Evaluation of Gaussian integrals for the modeling of two-dimensional quantum systems

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We have developed a McMurchie-Davidson-like recursion formula for efficient evaluation of the Coulomb attraction and interaction matrix elements between two-dimensional primitive Cartesian Gaussian type orbitals. We also present recurrence schemes for combined position and differential operator integrals, and three-center Gaussian integrals. The Cartesian Gaussian orbitals are isotropic in the exponent, but with arbitrary centers and angular momentum.

I. INTRODUCTION

Two-dimensional many-particle quantum mechanical systems are routinely studied both theoretically and experimentally. Systems such as lateral quantum dots and rings,1 2 and electrons on liquid helium3 are confined to planar motion and can be treated as two-dimensional structures. They are seen as promising candidate models for qubits, and can be used to build quantum computers.

Solving these systems theoretically requires the use of approximations for all but the simplest problems. In particular for the case of multiparticle problems and arbitrary potential geometries. A large number of ab initio many-body methods are formulated in terms of a known single-particle basis. For some problems the eigenstates of the single-particle Hamiltonian can be used, but these states can make the solution of the interaction terms more involved. In three dimensions the usage of Cartesian Gaussian type orbitals have been the prevalent choice for ab initio methods applied to atomic and molecular systems3. The main reason for this is that there exists several efficient algorithms10 11 for the evaluation of matrix elements between Gaussian type functions and relevant one- and two-body operators. In particular, the computationally expensive, non-separable Coulomb attraction and interaction integrals can be expressed in terms of fast recursion relations for arbitrary angular momentum values.

However, due to the non-separability of the Coulomb integrals, the results for three dimensions do not generalize to lower dimensions. In particular, the computationally most intensive task of computing the two-body Coulomb interaction integrals are completely different in the two-dimensional case compared to the three-dimensional one. A common basis in two dimensions are the polar harmonic oscillator eigenstates, or the Fock-Darwin states, as the Coulomb interaction integrals can be solved analytically12 13. These are well suited for a single potential well located at the origin. In more complicated geometries with several well-centers, e.g., a double quantum dot, the quality of this basis rapidly deteriorates.

We propose the use of primitive Cartesian Gaussian type orbitals in two-dimensions as they are much more flexible in their shape and placement allowing for a wider range of potential geometries. In two dimensions the Coulomb integrals between s-Gaussians, i.e., spherical charge distributions, have a closed-form solution in terms of a modified Bessel function3. Previous work has often utilized several s-Gaussian orbitals to create a good enough single-particle basis3 8. Using p, d, and higher-order Gaussians gives a better single-particle basis and lowers the number of states needed to build a sufficiently good representation of the problem. In this article, we have extended these solutions by introducing McMurchie-Davidson-like recursion relations for the evaluation of Coulomb integrals between Gaussian type orbitals of arbitrary angular momentum. This creates a general framework for studying two-dimensional systems using ab initio methods.

In section II we define the primitive Cartesian Gaussian type orbitals and their properties. We also demonstrate how they can be expanded in Hermite-Gaussian functions and list the overlap integral between two Gaussian orbitals. We proceed to present recursion relations for the calculation of matrix elements between two Gaussian orbitals and the combined position and differential operator14, and a “Gaussian well”-operator15. Next, we derive the recursion relations for the integral over two Gaussian orbitals and the Coulomb attraction operator, before we demonstrate how these relations can be reused for the integral over four Gaussian orbitals and the Coulomb interaction operator. Finally, we conclude with some remarks on the application of these basis functions and future prospects.

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A two-dimensional primitive unnormalized Cartesian Gaussian orbital may be written \[ g_{a}(r; a, A) = x_{a}^{i}y_{A}^{j} \exp(-ar^{2}), \] where \( \alpha = (i, k) \in \mathbb{N}_{0}^{2} \) are the Cartesian quantum numbers with the total orbital angular moment given by \( l = i + k, r_{A} \equiv r - A, r_{A} = ||r_{A}|| \) with \( A = (A_{x}, A_{y}) \in \mathbb{R}^{2} \) the center of the Gaussian, and \( a \in \mathbb{R}^{2} \) as the spread of the Gaussian. We will restrict \( a \) to be a scalar making the Gaussians separable in each Cartesian direction, viz., \( g_{a}(r; a, A) = g_{x}(x; a, A_{x})g_{y}(y; a, A_{y}) \). This lets us calculate many of the integrals as products of one-dimensional Gaussians. Multiplying two one-dimensional Gaussians results in a new Gaussian called the overlap distribution, \( g_{i}(x; a, A)g_{j}(x; b, B) = Kx^{i}y^{j} \exp(-px^{2}) \equiv \Omega_{ij}(x; p, \mu, P, X_{AB}), \) where the parameters are the total exponent \( p = a + b \), the center of mass \( P = (aA + bB)/p, K \equiv \exp(-\mu X^{2}_{AB}) \) with the reduced exponent \( \mu = ab/(a + b) \), and the center difference \( X_{AB} = A - B \). For notational brevity, we will not include the parameters, i.e., \( p, \mu, P, X_{AB} \), when writing the overlap distribution. It should be understood that the parameters for \( \Omega_{ij}(x) \) are built from \( g_{i}(x) \) and \( g_{j}(x) \). In two dimensions the overlap distribution is given by a product of the overlap distribution between the \( x \)- and \( y \)-components of the Gaussian functions, viz., \( g_{a}(r)g_{\beta}(r) = \Omega_{a\beta}(r) = \Omega_{ij}(x)\Omega_{kl}(y) \), where \( \alpha = (i, k) \) and \( \beta = (j, l) \), and the \( x \)- and \( y \)-components of the center of mass \( P = (P_{x}, P_{y}) \) and center difference \( R_{AB} = A - B = (X_{AB}, Y_{AB}) \) are parameters to \( \Omega_{ij}(x) \) and \( \Omega_{kl}(y) \) separately.

Now, as in the McMurchie-Davidson scheme \[ \Omega_{ij}(x) = \int \sum_{t=0}^{i+j} \sum_{u=0}^{i+j} E^{ij}_{tu} \Lambda_{t}(x), \] where \( E^{ij}_{tu} \) are the expansion coefficients and \( \Lambda_{t}(x; p, P) \) the Hermite-Gaussian functions given by \[ \Lambda_{t}(x; p, P) = \frac{\partial^{t}}{\partial P^{t}} \exp(-px^{2}), \] where the parameters \( p, \mu, P \) and \( X_{AB} \) in the expansion coefficients and the Hermite-Gaussian functions are the parameters from the overlap distribution. This expansion is exact as the overlap distribution \( \Omega_{ij}(x) \) is a polynomial of degree \( i + j \) in \( x_{P} \) and we expand it in a linear combination of Hermite-Gaussian functions of degree \( t \leq i + j \) in \( x_{P} \). The two-dimensional expansion is then a product of the two one-dimensional expansions \[ \Omega_{\alpha\beta}(r) = \sum_{t=0}^{i+j} \sum_{u=0}^{i+j} E^{\alpha\beta}_{tu} \Lambda_{tu}(r), \] with \( E^{\alpha\beta}_{tu} = E^{ij}_{tu} E^{kl}_{tu} \) and \( \Lambda_{tu}(r) = \Lambda_{t}(x)\Lambda_{u}(y) \). The recurrence scheme for the expansion coefficients are given by \[ E_{i}^{i+1,j} = \frac{1}{2p} E_{i-1}^{ij} + X_{PA} E_{i}^{ij} + (t + 1)E_{i+1}^{ij}, \] \[ E_{i}^{i,j+1} = \frac{1}{2p} E_{i-1}^{ij} + X_{PB} E_{i}^{ij} + (t + 1)E_{i+1}^{ij}, \] where \( E_{00}^{00} = K = \exp(-\mu X^{2}_{AB}) \), and \( E_{t}^{ij} = 0 \) for \( t < 0 \) and \( t > i + j \), with the derivation being listed in appendix A. The overlap integral between two one-dimensional Gaussian primitives can now be found by using the expansion of the overlap distribution as Hermite-Gaussian functions, \[ s_{ij} = \langle g_{i}(a, A)|g_{j}(b, B) \rangle = E_{0}^{ij} \sqrt{\frac{\pi}{p}}, \] where \( p = a + b \) is the total exponent. The derivation of this expression can be found in appendix B. The normalization factor of a one-dimensional primitive Gaussian orbital \( g_{i}(x; a, A) \) is found by \( N = \sqrt{s_{ii}} \). Extending to two dimensions is done by evaluating a product of two overlap integrals; one for each Cartesian direction, viz., \[ s_{\alpha\beta} = \langle g_{\alpha}(a)|g_{\beta}(b) \rangle = s_{ij}s_{kl}. \]

A. Combined position and differential operator integral

The second integral we wish to solve using arbitrary primitive Gaussian functions is the combined position and differential operator. We derive this expression for the differential operator instead of the momentum operator to ensure that all integrals are real. To ensure full generality we allow for arbitrary centers of the position operator, and arbitrary exponents for both the position and differential operator. The integral we wish to solve is thus \[ L^{ef}_{ij} = \langle g_{i}(a, A)|\tilde{z}^{f}_{\epsilon} \frac{df}{dx_{\epsilon}} |g_{j}(b, B) \rangle, \] with extensions to two dimensions by taking the product of a similar integral with \( x \to y \). There are two downward recursion formulas – one for the position term and another for the differential operator – for this integral. They are given by \[ L_{ij}^{e+1,f} = L_{i+1,j}^{ef} + X_{AC} L_{ij}^{ef}, \] \[ L_{ij}^{c,f+1} = jL_{i+1,j-1}^{ef} - 2bL_{i,j+1}^{ef}, \] where \( X_{AC} = A - C \). The base case of the recursion is the overlap integral, i.e., \( L_{00}^{00} = s_{ij} \) from Equation 8. See appendix C for the derivation – which is based on the work of Tellgren et al. [10] – of these matrix elements. As
an example, from Equation 10 we can build the kinetic operator matrix elements in two dimensions by

\[ T_{\alpha\beta} = \langle g_\alpha (a, A) \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \rangle | g_\beta \rangle = L_{ij}^{02} s_{kl} + s_{kj} L_{ki}^{02}, \quad (13) \]

where \( \alpha = (i, k) \), \( \beta = (j, l) \), and we have inserted the one-dimensional overlap integral between the Cartesian components that are not dependent on the derivative operator.

### B. Gaussian well integral

When modeling \( n \)-well systems it is useful to tune the placement and shape of the various wells. One of the easiest potential wells that allow for this flexibility are Gaussian potential wells. This results in three-center integrals \[11\]. For the sake of full generality we therefore define the one-dimensional Gaussian potential well to be on the form

\[ \hat{v}_k(x; c, C, w) = -wx^k_c \exp(-cx^2_c), \quad (14) \]

that is, the Gaussian well is itself a primitive Gaussian with an exponent \( c > 0 \) and \( w > 0 \) giving a negative weight. Extending to higher dimensions results in a product between two one-dimensional wells. The matrix elements for this operator is given by

\[ G^{k}_{ij} = \langle g_i (a, A) | \hat{v}_k | g_j (b, B) \rangle = -w \sum_{t=0}^{i+j} E^{ij}_t P^k_{t0}, \quad (15) \]

where \( E^{ij}_t \) are the Hermite-Gaussian expansion coefficients between \( g_i(x; a, A) \) and \( g_j(x; b, B) \), and \( P^k_{t0} \) can be found from the recurrence scheme

\[ P^k_{t0} = \langle g_k (c, C) | \frac{\partial^t}{\partial P^t} | g_0 (p, P) \rangle = 2p P^{t-1}_{k,t+1} - l P^{t-1}_{k,t-1}, \quad (16) \]

with the base case \( P^0_{k0} = \langle g_k (c, C) | g_0 (p, P) \rangle = s_{kt} \), i.e., the overlap integral. It is worth noting which Gaussians are included in the recurrence scheme for \( P^t_{k0} \) as this is a two-center integral. The \( g_i(x; p, P) \) Gaussian is the result from the product of \( g_i(x; a, A) \) and \( g_j(x; b, B) \) with \( p \) as the total exponent and \( P \) the center of mass from the product. The Gaussian \( g_k(x; c, C) \) is the potential well. The derivation of this expression can be found in appendix \[D\].

### C. Coulomb attraction integral

Next, we look at the Coulomb attraction integral in two dimensions. This integral is not separable in the two Cartesian directions and we have to treat the integral in full. We wish to compute

\[ v_{\alpha\beta} = \langle g_\alpha (a, A) | \frac{1}{r_C} | g_\beta (b, B) \rangle = \int d^2r \frac{\Omega_{\alpha\beta}(r)}{r_C}, \quad (17) \]

where \( r_C \equiv \|r - C\| \), and \( \alpha = (i, k) \) and \( \beta = (j, l) \) are the compound indices of the two-dimensional Gaussians. We have set \( s = (4\pi \alpha_0)^{-1} = 1 \) to keep the expressions as clean as possible. We expand the Gaussian overlap distribution in Hermite-Gaussian functions and move the derivative operators in front of the integral. This gives

\[ v_{\alpha\beta} = \sum_{tu} E^{\alpha\beta}_{tu} \frac{\partial^t}{\partial P^t_x} \frac{\partial^u}{\partial P^u_y} \int d^2r \frac{\exp(-pt^2)}{r_C}, \quad (18) \]

where \( \sum_{tu} = \sum_{t=0}^{i+j} \sum_{n=0}^{k+l} \). The closed form expression for the two-dimensional integral over the spherical Gaussian is derived in appendix \[E\] and is given by

\[ \int d^2r \frac{\exp(-pt^2)}{r_C} = \pi \frac{\gamma}{p} \exp(-p\Sigma^2/2), \quad (19) \]

where \( \gamma_0 (z) \equiv \exp(z) I_0(z) \) is the exponentially scaled modified Bessel function of the first kind, and \( \Sigma = P - C \). Inserted back into Equation 18 we have

\[ v_{\alpha\beta} = \pi \frac{\gamma}{p} \sum_{tu} E^{\alpha\beta}_{tu} \tilde{I}_{tu}(-p\Sigma^2/2), \quad (20) \]

where we have defined

\[ \tilde{I}_{tu}(-p\Sigma^2/2) = \frac{\partial^t}{\partial P^t_x} \frac{\partial^u}{\partial P^u_y} \frac{\gamma}{p} \tilde{I}_0(-p\Sigma^2/2), \quad (21) \]

Now the task is to find a recurrence formula for the evaluation of Equation 21. We therefore introduce the intermediate integrals

\[ \tilde{I}^n_{tu}(-p\Sigma^2/2) = \frac{\partial^t}{\partial P^t_x} \frac{\partial^u}{\partial P^u_y} \tilde{I}_0(-p\Sigma^2/2), \quad (22) \]

where \( \tilde{I}_{tu}(z) \equiv \tilde{I}^0_{tu}(z) \) and \( \tilde{I}^n_{tu}(z) \equiv \tilde{I}_n(z) \). Incrementing \( t \) and \( u \) (separately) by one in Equation 22 and collecting terms we find the relations

\[ \tilde{I}^n_{t+1,u} - \frac{p}{2} \left[ t \left( \tilde{I}^{n-1}_{t-1,u} + 2\tilde{I}^{n-1}_{t-1,u} + \tilde{I}^{n+1}_{t-1,u} \right) + \Sigma_z \left( \tilde{I}^{n-1}_{tu} + 2\tilde{I}^{n}_{tu} + \tilde{I}^{n+1}_{tu} \right) \right], \quad (23) \]

\[ \tilde{I}^n_{t,u+1} = \frac{p}{2} \left[ u \left( \tilde{I}^{n-1}_{t,u-1} + 2\tilde{I}^{n}_{t,u-1} + \tilde{I}^{n+1}_{t,u-1} \right) + \Sigma_u \left( \tilde{I}^{n-1}_{tu} + 2\tilde{I}^{n}_{tu} + \tilde{I}^{n+1}_{tu} \right) \right], \quad (24) \]
where we have removed the function arguments (they are the same as in Equation 20) for brevity. These expressions apply for $n \geq 0$ and with $i_{-1}(z) = i_1(z)$. The recursion schemas are derived in [17].

The Coulomb attraction potential is seldom used in two-dimensional systems. They are similar to the Gaussian wells in that they are flexible in their placement and strength. We have included it in this article as recursion relations can be completely reused in the two-body interaction integral.

D. Coulomb interaction integral

We are now in a position to set up the Coulomb interaction matrix elements between two-dimensional primitive Cartesian Gaussian functions. The Coulomb interaction operator is given by

$$\hat{u}(r_1, r_2) = \frac{1}{\|r_1 - r_2\|},$$

(25)

where we again have set all physical constants to unity, and we define $r_{12} \equiv \|r_1 - r_2\|$. The matrix elements are then given by

$$u_{\alpha \beta}^{\gamma \delta} = \langle g_\alpha(a, A)g_\beta(b, B)|u|g_\gamma(c, C)g_\delta(d, D) \rangle = \int d^2r_1 d^2r_2 \frac{\Omega_{\alpha \gamma}(r_1)\Omega_{\beta \delta}(r_2)}{r_{12}}$$

$$= \sum_{\nu \tau} E_{t \nu}^{\alpha \gamma} E_{t \nu}^{\beta \delta} \frac{\Lambda_{t \nu}(r_1)\Lambda_{t \nu}(r_2)}{r_{12}}.$$ (26)

For the integrals over the Hermite-Gaussians (sans the expansion coefficients) we have

$$V_{t_\nu; t_\nu} = \frac{\partial^\tau \partial^\nu}{\partial P_2^\nu \partial P_2^\nu} \frac{\partial^\alpha \partial^\beta}{\partial Q_2^\nu \partial Q_2^\nu} \frac{\partial^\gamma \partial^\delta}{\partial Q_2^\nu \partial Q_2^\nu} V_0,$$

(27)

where we have denoted the integral over the two spherical Gaussian charge distributions by $V_0$. This integral has a closed form solution given by

$$V_0 = \int d^2r_1 d^2r_2 \frac{\exp(-pr_1^2/4\sigma)\exp(-pr_2^2/4\sigma)}{r_{12}}$$

$$= \frac{\pi^2}{pq} \sqrt{\frac{\pi}{4\sigma}} \tilde{\Omega}_{t_\nu}(\Delta^2/(8\sigma)),$$

(28)

with $r_{1\nu} \equiv r_1 - P$ and similarly for $r_{2\nu}, \sigma = (p+q)/(4pq)$, and $\Delta = Q - P$. This derivation is demonstrated in appendix [26]. Except for the function argument and the prefactor, the Coulomb interaction integral between two spherical Gaussian orbitals is identical to the Coulomb attraction integral. To be able to use the recursion formula in Equation 23 we look at the function argument to the exponentially scaled Bessel function. Defining $f = -\Delta^2/(8\sigma)$ for the function argument, we have that

$$\frac{\partial f}{\partial Q_2^\nu} = -\frac{\Delta_i}{4\sigma} = -\frac{\partial f}{\partial P_2^\nu}, \quad \frac{\partial^2 f}{\partial Q_2^\nu \partial Q_2^\nu} = \frac{\partial^2 f}{\partial P_2^\nu \partial P_2^\nu} = -\frac{1}{4\sigma}.$$ (29)

This lets us write the integral over the two Hermite-Gaussian functions as

$$V_{t_\nu; t_\nu} = (-1)^{t+u} \frac{\pi^2}{pq} \sqrt{\frac{\pi}{4\sigma}} \tilde{I}_{t + \tau, u + \nu}(-\Delta^2/(8\sigma)),$$

(30)

where $\tilde{I}_{t + \tau, u + \nu}$ is the same as in Equation 21 with $P_i \rightarrow Q_i$ in the center derivatives shown in Equation 22 [17]. We therefore find the same recursion formula as in Equation 23 but with $p \rightarrow (4\sigma)^{-1}$ and $\Sigma_i \rightarrow \Delta_i$. This leaves us with the full expression for the Coulomb interaction matrix elements

$$u_{\alpha \beta}^{\gamma \delta} = \frac{\pi^2}{pq} \sqrt{\frac{\pi}{4\sigma}} \sum_{\nu \tau} (-1)^{t+u} E_{t \nu}^{\alpha \gamma}$$

$$\times \sum_{\tau \nu} E_{t \nu}^{\beta \delta} \tilde{I}_{t + \tau, u + \nu}(-\Delta^2/(8\sigma)).$$ (31)

III. SUMMARY REMARKS

In this report, we have derived recursion formulas for the evaluation of atomic integrals using two-dimensional primitive Gaussian orbitals. The formulas are an extension of the three-dimensional McMurchie-Davidson method to two-dimensional systems. These integrals allow for more flexibility in the construction of basis sets for $ab$ initio methods applied to two-dimensional systems, e.g., lateral quantum dots.

In this paper, we have only been concerned with the technicalities of computing matrix elements, but studies as to how the Gaussian basis elements should be chosen, e.g., placement of the centers, width of the peak, is of interest. Much can be learned from previous work on contracted primitive Gaussian orbitals in the three-dimensional case.

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Appendix A: Hermite-Gaussian expansion coefficients

The expansion coefficients $E_{t \nu}^{\alpha \beta}$ in Equation 5 are separable in each Cartesian direction. That is,

$$E_{t \nu}^{\alpha \beta} = E_{t \nu}^{ij} E_{t \nu}^{kl},$$ (A1)

with $\alpha = (i, k)$ and $\beta = (j, l)$. Restricting our attention to the one-dimensional case we expand the overlap distribution in Hermite-Gaussians

$$\Omega_{ij}(x) = g_i(x; a, A)g_j(x; b, B) = \sum_{i=0}^{\infty} E_{t \nu}^{ij} \Lambda_t(x),$$ (A2)
where this will be an exact expansion as $\Omega_{ij}(x)$ is a polynomial of degree $i + j$ in $x_p$. To build a recurrence scheme we increment one of the Gaussians in the overlap distribution and write

$$\Omega_{i+1,j}(x) = g_{i+1}(x)g_j(x) = x_A g_i(x)g_j(x) = x_p \Omega_{ij}(x) + X_{PA} \Omega_{ij}(x).$$  \hspace{1cm} (A3)

Looking at the first term, we expand the overlap distribution in Hermite-Gaussian functions. We can then find a relation for the Hermite-Gaussian multiplied by $x_p$, viz.,

$$x_p \Lambda_t(x) = x_p \frac{\partial^t}{\partial x_p^t} \exp(-px_p^2) = \frac{1}{2p} \Lambda_{t+1}(x) + t \Lambda_{t-1}(x),$$  \hspace{1cm} (A4)

where we have used the commutation relation

$$\left[x_p, \frac{\partial^t}{\partial x_p^t}\right] = t \frac{\partial^t}{\partial x_p^{t-1}}.$$  \hspace{1cm} (A5)

Inserted back into Equation A3 (and removing function arguments) we get

$$\Omega_{i+1,j} = \sum_{t=0}^{i+j+1} E_t^{i+1,j} \Lambda_t = [x_p + X_{PA}] \Omega_{ij}$$

$$= \sum_{t=0}^{i+j} E_t^{ij} \left[ \frac{1}{2p} \Lambda_{t+1} + X_{PA} \Lambda_t + t \Lambda_{t-1} \right]$$

$$= \sum_{t=1}^{i+j+1} \frac{1}{2p} E_t^{ij} \Lambda_t + \sum_{t=-1}^{i+j-1} (t+1) E_t^{ij} \Lambda_t$$

$$+ \sum_{t=0}^{i+j} X_{PA} E_t^{ij} \Lambda_t,$$  \hspace{1cm} (A6)

where we have changed the limits on the two first sums. Now we wish to collect all terms under a common sum and factor out the Hermite-Gaussian functions on both sides of the equation. First, we require that $E_t^{ij} = 0$ for $t < 0$ allowing the starting limit on the first sum to be set to 0. We can also set $t = 0$ to the starting limit for the second sum as the $(t + 1)$-term removes the $t = -1$ limit. Second, as we restrict our attention to polynomials of at most degree $i + j$ we require that $E_t^{ij} = 0$ for $t > i + j$. This lets us shift the limits on the second and third sums to $i + j + 1$. Collecting we then get

$$\Omega_{i+1,j} = \sum_{t=0}^{i+j+1} E_t^{i+1,j} \Lambda_t$$

$$= \sum_{t=0}^{i+j+1} \left[ \frac{1}{2p} E_t^{ij} \Lambda_t + (t+1) E_{t+1}^{ij} + X_{PA} E_t^{ij} \right] \Lambda_t.$$  \hspace{1cm} (A7)

Equating terms at each order of Hermite-Gaussian then gives the recurrence scheme for the expansion coefficients at every step in the sum. We are then left with

$$E_t^{i+1,j+1} = \frac{1}{2p} E_t^{ij} + X_{PA} E_t^{ij} + (t+1) E_{t+1}^{ij},$$  \hspace{1cm} (A8)

$$E_t^{i+1,j+1} = \frac{1}{2p} E_t^{ij} + X_{PB} E_t^{ij} + (t+1) E_{t+1}^{ij},$$  \hspace{1cm} (A9)

where $E_0^{00} = \exp(-\mu X_{AB}^2)$, and $E_t^{ij} = 0$ for $t < 0$ and $t > i + j$.

Appendix B: Overlap integral between two Gaussian orbitals

Here we derive the overlap integral – as seen in Equation 8 – between two one-dimensional primitive Gaussians.

$$s_{ij} = \langle g_i(a, A) | g_j(b, B) \rangle$$

$$= \int dx g_i(x)g_j(x)$$

$$= \int dx \Omega_{ij}(x) = \sum_{t=0}^{i+j} E_t^{ij} \int dx \Lambda_t(x)$$

$$= \sum_{t=0}^{i+j} E_t^{ij} \frac{\partial^t}{\partial x_p^t} \int dx \exp(-px_p^2)$$

$$= \sum_{t=0}^{i+j} E_t^{ij} \frac{\partial^t}{\partial x_p^t} \sqrt{\frac{\pi}{p}} = \sum_{t=0}^{i+j} E_t^{ij} \delta_{t0} \sqrt{\frac{\pi}{p}}$$

$$= E_0^{ij} \sqrt{\frac{\pi}{p}},$$  \hspace{1cm} (B1)

where $p$ is the total exponent from the product of the two Gaussian functions. The overlap between two two-dimensional primitive Gaussians will be a product of the overlap between each Cartesian direction. That is,

$$s_{\alpha\beta} = \langle g_{\alpha}(a) | g_{\beta}(b) \rangle = \langle g_{ak} | g_{bj} \rangle = s_{ij} \delta_{kl}.$$  \hspace{1cm} (B2)

Appendix C: Combined position and differential operator integrals

We derive the recursion formulas for the combined position and differential operator integrals shown in Equation 11 and Equation 12. We start by finding a relation for the position term on a single Gaussian orbital. That is,

$$g_i(x; a, A)x_C = x_i' \exp(-ax_i'^2) (x - C - A + A)$$

$$= x_i' \exp(-ax_i'^2) (x_A + X_{AC})$$

$$= g_{i+1}(x) + X_{AC} g_i(x).$$  \hspace{1cm} (C1)

The downward recurrence scheme for the position is then

$$L_{ij}^x = \langle g_i | x_C \frac{df}{dx} \rho | g_j \rangle$$

$$= \left[ \langle g_{i+1} | + X_{AC} \langle g_i | \right] x_{C}^{i-1} \frac{df}{dx} \rho | g_j \rangle$$

$$= L_{i+1,j}^x + X_{AC} L_{ij}^{x-1},$$  \hspace{1cm} (C2)
We thus find Equation 12 from the action of a derivative operator on a primitive one-dimensional Gaussian orbital is

\[
\frac{d}{dx} g_j(x; b, B) = \frac{d}{dx_B} \left[ x^i_B \exp(-bx^2_B) \right] = \delta g_{j-1}(x) - 2b g_{j+1}(x). \tag{C3}
\]

We thus find Equation 12 from

\[
L_{ij}^f = (g_i | \hat{x}^f \frac{d}{dx_j} | g_j) = (g_i | x_j \frac{d^{f-1}}{dx} | j | g_{j-1}) - 2b | g_{j+1} \rangle = jL_{i,j-1}^f - 2bL_{i,j+1}^f. \tag{C4}
\]

**Appendix D: Gaussian well integral**

The matrix element between two primitive Gaussians over a Gaussian well is given by

\[
G_{ij}^f = \langle g_i(a, A)| \hat{v}_c(c, w)| g_j(b, B) \rangle = -w \int dx x_k^c \exp(-cx^2_C) \Omega_{ij}(x)
\]

\[
= -w \sum_{i+j} E_t^{ij} \int dx g_k(x) \Lambda_{i}(x)
\]

\[
= -w \sum_{i+j} E_t^{ij} \langle g_k(c, C) | \frac{\partial^x}{\partial t^x} | g_0(p, P) \rangle
\]

\[
= -w \sum_{i+j} E_t^{ij} P_{k}^{i}. \tag{D1}
\]

Here it is worth noting that the choice of creating an overlap distribution between \(g_k(x; aA)\) and \(g_j(x; bB)\) is somewhat arbitrary. This can very well be done between the well and any of the two. However, this requires some care when manipulating the indices for the expansion coefficients and the matrix elements \(P_{k}^{i}\). We observe that \(P_{k}^{0} = \langle g_k|g_0 \rangle = s_{kl}\). For \(t > 0\) we get

\[
P_{k}^{t} = \langle g_k(c, C) | \frac{\partial^x}{\partial t^x} | g_0(p, P) \rangle
\]

\[
= \langle g_k | \frac{\partial^{t-1}}{\partial P_{t-1}} \left[ 2p | g_{i+1} \rangle - l | g_{i-1} \rangle \right]
\]

\[
= 2pP_{k,l+1}^{t-1} - lP_{k,l-1}^{t-1}. \tag{D2}
\]

**Appendix E: Coulomb attraction integral over a spherical Gaussian charge distribution**

We derive a closed-form expression for the Coulomb attraction integral over a two-dimensional spherical Gaussian charge distribution. We have

\[
v_0(C) = \int d^2r \frac{\exp(-pr^2)}{r_C}
\]

\[
= \int d^2r \exp(-pr^2) \int dt \frac{1}{\sqrt{\pi}} \exp(-r_C^2t^2), \tag{E1}
\]

where we in the last equation have used that

\[
\int dt \exp(-r_C^2t^2) = \frac{\sqrt{\pi}}{r_C}, \tag{E2}
\]

for \(\text{Re}(r_C) > 0\), in reverse. We now use the Gaussian product rule

\[
\exp(-pr^2) \exp(-r_C^2t^2) = \exp(-\nu \Sigma^2), \tag{E3}
\]

where we have introduced \(s = p + t^2, \nu = pt^2/s, \Sigma = (pP + t^2C)/(p + t^2)\), and \(\Sigma = P - C\). Inserting this back into the integral we can now solve the integral over \(d^2r\) for the last term in the Gaussian product. This leaves us with

\[
v_0(C) = \frac{1}{\sqrt{\pi}} \int dt \exp\left(-\frac{pt^2}{p + t^2} \Sigma\right) \frac{\pi}{p + t^2}. \tag{E4}
\]

This integral is symmetric for \(t \in (-\infty, \infty)\). We therefore shift the limits of integration to \(t \in (0, \infty)\) and multiply with a factor of 2. Then

\[
v_0(C) = \frac{2}{\sqrt{\pi}} \int_0^\infty dt \frac{p}{p + t^2} \exp\left(-\frac{pt^2}{p + t^2} \Sigma\right), \tag{E5}
\]

where we have moved a factor of \(p\) into the numerator of the integral. Next, we perform a variable substitution introducing

\[
u^2 = \frac{t^2}{p + t^2} \quad \Rightarrow \quad u = \frac{t}{\sqrt{p + t^2}}, \tag{E6}
\]

where \(t \geq 0\) from the integration limits and \(\text{Re}(p) > 0\). The new integration limits are \(\lim_{t \to \infty} u(t) = 1\) and \(u(0) = 0\). We also have

\[
\frac{du}{dt} = \frac{(1 - u^2)^{3/2}}{\sqrt{p}} \quad \Rightarrow \quad dt = \frac{\sqrt{p}}{(1 - u^2)^{3/2}} \, du, \tag{E7}
\]

for the variable substitution. Inserted back into \(v_0(C)\) we are then left with

\[
v_0(C) = \frac{2}{\sqrt{\pi}} \int_0^1 du \frac{\sqrt{p}}{(1 - u^2)^{3/2}} \exp(-pu^2\Sigma^2) \]

\[
= \sqrt{\frac{\pi}{p}} \exp\left(-\frac{pu^2\Sigma^2}{2}\right) \, I_0\left(\frac{pu^2\Sigma^2}{2}\right), \tag{E8}
\]

where \(I_0(z)\) is the modified Bessel function of the first kind. Note that \(I_0(z) = I_0(-z)\) making the choice of sign in the argument arbitrary. However, when we create a recursion formula for higher-order Hermite Gaussians we benefit from using the same sign in the exponential function and the Bessel function.
Appendix F: Recursion formula for the Coulomb attraction integral

Defining \( z = -p\Sigma^2 / 2 \) as the argument of the exponential function and the Bessel function in Equation 19 we find the derivative of this quantity with respect to \( P_z \) and \( P_y \) to be

\[
\frac{\partial z}{\partial P_i} = -p\Sigma_i, \quad \frac{\partial^2 z}{\partial P_i^2} = -p. \tag{F1}
\]

Furthermore, we define the exponentially scaled modified Bessel function of the first kind to be

\[
\tilde{i}_n(z) = \exp(z) I_n(z), \tag{F2}
\]

where \( n \) denotes the order of the modified Bessel function. The derivative of this function is

\[
\frac{d\tilde{i}_n}{dz} = \tilde{i}_n(z) + \frac{1}{2} [\tilde{i}_{n-1}(z) + \tilde{i}_{n+1}(z)], \tag{F3}
\]

where \( n \geq 0 \) and \( \tilde{i}_{-1}(z) = \tilde{i}_1(z) \). Using the intermediate integrals defined in Equation 22 and in the text below, we compute

\[
\tilde{I}_{t+1,u}(p, \Sigma) = \frac{\partial^t}{\partial P_z^t} \frac{\partial^u}{\partial P_y^u} \left. \frac{\partial \tilde{i}_n}{\partial z} \right|_{z = -p\Sigma^2 / 2} = \frac{\partial^t}{\partial P_z^t} \frac{\partial^u}{\partial P_y^u} \left. \tilde{i}_n \right|_{z = -p\Sigma^2 / 2} = \frac{\partial^t}{\partial P_z^t} \frac{\partial^u}{\partial P_y^u} (-p\Sigma_x) \left. \frac{1}{2} [\tilde{i}_{n-1} + \tilde{i}_{n+1}] \right. \\
+ 2\tilde{i}_n + \tilde{i}_{n+1}, \tag{F4}
\]

where we for brevity have removed the function arguments to the exponentially scaled modified Bessel functions. To proceed from here we note that the derivative with respect to \( P_y \) “passes through” the term in front of the Bessel functions. For the derivatives with respect to \( P_x \) we have that

\[
\left[ \frac{\partial^t}{\partial P_x^t}, \Sigma_x \right] = t \frac{\partial^{t-1}}{\partial P_x^{t-1}}. \tag{F5}
\]

We then have

\[
\tilde{I}_{t+1,u}(p, \Sigma) = \frac{p}{2} \left[ \tilde{i}_n^{t-1} + \Sigma_x \frac{\partial}{\partial P_x} \right] \\
\times \left[ \tilde{i}_0^{t-1} + 2\tilde{i}_0 + \tilde{i}_{0+1} \right] \\
= \frac{p}{2} \left[ \left( \tilde{i}_0^{t-1} + 2\tilde{i}_0 + \tilde{i}_{0+1} \right) \\
+ \Sigma_x \left( \tilde{i}_0^{t-1} + 2\tilde{i}_0 + \tilde{i}_{0+1} \right) \right]. \tag{F6}
\]

For the \( y \)-direction we have a similar relation given by

\[
\tilde{I}_{t,u+1}(p, \Sigma) = \frac{-p}{2} \left[ u \left( \tilde{i}_0^{t-1} + 2\tilde{i}_0 + \tilde{i}_{0+1} \right) \\
+ \Sigma_y \left( \tilde{i}_0^{t-1} + 2\tilde{i}_0 + \tilde{i}_{0+1} \right) \right]. \tag{F7}
\]

Appendix G: Coulomb interaction between two spherical Gaussian charge distributions

We derive the closed-form expression for the Coulomb interaction integral between two two-dimensional spherical Gaussian charge distributions. This is based on the work done by Nielsen et al. [3]. The integral we wish to solve is on the form

\[
V_0 = \int d^2r_1 d^2r_2 \frac{\exp(-pr_1^2) \exp(-qr_2^2)}{r_{12}}, \tag{G1}
\]

where we have defined \( r_{1P} \equiv r_1 - \mathbf{P} \) and similarly \( r_{2Q} \). We also have \( r_{12} \equiv \| r_1 - r_2 \| \). To proceed we use the momentum representation of the exponential functions and the Coulomb operator. They are given by

\[
\exp(-pr_{1P}^2) = \frac{\pi}{p} \int \frac{d^2k}{(2\pi)^2} \exp\left(-\frac{k^2}{4p} \right) \exp(-ik \cdot r_{1P}), \\
\frac{1}{r_{12}} = \int \frac{d^2k}{(2\pi)^2} \frac{k}{k} \exp(-ik \cdot r_{12}). \tag{G2}
\]

Inserted into the full integral we have

\[
V_0 = \frac{\pi^2}{pq} \int d^2r_1 d^2r_2 d^2k_1 d^2k_2 d^2k_3 \left( \frac{2\pi}{(2\pi)^2} \right)^2 \left( \frac{2\pi}{(2\pi)^2} \right)^2 \left( \frac{2\pi}{(2\pi)^2} \right)^2 \\
\exp(-\sigma k^2) \exp(ik \cdot \Delta), \tag{G4}
\]

where we have defined \( \sigma = (p+q)/(4pq) \) and \( \Delta = \mathbf{Q} - \mathbf{P} \). We now transform to polar coordinates and choose the \( x \)-axis to be along the \( \Delta \) such that we can write the scalar product between \( k \) and \( \Delta \) as \( k \cdot \Delta = k \Delta \cos(\phi) \), in the normal way. We let \( k \in (-\infty, \infty) \) and \( \phi \in [0, \pi) \) as this covers the same area as the conventional limits.
The integral can now be written

\[ V_0 = \frac{\pi}{2pq} \int_0^{\pi} d\phi \int_{-\infty}^{\infty} dk \exp(-\sigma k^2) \exp(i k \Delta \cos(\phi)), \]  
\[ \text{G5} \]

where the Coulomb momentum term has been cancelled by the Jacobian from changing to polar coordinates. Completing the square in the exponentials we get

\[ -\sigma k^2 + i k \Delta \cos(\phi) = -\sigma \left( k - \frac{i \Delta \cos(\phi)}{2\sigma} \right)^2 - \frac{\Delta^2 \cos^2(\phi)}{4\sigma}, \]  
\[ \text{G6} \]

where only the squared term is dependent on \( k \). We can then solve the \( k \)-integral leaving us with

\[ V_0 = \frac{\pi}{pq} \sqrt{\frac{\pi}{4\sigma}} \int_0^\pi d\phi \exp\left( -\frac{\Delta^2 \cos^2(\phi)}{4\sigma} \right). \]  
\[ \text{G7} \]

Next we rewrite the argument in the exponential using a trigonometric identity for double angles. That is

\[ -\frac{\Delta^2 \cos^2(\phi)}{4\sigma} = -\frac{\Delta^2}{8\sigma} (1 + \cos(2\phi)), \]  
\[ \text{G8} \]

and we move the constant term outside the integral. Defining \( \theta = 2\phi \) we have \( d\theta / 2 = d\phi \) and a doubling of the integral limits. We are then left with

\[ V_0 = \frac{\pi}{pq} \sqrt{\frac{\pi}{4\sigma}} \int_0^{2\pi} d\phi \exp\left( -\frac{\Delta^2}{8\sigma} \cos(\theta) \right). \]  
\[ \text{G9} \]

The last term in the integral is symmetric over the integration interval. This then gives

\[ V_0 = \frac{\pi^2}{pq} \sqrt{\frac{\pi}{4\sigma}} I_0\left( -\frac{\Delta^2}{8\sigma} \right), \]  
\[ \text{G10} \]

with \( I_0(z) \) being the modified Bessel function of the first kind. Collecting terms we are then left with

\[ V_0 = \frac{\pi^2}{pq} \sqrt{\frac{\pi}{4\sigma}} I_0\left( -\frac{\Delta^2}{8\sigma} \right), \]  
\[ \text{G11} \]

where we have inserted the definition of the exponentially scaled Bessel function from [Equation F2](#).

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