Molecular intergrals from Fast Fourier Transformations (FFT) and matrix multiplications instead of recurrences

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Quantum chemical methods crucially depend on techniques to compute integrals over Gaussian basis functions (GTOs), and efficient algorithms for almost any GTO-based integral quantity are nowadays available. However, such techniques either rely on very deep many-index recurrence relations (McMurchie-Davidson, Obara-Saika, Head-Gordon-Pople, Gill, etc.), recurrence relations coupled with a numerical integration over an auxiliary variable (Rys), or both. They therefore face challenges on hardware platforms like FPGAs, GPUs, or accelerators, which are caused by high numerical accuracy requirements or the complex relations of the very many intermediates, which are hard to reconcile with these platforms' parallel execution capabilities. We here present a completely different approach to computing interal intermediates, for the example of the core-intermediate[1,2] $[\mathbf{r}]^0$ of the McMurchie-Davidson (MD) method. We show that $[\mathbf{r}]^0$ can be expressed with a closed formula directly in terms of the inputs vector elements $[\mathbf{0}]^m$, and that this formula can be evaluated, without any recurrences and only dense Fast Fourier Transformations (FFTs) and matrix multiplications as computational kernels, at a cost of $\mathcal{O}(L)$ for each output $[\mathbf{r}]^0$ (and this highest scaling step is evaluated with a dense matrix multiplication). While the approach's full scope of applicability is not yet obvious, it is clearly not limited to MD, and its core idea may enable integral computation with an unprecedented performance, particularly on novel platforms.

1. McMurchie, Davidson; *"One-and two-electron integrals over Cartesian Gaussian functions"*; J. Comput. Phys. **26**, 218 (1978)
2. Gill, Head-Gordon, Pople; *"An efficient algorithm for the generation of two-electron repulsion integrals over gaussian basis functions"*; Int. J. Quantum. Chem. Symposium **23**, 269 (1989)

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