{2l_1+\lambda_1,2l_2+\lambda_2;2k_1+\kappa_1,2k_2+\kappa_2}. \tag{55}$$

Substituting the cusp conform expression (44) for $m=1$ into the right-hand side of Eq. (55) we arrive at terms containing $g^{A,B,1}_{L'L'K'K'}$, where L' is restricted to l_1+l_2 or l_1+l_2+1 and a similar constraint is true for K'. A smooth dependence on indices can be expressed as $g_{L'L'K'K'}\approx g_0+(L'-K')\Delta g+\cdots$, which results in the first order

$$g^{A,B,1}_{L'+1L'+1K'K'}\approx g^{A,B,1}_{L'L'K'K'}+\Delta g^{A,B},$$

$$g^{A,B,1}_{L'L'K'+1K'+1}\approx g^{A,B,1}_{L'L'K'K'}-\Delta g^{A,B},$$

$$g^{A,B,1}_{L'+1L'+1K'+1K'+1}\approx g^{A,B,1}_{L'L'K'K'}. \tag{56}$$

Using these approximations in Eq. (55) it can be realized that a relation similar to Eq. (44) holds for almost all index com-

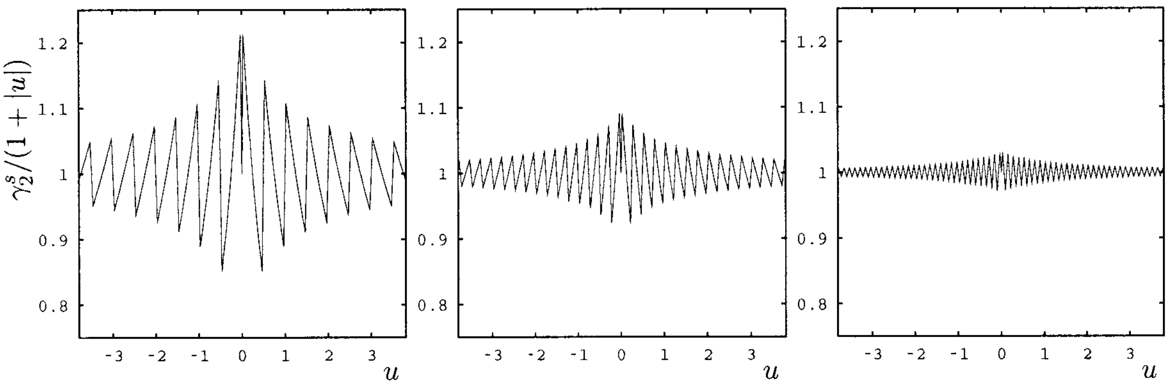

FIG. 2. Ratio of the calculated second-order density matrix to the theoretical expectation $1+|u|$ at resolutions $m=1$, $m=2$ and $m=3$, from left to right. The center of electron pairs was chosen as $R=0$, $R'=0.5(2^{-m}b)$. Distances are given in atomic units.

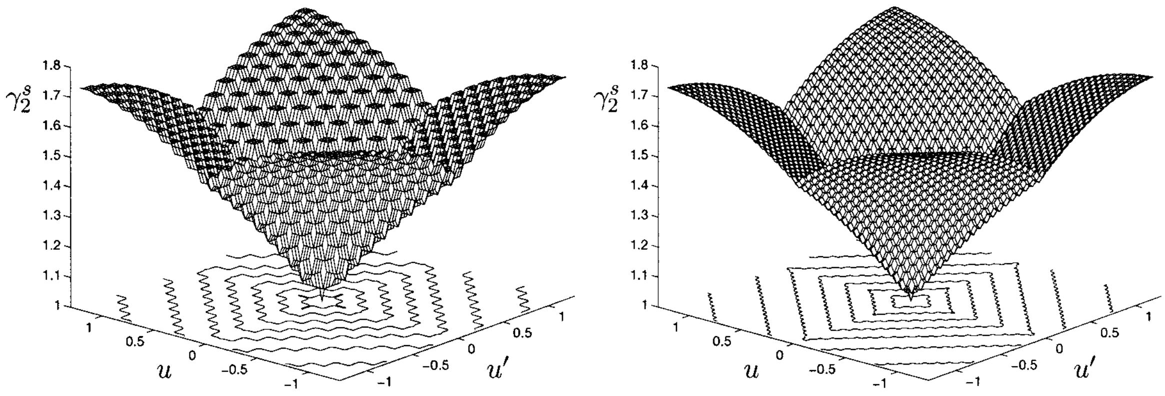

FIG. 3. Second-order density matrix as a function of electron separations u and u' at $R=R'=0$, in atomic units. The coarse resolution plot corresponds to $m=4$, whereas the fine resolution picture was calculated for $m=5$.

binations l_1,l_2,k_1,k_2 of the first-level expansion coefficients, supposing that the following scaling transformation equation is valid for the fine→coarse transition

$$g^{A,B,0}_{LLKK}=4\cdot g^{A,B,1}_{2L\,2L\,2K\,2K}. \tag{57}$$

This result indicates that the local form of the correlation cusp condition (19) is satisfied in a scale-invariant way by Eqs. (44) and (57).

The opposite, coarse→fine, transformation is trivial for even index values, however, continuity on indices implies a linear interpolation formula for odd labels

$$g^{A,B,1}_{2L+1\,2L+1\,2K\,2K}=\tfrac{1}{2}\left(g^{A,B,1}_{2L\,2L\,2K\,2K}+g^{A,B,1}_{2L+2\,2L+2\,2K\,2K}\right), \tag{58}$$

$$g^{A,B,1}_{2L\,2L\,2K+1\,2K+1}=\tfrac{1}{2}\left(g^{A,B,1}_{2L\,2L\,2K\,2K}+g^{A,B,1}_{2L\,2L\,2K+2\,2K+2}\right), \tag{59}$$

$$g^{A,B,1}_{2L+1\,2L+1\,2K+1\,2K+1}=\tfrac{1}{2}\left(g^{A,B,1}_{2L\,2L\,2K\,2K}+g^{A,B,1}_{2L+2\,2L+2\,2K+2\,2K+2}\right). \tag{60}$$

Knowing the scaling function expansion coefficients (57)–(60), the wavelet coefficients can be calculated using Eq. (44) and transformation rules (53) and (54).

The above considerations are also applicable to the transitions between arbitrary resolution levels m and $m+1$ leading to the scaling transformation (57) for $g^{A,B,m}_{LLKK}$ and $g^{A,B,m+1}_{2L\,2L\,2K\,2K}$.

Since in the previous considerations we made some simplification and assumptions, a numerical check of the results is appropriate. In order to illustrate the validity of predictions for a general coefficient $g^{A,B,m}_{l_1l_2k_1k_2}$ using expressions (44) and (57), we have calculated $\gamma_2^s(\mathbf{r}_1,\mathbf{r}_2|\mathbf{r}_1',\mathbf{r}_2')$ by Eq. (30) for several combinations of the one-dimensional variables r_1,r_2,r_1',r_2'.

As the first step we considered the spherically averaged cusp condition where we have used in expression (36) $R=R'$, $u=u'$ as the arguments of γ_2^s, moreover, we have chosen the grid point $R=0$. In Fig. 1 we have plotted $\gamma_2^s(R+u/2,R-u/2\,|R'+u/2,R'-u/2)$ for various separations of the electron pairs, in order to show that the above specific choice for R and R' does not affect the validity of our results. As we can realize, the most unfavorable case is, in fact, the choice $R=R'=0$ used in our theoretical considerations, all other possibilities, not taken explicitly into account, follow the behavior $(1+|u|)$ in a much smoother (although sometimes irregular) way.

We also wish to prove the reduction of errors by increasing the resolution level. In Fig. 2 the values of the calculated $\gamma_2^s(R+u/2,R-u/2\,|R'+u/2,R'-u/2)$ at different resolution levels are compared to the theoretical expectation $(1+|u|)$. Indeed, the ratio $\gamma_2^s/(1+|u|)$ tends to the constant $\gamma_2^s(R,R\,|R',R')$ that has been chosen as 1 for the present model calculation. The maximum errors appearing around $u\approx 0$ decrease exponentially with increasing m.

Finally, the fulfilment of the general (i.e., not spherically averaged) cusp condition (19) is illustrated by plots of $\gamma_2^s(u/2,-u/2\,|u'/2,-u'/2)$. In Fig. 3 we have applied an exponential long-range decay of the coefficients $g^{A,m}$, according to our previous note at the end of Sec. VII. The behavior $(1+|u|/2+|u'|/2)$ is clearly detectable.

IX. DISCUSSION

It has been shown in this work that the introduction of multiresolution analysis into the expansion of wave functions leads to a systematic refinement of representation of density matrices. This process, in a certain sense, is a backward renormalization transformation. The possibility of inverse transformation is permitted by the fact that the two-particle density operator must satisfy Bingel's cusp condition in the small electron-separation limit. We would like to emphasize, however, that even if the concepts used throughout this contribution are focused on density matrices and are also connected to renormalization, they differ completely from the technique of ‘‘density-matrix renormalization’’ developed by White [18,19] for other purposes.

Due to the well-localized behavior of scaling functions the algebraic form of the two-electron density-operator cusp condition is faithfully reflected by the index dependence of the expansion coefficients $g^{A,B,m}_{l_1 l_2 k_1 k_2}$. As we have also shown $g^{A,B,m}$ scales with increasing resolution level as $\sim 4^{-m}$. These behaviors allow a prediction of the $(m+1)$th level scaling function expansion coefficients from those of the mth level. The expected deviation of the predicted coefficients from the exact values becomes negligible if the resolution level tends to infinity.

Although, we have based the derivation and illustration of the results on Haar scaling functions, preliminary calculations using more sophisticated continuous scaling functions show an even better agreement with the general correlation cusp condition (19).

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