Electronic Supplementary Information for
Fully relativistic self-consistent field under a magnetic field

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1 Integral Evaluation

The primitive Cartesian gauge-including atomic orbitals (GIAOs) are defined as

$$\phi_p(\mathbf{r}) = N_A(x-A_x)^{a_x}(y-A_y)^{a_y}(z-A_z)^{a_z}e^{-\zeta_a(\mathbf{r}-\mathbf{A})^2}e^{-i\mathcal{A}_A\cdot\mathbf{r}}, \tag{1}$$

$$\phi_q(\mathbf{r}) = N_B(x-B_x)^{b_x}(y-B_y)^{b_y}(z-B_z)^{b_z}e^{-\zeta_b(\mathbf{r}-\mathbf{B})^2}e^{-i\mathcal{A}_B\cdot\mathbf{r}}, \tag{2}$$

$$\phi_r(\mathbf{r}) = N_C(x-C_x)^{c_x}(y-C_y)^{c_y}(z-C_z)^{c_z}e^{-\zeta_c(\mathbf{r}-\mathbf{C})^2}e^{-i\mathcal{A}_C\cdot\mathbf{r}}, \tag{3}$$

$$\phi_s(\mathbf{r}) = N_D(x-D_x)^{d_x}(y-D_y)^{d_y}(z-D_z)^{d_z}e^{-\zeta_d(\mathbf{r}-\mathbf{D})^2}e^{-i\mathcal{A}_D\cdot\mathbf{r}}, \tag{4}$$

where ζ_a is the Gaussian exponent, N_A the normalization constant, and $\mathcal{A}_A$ the magnetic vector potential at center $\mathbf{A}$, which for a uniform magnetic field in the Coulomb gauge is given by

$$\mathcal{A}_A = \frac{1}{2}\mathcal{B}\times(\mathbf{A}-\mathbf{r}_G), \tag{5}$$

where $\mathbf{r}_G$ is the gauge origin. The exponential part of the basis function can be factored so that it resembles a Gaussian orbital:

$$e^{-\zeta_a(\mathbf{r}-\mathbf{A})^2}e^{-i\mathcal{A}_A\cdot\mathbf{r}} = \chi_a e^{-\zeta_a(\mathbf{r}-\tilde{\mathbf{A}})^2}, \tag{6}$$

with

$$\tilde{\mathbf{A}} = \mathbf{A} - i\frac{\mathcal{A}_A}{2\zeta_a}, \tag{7}$$

$$\chi_a = e^{-\mathcal{A}_A{}^2/4\zeta_a}e^{-i\mathbf{A}\cdot\mathcal{A}_A}. \tag{8}$$

The same approach can be used for the other three basis functions as well. In accord with the Gaussian product theorem, the product of two s-type GIAOs is another GIAO:

$$\left(e^{-\zeta_a(\mathbf{r}-\mathbf{A})^2}e^{-i\mathcal{A}_A\cdot\mathbf{r}}\right)^*\left(e^{-\zeta_b(\mathbf{r}-\mathbf{B})^2}e^{-i\mathcal{A}_B\cdot\mathbf{r}}\right) = E_{AB}e^{-p(\mathbf{r}-\mathbf{P})^2}e^{-i\mathcal{A}_{BA}\cdot\mathbf{r}} = \tilde{E}_{ab}e^{-p(\mathbf{r}-\tilde{\mathbf{P}})^2}, \tag{9}$$

$$\left(e^{-\zeta_c(\mathbf{r}-\mathbf{C})^2}e^{-i\mathcal{A}_C\cdot\mathbf{r}}\right)^*\left(e^{-\zeta_d(\mathbf{r}-\mathbf{D})^2}e^{-i\mathcal{A}_D\cdot\mathbf{r}}\right) = E_{CD}e^{-q(\mathbf{r}-\mathbf{Q})^2}e^{-i\mathcal{A}_{DC}\cdot\mathbf{r}} = \tilde{E}_{cd}e^{-q(\mathbf{r}-\tilde{\mathbf{Q}})^2}, \tag{10}$$

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where we introduce the variables

$$p = \zeta_a + \zeta_b, \qquad q = \zeta_c + \zeta_d, \tag{11}$$

$$\mathbf{P} = \frac{\zeta_a \mathbf{A} + \zeta_b \mathbf{B}}{p}, \qquad \mathbf{Q} = \frac{\zeta_c \mathbf{C} + \zeta_d \mathbf{D}}{q}, \tag{12}$$

$$\tilde{\mathbf{P}} = \mathbf{P} - i\frac{\mathcal{A}_{BA}}{2p}, \qquad \tilde{\mathbf{Q}} = \mathbf{Q} - i\frac{\mathcal{A}_{DC}}{2q}, \tag{13}$$

$$\mathcal{A}_{BA} = \mathcal{A}_B - \mathcal{A}_A, \qquad \mathcal{A}_{DC} = \mathcal{A}_D - \mathcal{A}_C, \tag{14}$$

$$E_{ab} = e^{-\zeta_a\zeta_b(\mathbf{A}-\mathbf{B})^2/p}, \qquad E_{cd} = e^{-\zeta_c\zeta_d(\mathbf{C}-\mathbf{D})^2/q}, \tag{15}$$

$$\tilde{E}_{ab} = \chi_a^* \chi_b E_{ab}, \qquad \tilde{E}_{cd} = \chi_c^* \chi_d E_{cd}. \tag{16}$$

1.1 Overlap Integral

The overlap integral is factored in terms of its Cartesian components:

$$\int_{-\infty}^{\infty} \phi_p^* \phi_q \mathrm{d}\mathbf{r} = N_A N_B I_x(a_x, b_x) I_y(a_y, b_y) I_z(a_z, b_z), \tag{17}$$

$$I_x(a_x, b_x) = \int_{-\infty}^{\infty} \tilde{E}_{ab}^x (x - A_x)^{a_x} (x - B_x)^{b_x} e^{-p(x-\tilde{P}_x)} \mathrm{d}x, \tag{18}$$

$$\tilde{E}_{ab}^x = e^{-\zeta_a\zeta_b(\tilde{A}_x - \tilde{B}_x)^2/p} e^{-\mathcal{A}_{BA}^2/4p}, \tag{19}$$

and likewise for I_y and I_z. As with real Gaussian functions, the overlap of two s-functions is obtained analytically, from which higher angular momentum integrals are evaluated using recurrence relations:

$$I_x(0,0) = \tilde{E}_{AB}^x \sqrt{\frac{\pi}{p}}, \tag{20}$$

$$I(a+1,b) = \frac{a}{2p} \cdot I(a-1,b) + \frac{b}{2p} \cdot I(a,b-1) + (\tilde{P} - A) \cdot I(a,b), \tag{21}$$

$$I(a,b+1) = \frac{a}{2p} \cdot I(a-1,b) + \frac{b}{2p} \cdot I(a,b-1) + (\tilde{P} - B) \cdot I(a,b). \tag{22}$$

1.2 Kinetic Energy and Momentum Integrals

When a magnetic field is present, the kinetic energy and momentum operators are defined as

$$\hat{T} = -\frac{1}{2}\nabla^2 - i\nabla \cdot \mathcal{A}(\mathbf{r}) + \frac{1}{2}\mathcal{A}^2(\mathbf{r}), \tag{23}$$

$$\hat{\pi} = -i\nabla + \mathcal{A}(\mathbf{r}). \tag{24}$$

We can factor the operators and basis functions as follows:

$$\hat{T} = \hat{T}_x + \hat{T}_y + \hat{T}_z, \tag{25}$$

$$\hat{\pi} = (\hat{\pi}_x, \hat{\pi}_y, \hat{\pi}_z)^T, \tag{26}$$

$$\phi_A(\mathbf{r}) = X_A(x) Y_A(y) Z_A(z), \tag{27}$$

using

$$\hat{T}_x = \frac{1}{2}\hat{\pi}_x^2, \tag{28}$$

$$X_A(x) = \sqrt[3]{N_A}(x - A_x)^{a_x} e^{-\alpha(x-A_x)^2} e^{-i\mathcal{A}_{Ax}x}. \tag{29}$$

Because we are working in the Coulomb gauge, the gradient and vector potential operators commute. We denote the overlap integrals over functions with incremented or decremented angular momentum as follows:

$$S_{AB}^{x\pm n} = \int_{-\infty}^{\infty} X_A (x - B_x)^{\pm n} X_B dx. \tag{30}$$

Five basic classes of integrals will be needed:

$$\langle X_A | X_B \rangle = S_{AB}^{x}, \tag{31}$$

$$\langle X_A | x | X_B \rangle = S_{AB}^{x+1} + B_x S_{AB}^{x}, \tag{32}$$

$$\langle X_A | x^2 | X_B \rangle = S_{AB}^{x+2} + 2B_x S_{AB}^{x+1} + B_x^2 S_{AB}^{x}, \tag{33}$$

$$\langle X_A | \frac{\partial}{\partial x} | X_B \rangle = b_x S_{AB}^{x-1} - 2\beta S_{AB}^{x+1} - i\mathcal{A}_{Bx} S_{AB}^{x}, \tag{34}$$

$$\begin{aligned} \langle X_A | \frac{\partial^2}{\partial x^2} | X_B \rangle &= b_x(b_x - 1) S_{AB}^{x-2} - i2b_x \mathcal{A}_{Bx} S_{AB}^{x-1} \\ &-\left(2\beta(2b_x{+}1) + \mathcal{A}_{Bx}^2\right) S_{AB}^{x} + i4\beta\mathcal{A}_{Bx} S_{AB}^{x+1} + 4\beta^2 S_{AB}^{x+2}. \end{aligned} \tag{35}$$

The kinetic energy and momentum integrals are then

$$\begin{aligned} \langle A | \hat{T}_x | B \rangle = \frac{-1}{2} & \langle X_A | \frac{\partial^2}{\partial x^2} | X_B \rangle \langle Y_A | Y_B \rangle \langle Z_A | Z_B \rangle \\ + \frac{1}{8} & \mathcal{B}_y^2 \langle X_A | X_B \rangle \langle Y_A | Y_B \rangle \langle Z_A | z^2 | Z_B \rangle \\ + \frac{1}{8} & \mathcal{B}_z^2 \langle X_A | X_B \rangle \langle Y_A | y^2 | Y_B \rangle \langle Z_A | Z_B \rangle \\ - \frac{1}{4} & \mathcal{B}_y \mathcal{B}_z \langle X_A | X_B \rangle \langle Y_A | y | Y_B \rangle \langle Z_A | z | Z_B \rangle \\ + \frac{i}{2} & \mathcal{B}_z \langle X_A | \frac{\partial}{\partial x} | X_B \rangle \langle Y_A | y | Y_B \rangle \langle Z_A | Z_B \rangle \\ - \frac{i}{2} & \mathcal{B}_y \langle X_A | \frac{\partial}{\partial x} | X_B \rangle \langle Y_A | Y_B \rangle \langle Z_A | z | Z_B \rangle, \end{aligned} \tag{36}$$

$$\begin{aligned} \langle A | \hat{\pi}_x | B \rangle = -i & \langle X_A | \frac{\partial}{\partial x} | X_B \rangle \langle Y_A | Y_B \rangle \langle Z_A | Z_B \rangle \\ + \frac{1}{2} & \mathcal{B}_y \langle X_A | X_B \rangle \langle Y_A | Y_B \rangle \langle Z_A | z | Z_B \rangle \\ - \frac{1}{2} & \mathcal{B}_z \langle X_A | X_B \rangle \langle Y_A | y | Y_B \rangle \langle Z_A | Z_B \rangle. \end{aligned} \tag{37}$$

The contributions of $\hat{T}_y$, $\hat{T}_z$, $\hat{\pi}_y$, and $\hat{\pi}_z$ can be obtained by cyclic permutation of the x, y, and z indices in the above equations.

1.3 Electron Repulsion Integral

For the electron repulsion integral (ERI), we use the Rys quadrature scheme, in which the ERI is represented as the product of three polynomials $I_\lambda(t)$ and a weighting function $w(t)$. Each polynomial's rank L_λ is equal to the sum of the angular momentum indices a_λ for the four Cartesian basis functions. This integral can be obtained exactly by Gaussian quadrature using any number of quadrature points greater than $(L_x + L_y + L_z)/2$.

$$\begin{aligned}(pq|rs) &= \iint_{-\infty}^{\infty} \phi_p^*(\mathbf{r}_1)\phi_q(\mathbf{r}_1)\frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}\phi_r^*(\mathbf{r}_2)\phi_s(\mathbf{r}_2)\mathrm{d}\mathbf{r}_1\mathrm{d}\mathbf{r}_2 \\ &= \frac{2\pi^2\sqrt{\pi}}{pq\sqrt{p+q}}\tilde{E}_{AB}\tilde{E}_{CD}\int_0^1 \tilde{I}_x(t)\tilde{I}_y(t)\tilde{I}_z(t)w(t)dt \\ &= \frac{2\pi^2\sqrt{\pi}}{pq\sqrt{p+q}}\tilde{E}_{AB}\tilde{E}_{CD}\sum_g \tilde{I}_x(t_g)\tilde{I}_y(t_g)\tilde{I}_z(t_g)w_g, \end{aligned} \tag{38}$$

$$w(t) = \frac{e^{-\tilde{T}t}}{2\sqrt{t}}, \tag{39}$$

$$\tilde{T} = \frac{pq}{p+q}(\tilde{\mathbf{P}} - \tilde{\mathbf{Q}})^2, \tag{40}$$

The derivation of Eq. (38) follows that of the conventional ERI.[1] The polynomials $\tilde{I}_\lambda(t)$ are obtained using recurrence relations, although the coefficients now include a contribution from the field-dependent wavevector. Starting from the $(ss|ss)$ term, $\tilde{I}_x(0,0,0,0,t) = 1$, the vertical and horizontal recurrence relations are given by

$$\begin{aligned}\tilde{I}_x(a+1,0,c,0,t) = \tilde{C}_{00x} &\cdot \tilde{I}_x(a,0,c,0,t) \\ &+ a_x \cdot B_{10} \cdot \tilde{I}_x(a-1,0,c,0,t) \\ &+ c_x \cdot B_{00} \cdot \tilde{I}_x(a,0,c-1,0,t),\end{aligned} \tag{41}$$

$$\begin{aligned}\tilde{I}_x(a,0,c+1,0,t) = \tilde{D}_{00x} &\cdot \tilde{I}_x(a,0,c,0,t) \\ &+ a_x \cdot B_{00} \cdot \tilde{I}_x(a-1,0,c,0,t) \\ &+ c_x \cdot B_{01} \cdot \tilde{I}_x(a,0,c-1,0,t),\end{aligned} \tag{42}$$

$$\begin{aligned}\tilde{I}_x(a,b+1,c,d,t) &= \tilde{I}_x(a+1,b,c,d,t) \\ &+ (A_x - B_x) \cdot \tilde{I}_x(a,b,c,d,t),\end{aligned} \tag{43}$$

$$\begin{aligned}\tilde{I}_x(a,b,c,d+1,t) &= \tilde{I}_x(a,b,c+1,d,t) \\ &+ (C_x - D_x) \cdot \tilde{I}_x(a,b,c,d,t),\end{aligned} \tag{44}$$

with the coefficients

$$\tilde{C}_{00x} = (\tilde{P}_x - A_x) - \frac{q(\tilde{P}_x - \tilde{Q}_x)}{p+q}t^2, \tag{45}$$

$$\tilde{D}_{00x} = (\tilde{Q}_x - C_x) + \frac{p(\tilde{P}_x - \tilde{Q}_x)}{p+q}t^2, \tag{46}$$

$$B_{00} = \frac{1}{2(p+q)} t^2, \tag{47}$$

$$B_{10} = \frac{1}{2p} - \frac{q}{2p(p+q)} t^2, \tag{48}$$

$$B_{01} = \frac{1}{2q} - \frac{p}{2q(p+q)} t^2. \tag{49}$$

For efficiency, we screen electron repulsion integrals using the following relation:

$$|(pq|rs)| \leq \frac{16(\zeta_a \zeta_b \zeta_c \zeta_d)}{pq\sqrt{\pi(p+q)}} E_{ab} E_{cd} F_0(T), \tag{50}$$

$$T = \frac{pq}{p+q} (\mathbf{P} - \mathbf{Q})^2, \tag{51}$$

where $F_0(T)$ is the Boys function, defined below. Integrals are evaluated in batches corresponding to all basis functions of total angular momenta l_a, l_b, l_c, and l_d on centers **A**, **B**, **C**, and **D**. ($l_a = a_x + a_y + a_z$.) For each batch, we identify $\zeta_a \zeta_b$ and $\zeta_c \zeta_d$ pairs whose corresponding integrals will all be below 5.0×10^{-13} and neglect them. This screening procedure is identical to that used for conventional Gaussian functions[1] and does not explicitly consider the field-dependent wavevector.

The algorithm for nuclear attraction integrals with a point nuclear charge is analogous to the above. When nuclei are represented with a Gaussian charge distribution as in this work, nuclear attraction integrals are evaluated using the ERI code with slight adjustments.

1.4 Complex Gaussian Quadrature

We determine the roots t_g and weights w_g of the quadrature using the Golub–Welsh algorithm[2], which requires as a starting point evaluation of the zeroth-order Boys function with complex argument $F_0(\tilde{T})$:

$$F_n(\tilde{T}) = \int_0^1 \frac{e^{-\tilde{T}t}}{2\sqrt{t}} t^n dt = \int_0^1 e^{-\tilde{T}t^2} t^{2n} dt, \tag{52}$$

$$F_0(\tilde{T}) = \frac{1}{2}\sqrt{\pi}\tilde{T}^{-1/2} \operatorname{erf}\left(\sqrt{\tilde{T}}\right), \tag{53}$$

where higher-order Boys functions are evaluated with well-known recurrence relations.[3] The error function with complex argument must be evaluated numerically; we use the following two relations:

$$\operatorname{erf}(z) \approx \frac{2}{\sqrt{\pi}} \sum_{j=0}^{\infty} \frac{(-1)^j z^{2j+1}}{j!(2j+1)}, \tag{54}$$

$$\operatorname{erf}(z) \approx 1 - e^{-z^2} \pi^{-1/2} \frac{1}{z + \frac{0.5}{z + \frac{1}{z + \frac{1.5}{z + \cdots}}}}. \tag{55}$$

Equation (54) is the Taylor expansion around $z = 0$; we use it when the real part of z is between -0.5 and 0.5. Equation (55) gives a continued fraction used outside of that range, which can be obtained from Eq. (13.2.20a) of Ref. 4. Since Eq. (55) is only valid for $\mathrm{Re}(z) > 0$, we also take advantage of the fact that $\operatorname{erf}(-z) = -\operatorname{erf}(z)$.

To handle the numerical instability and inefficiency of the Golub–Welsh algorithm, we have tabulated quadrature roots and weights using 1024-bit multiple-precision arithmetic.[5,6] At runtime, t_g and w_g are determined by 2-dimensional Chebyshev interpolation, using an algorithm similar to that described in Ref. 7. The current interpolation covers the range of $-2.0 < \mathrm{Re}(\tilde{T}) < \infty$, $0.0 < \mathrm{Im}(\tilde{T}) < 0.5$. For large values of $\mathrm{Re}(\tilde{T})$ we use the approximation

$$t_g \approx t'_g \tilde{T}^{-1}, \tag{56}$$

$$w_g \approx w'_g \tilde{T}^{-1/2}, \tag{57}$$

where the constants t'_g and w'_g are determined numerically. Interpolation across this range requires approximately 30 MB of storage, and is sufficient for molecules of radius 100 Å in 100 T fields (or smaller molecules in stronger fields).

To address arbitrarily strong magnetic fields or use this integral code for a general mixed Gaussian and plane wave basis set, a wider interpolation range would need to be generated. One substantial complication arises due to the presence of poles in the complex function defining the roots and weights. The details of the algorithm are given elsewhere,[1,8] but one key step is the determination of eigenvalues and eigenvectors of a tridiagonal matrix. In the two-root case, this matrix is given by

$$\mathbf{J} = \begin{pmatrix} a_0 & \sqrt{b_1} \\ \sqrt{b_1} & a_1 \end{pmatrix}, \tag{58}$$

with

$$a_0 = \frac{F_1}{F_0}, \tag{59}$$

$$a_1 = \frac{F_3 - 2F_2F_1/F_0 + F_1^3/F_0^2}{F_2 - F_1^2/F_0}, \tag{60}$$

$$b_1 = \frac{F_2}{F_0} - \frac{F_1^2}{F_0^2}. \tag{61}$$

The denominator of a_1 falls to zero at approximately $\tilde{T} = 3.375 + 7.977i$ and $\tilde{T} = -6.015 + 7.131i$, causing the values for the roots to diverge. Close to these poles, the roots cannot be obtained by polynomial interpolation. The simplest solution would be to use 3-root quadrature for this case, but this has not been necessary for the current work due to the moderate field strengths used.

1.5 Density Fitting with GIAOs

The density fitting approximation expands the 4-index integrals as a product of 2- and 3-index integrals:

$$(pq|rs) = \sum_{\gamma\delta} (pq|\gamma) \mathbf{J}^{-1}_{\gamma\delta} (\delta|rs), \tag{62}$$

$$(pq|\gamma) = \iint_{-\infty}^{\infty} \phi_p^*(\mathbf{r}_1)\phi_q(\mathbf{r}_1) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \chi_\gamma^*(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2, \tag{63}$$

$$(\delta|rs) = \iint_{-\infty}^{\infty} \chi_\delta(\mathbf{r}_1) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \phi_r^*(\mathbf{r}_2)\phi_s(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2, \tag{64}$$

$$\mathbf{J}_{\gamma\delta} = \iint_{-\infty}^{\infty} \chi_\gamma^*(\mathbf{r}_1) \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \chi_\delta(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2, \tag{65}$$

which follows from the expansion of orbital products in a fitting basis

$$\phi_p^*(\mathbf{r}_1)\phi_q(\mathbf{r}_1) \approx \sum_\delta C_\delta^{pq}\chi_\delta(\mathbf{r}_1). \tag{66}$$

Using $|\widetilde{pq})$ to denote the right-hand side of Eq. (66), we minimize the Coulomb integral of the residual[9] given by

$$(qp - \widetilde{qp}|pq - \widetilde{pq}), \tag{67}$$

which leads to the optimized coefficients

$$C_\delta^{pq} = \sum_\gamma (pq|\gamma)\mathbf{J}^{-1}_{\gamma\delta}. \tag{68}$$

Any fitting basis with sufficient flexibility should work, but the use of real Gaussian functions improves the efficiency of the 2-index matrix inversion and simplifies the computation of Coulomb and exchange matrices due to the identity

$$(\delta|pq) = (\delta|qp)^*. \tag{69}$$

2 Magnetizabilities

Isotropic and anisotropic magnetizabilities were computed using a 7-point finite difference formula at magnetic field increments of 0.002 a.u. (Tables S1-S10). XZ denotes cc-pVXZ for the F, N, P, and As atoms and Dyall's XZ spin-free basis sets for Sb and Bi. AXZ denotes addition of diffuse functions. The fitting basis for F, N, P, and As atoms was cc-pV5Z-JKFIT+1 with the diffuse functions from aug-cc-pV5Z, while an even-tempered fitting basis was used for Sb and Bi. All basis sets were used in uncontracted form.

2.1 Isotropic Magnetizabilities

Table S1 NF_3 Isotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	−5.06	−5.06	−5.07	−5.10	−5.09	−5.08
Coulomb	RMB	−5.05	−5.05	−5.06	−5.09	−5.08	−5.07
Gaunt	RMB	−5.06	−5.05	−5.06	−5.09	−5.08	−5.07
Coulomb*	sMB	−4.67	−4.76	−4.83	−5.00	−5.04	−5.04

* Taken from Ref. 10.

Table S2 PF_3 Isotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	−6.73	−6.60	−6.57	−6.77	−6.65	−6.59
Coulomb	RMB	−6.72	−6.59	−6.56	−6.76	−6.63	−6.58
Gaunt	RMB	−6.72	−6.59	−6.56	−6.76	−6.64	−6.58
Coulomb*	sMB	−6.43	−6.40	−6.41	−6.69	−6.61	−6.54

* Taken from Ref. 10.

Table S3 AsF_3 Isotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	−8.46	−8.44	−8.42	−8.52	−8.46	−8.44
Coulomb	RMB	−8.40	−8.38	−8.37	−8.46	−8.41	−8.38
Gaunt	RMB	−8.40	−8.39	−8.37	−8.46	−8.41	−8.38
Coulomb*	sMB	−8.11	−8.21	−8.24	−8.37	−8.37	−8.36

* Taken from Ref. 10.

Table S4 SbF_3 Isotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	−11.35	−11.35	−11.32	−11.36	−11.28	−11.26
Coulomb	RMB	−11.18	−11.19	−11.17	−11.19	−11.13	−11.10
Gaunt	RMB	−11.18	−11.20	−11.17	−11.20	−11.13	−11.11
Coulomb*	sMB	−10.86	−11.01	−10.98	−11.08	−11.08	−11.06

* Taken from Ref. 10.

Table S5 BiF_3 Isotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	−13.66	−13.48	−13.45	−13.64	−13.44	−13.42
Coulomb	RMB	−12.96	−12.83	−12.83	−12.97	−12.80	−12.79
Gaunt	RMB	−12.97	−12.84	−12.84	−12.98	−12.81	−12.80
Coulomb*	sMB	−12.67	−12.72	−12.74	−12.90	−12.79	−12.79

* Taken from Ref. 10.

2.2 Anisotropic Magnetizabilities

Table S6 NF_3 Anisotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	−0.61	−0.60	−0.60	−0.63	−0.60	−0.60
Coulomb	RMB	−0.61	−0.60	−0.60	−0.64	−0.60	−0.60
Gaunt	RMB	−0.61	−0.60	−0.60	−0.64	−0.60	−0.60
Coulomb*	sMB	−0.60	−0.60	−0.61	−0.65	−0.60	−0.60

* Taken from Ref. 10.

Table S7 PF_3 Anisotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	0.33	0.37	0.36	0.25	0.33	0.34
Coulomb	RMB	0.33	0.36	0.36	0.24	0.32	0.33
Gaunt	RMB	0.33	0.36	0.36	0.24	0.32	0.33
Coulomb*	sMB	0.37	0.39	0.37	0.23	0.33	0.33

* Taken from Ref. 10.

Table S8 AsF_3 Anisotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	0.29	0.26	0.24	0.22	0.23	0.23
Coulomb	RMB	0.26	0.23	0.21	0.19	0.20	0.20
Gaunt	RMB	0.26	0.23	0.21	0.19	0.20	0.20
Coulomb*	sMB	0.29	0.25	0.22	0.17	0.20	0.20

* Taken from Ref. 10.

Table S9 SbF_3 Anisotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	0.42	0.38	0.35	0.33	0.33	0.33
Coulomb	RMB	0.33	0.29	0.27	0.26	0.25	0.25
Gaunt	RMB	0.33	0.29	0.27	0.26	0.25	0.25
Coulomb*	sMB	0.38	0.30	0.26	0.23	0.26	0.26

* Taken from Ref. 10.

Table S10 BiF_3 Anisotropic Magnetizability / a.u.

Hamiltonian	Approach	DZ	TZ	QZ	ADZ	ATZ	AQZ
Non-Rel	N/A	0.40	0.31	0.28	0.30	0.27	0.27
Coulomb	RMB	0.19	0.11	0.10	0.11	0.09	0.09
Gaunt	RMB	0.19	0.11	0.10	0.11	0.09	0.09
Coulomb*	sMB	0.24	0.13	0.10	0.10	0.10	0.10

* Taken from Ref. 10.

3 Wall-clock times for Dirac–Hartree–Fock calculations

Startup timings include the computation of all 1-electron integrals, the 2-index and 3-index electron repulsion integrals, and inversion of the 2-index integral matrix.

Table S11 Wall-clock times, in seconds, for parallel Hartree–Fock computations across 1024 CPU cores.

Molecule	Orb. basis	Aux. basis	Startup, 0 *T*	Startup, 100 *T*	One Iter., 0 *T*	One Iter., 100 *T*
			Non-Relativistic			
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT	4.4	8.2	3.0	7.6
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT+1	14.8	20.4	7.8	18.3
$C_{36}H_{62}B_2Cl_3In_2LiN_4O_2$	SVP + DZ	TZVPP-JKFIT	8.6	15.6	8.0	19.7
			Dirac–Coulomb			
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT	12.8	37.5	218	245
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT+1	28.9	69.0	628	682
$C_{36}H_{62}B_2Cl_3In_2LiN_4O_2$	SVP + DZ	TZVPP-JKFIT	27.5	75.7	652	721
			Dirac–Coulomb–Gaunt			
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT+1	34.8	95.6	1330	1410

4 Converged Hartree–Fock and Dirac–Hartree–Fock energies

Table S12 Total energies (in E_h) for the molecules used in timing tests.

Molecule	Orb. basis	Aux basis	$\mathcal{B}/T$	Non-Rel.	Coulomb	Gaunt
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT	0.0	−21624.842168	−23096.426979	
			100.0	−21624.842159	−23096.426971	
$C_{32}H_{47}BiN_4O$	SVP + DZ	TZVPP-JKFIT+1	0.0	−21624.845128	−23096.429834	−23067.593564
			100.0	−21624.845120	−23096.429326	−23067.593556
$C_{36}H_{62}B_2Cl_3In_2LiN_4O_2$	SVP + DZ	TZVPP-JKFIT	0.0	−14680.457214	−14966.094341	
			100.0	−14680.457203	−14966.094330	

5 Geometries

The coordinates (in Å) of the large molecules used for timing benchmarks are below. The magnetic field vector used for timing tests was $(60.0, 0.0, -80.0)^T$ T. The results shown in Fig. 3 in the main text were obtained with a magnetic field oriented along the x-axis.

$C_{37}H_{47}BiN_4O$

Atom	x	y	z
Bi	-1.133828616	0.315661652	-0.550328248
O	-0.153516644	-0.576642336	-2.340563620
N	-1.724857169	1.067961666	1.982349101
N	-1.899112511	-3.067714482	-1.312399052
N	3.722865597	-0.639488822	0.413820443
N	-1.277164936	2.861336218	-2.227564550
C	-0.529997877	-1.364763396	0.914063515
C	-0.392450589	-0.986958770	2.264719810
C	-0.003322961	-1.915867570	3.227624241
H	0.076103743	-1.650402852	4.136695407
C	0.266697626	-3.224467842	2.862726122
H	0.569687893	-3.849304152	3.512026394
C	0.093098319	-3.617076952	1.545334722
H	0.261107580	-4.519860192	1.300291795
C	-0.323259384	-2.707181754	0.568464113
C	-0.588554329	0.448350468	2.677864722
H	-0.742975600	0.487538808	3.654227470
H	0.234693426	0.960701728	2.478529285
C	-2.988093520	0.415258092	2.356134787
H	-3.145909764	0.535435668	3.316499914
H	-2.934313850	-0.541517972	2.150958949
H	-3.724786216	0.817011148	1.851320966
C	-1.795914164	2.500077780	2.312204812
H	-1.937642634	2.606612008	3.275871035
H	-2.541644175	2.909958788	1.825927917
H	-0.957849983	2.937535768	2.054465361
C	-0.517241555	-3.202406258	-0.844405152
H	0.079541502	-2.692522412	-1.449775449
H	-0.252527558	-4.155553772	-0.891128363
C	-2.799832378	-3.917956318	-0.533086367
H	-2.768818340	-3.649008192	0.408995763
H	-2.518060200	-4.853686792	-0.614344125
H	-3.713950742	-3.820275752	-0.870813924
C	-1.947424099	-3.460759018	-2.722475082
H	-2.860604168	-3.352918512	-3.059694778
H	-1.674982761	-4.397940912	-2.810842894
H	-1.337673354	-2.894269792	-3.239985428
C	0.744782128	1.635321744	-0.310389325
C	2.044542847	1.177108450	-0.023701798
C	3.101140689	2.095712168	0.025814649
H	3.976729018	1.790913968	0.233783723
C	2.886875617	3.442920212	-0.225322609
H	3.612720201	4.055129168	-0.187740896
C	1.618058625	3.891844418	-0.531562785
H	1.474250748	4.814221828	-0.710837713
C	0.544278407	3.000091970	-0.578793856
C	2.313072685	-0.287229188	0.262985730
H	1.836060955	-0.537163712	1.094608096
H	1.931486675	-0.827447712	-0.474682354
C	3.831084100	-1.890612862	1.164692912
H	3.395846234	-1.786836332	2.036690226
H	4.777153876	-2.111954412	1.295213186
H	3.393913923	-2.611242892	0.665465562
C	4.379208548	-0.792903916	-0.881732935
H	5.329911181	-0.987103912	-0.746387982
H	4.284910852	0.036147188	-1.396450045
H	3.961827220	-1.531386412	-1.373596301
C	-0.813677451	3.522748312	-1.007428529
H	-1.468724139	3.368607508	-0.281695179
H	-0.754449924	4.497812268	-1.165373296
C	-0.399380107	3.167150412	-3.362633856
H	-0.737401681	2.719822768	-4.166831729
H	0.507015180	2.848999148	-3.168884890
H	-0.381352416	4.136408688	-3.509151751
C	-2.646954664	3.271072084	-2.524917158
H	-2.945392478	2.837387788	-3.351714845
H	-2.680265239	4.243813768	-2.635630853
H	-3.234757222	3.005752508	-1.787503005
C	-0.335957431	-0.197676574	-3.609962157
C	0.825379919	0.053854512	-4.380133344
C	0.703959481	0.383181710	-5.729012125
H	1.486785337	0.539789928	-6.243983166
C	-0.540061663	0.487393666	-6.334382421
H	-0.612607691	0.719766008	-7.252087225
C	-1.677270834	0.246603088	-5.577669551
H	-2.529541272	0.316271248	-5.990052672
C	-1.603507575	-0.096222316	-4.225235743
C	2.160976169	-0.013201092	-3.710264702
H	2.864619230	0.179837768	-4.364897514
H	2.297071530	-0.910178652	-3.341557625
H	2.197578197	0.647195008	-2.986054934
C	-2.885467180	-0.326562670	-3.462682471
H	-3.584062836	-0.625700332	-4.080495361
H	-3.163840337	0.509310108	-3.031762423
H	-2.739405186	-1.013229472	-2.777831930

$C_{36}H_{62}B_2Cl_3In_2LiN_4O_2$			
Atom	*x*	*y*	*z*
In	-2.966130077	0.435211239	0.941685939
In	0.119550696	0.015359619	0.383783387
Cl	-4.697430952	1.286872599	2.323137409
Cl	1.664157202	-0.042100161	2.291024891
Cl	2.164020455	0.729097239	-0.944916414
N	-3.174289893	0.976772199	-1.081434958
N	-1.270005184	1.699731759	0.197095086
N	-0.661898934	-1.751503281	-0.435795843
N	-1.786102785	-1.182783201	1.617146125
C	-4.262877507	0.671536119	-2.018507274
C	-4.812292036	-0.710336121	-1.669949292
H	-5.112402763	-0.716416521	-0.735620256
H	-4.109331044	-1.383233721	-1.790976369
H	-5.569076438	-0.919096521	-2.255720345
C	-5.373864750	1.711689879	-1.850683060
H	-5.663012883	1.731957879	-0.914740330
H	-6.133753300	1.474554279	-2.421930865
H	-5.036789619	2.595374679	-2.107260464
C	-3.807274502	0.637485879	-3.470832200
H	-3.096455062	-0.031358121	-3.575722334
H	-3.464771771	1.519143879	-3.727409604
H	-4.563414535	0.402377079	-4.046921088
C	-0.908741680	3.004180239	0.874846718
C	-0.747804042	2.761572279	2.375582475
H	-1.578594613	2.388641079	2.737050013
H	-0.549692695	3.610801479	2.824189508
H	-0.011767433	2.131237479	2.527269745
C	0.373865687	3.580399479	0.329418024
H	1.099505605	2.933850279	0.447217712
H	0.591872024	4.407333879	0.811912638
H	0.264260776	3.779025879	-0.624275345
C	-2.044283095	4.008054279	0.694112950
H	-2.875395850	3.629042679	1.050739404
H	-2.157958705	4.204653879	-0.259580419
H	-1.829097952	4.834988679	1.174993870
C	-0.254084890	-2.333802921	-1.745792927
C	-1.255355013	-3.345176121	-2.294449010
H	-2.132713710	-2.917521321	-2.388043283
H	-1.324346505	-4.103199321	-1.679631458
H	-0.951378074	-3.661356921	-3.170685049
C	1.122623308	-2.974271721	-1.590878268
H	1.752784458	-2.313534921	-1.235865508
H	1.436313336	-3.288425721	-2.463886918
H	1.063344906	-3.732294921	-0.972833327
C	-0.176701924	-1.204875321	-2.785012096
H	0.458271388	-0.521843721	-2.480023862
H	-1.063275968	-0.801542121	-2.894743313
H	0.124493052	-1.571726121	-3.641883803
C	-1.927227174	-1.589967321	3.064629968
C	-1.721722244	-0.359699721	3.950548173
H	-2.410882791	0.307117479	3.748836378
H	-0.836302585	0.021338679	3.777882876
H	-1.786552297	-0.621156921	4.892945681
C	-3.298679383	-2.198007321	3.376072980
H	-3.439284559	-2.988459321	2.812893648
H	-3.998870687	-1.537270521	3.192111823
H	-3.334907141	-2.457437721	4.320084182
C	-0.826329119	-2.597286921	3.418029034
H	-0.930795736	-3.401926521	2.867759256
H	-0.894907688	-2.838476121	4.365267624
H	0.051056535	-2.195980521	3.246977431
C	-1.553278178	2.674014519	-2.367065256
C	-0.366426177	2.414989479	-3.046430583
H	0.200594741	1.717770279	-2.743056043
C	0.008283146	3.162878679	-4.164720776
H	0.810812236	2.946011079	-4.624623669
C	-0.764831616	4.198573479	-4.608486726
H	-0.501755864	4.709327079	-5.365309382
C	-1.934580891	4.490432679	-3.937189871
H	-2.481960839	5.207919879	-4.230882245
C	-2.318505562	3.738489879	-2.835036621
H	-3.127214015	3.957384279	-2.386429589
C	-2.320291371	-3.605214561	0.563403706
C	-1.643274762	-4.820686521	0.745751169
H	-0.696538608	-4.816632921	0.821594804
C	-2.324926002	-6.028659321	0.819981110
H	-1.840667657	-6.833298921	0.960372519
C	-3.682168789	-6.071222121	0.692499255
H	-4.141757241	-6.900183321	0.739296392
C	-4.388754246	-4.893651321	0.494014849
H	-5.334885264	-4.913919321	0.406875353
C	-3.708245116	-3.681624921	0.423012297
H	-4.202431341	-2.885092521	0.276166110
O	4.429149938	2.378507079	1.239541644
C	3.996871098	3.744570279	0.819981110
H	3.217351446	4.030349079	1.360568721
H	3.727259382	3.732409479	-0.132098564
C	5.133593855	4.707300279	1.018465516
H	5.138761513	5.408573079	0.321349552
H	5.098000164	5.132928279	1.910838499
C	6.261157152	3.837803079	0.895824745
H	7.050808736	4.210734279	1.363796110
H	6.492348931	3.685793079	-0.054641235
C	5.782392161	2.518356279	1.571962683
H	6.302814049	1.746145479	1.236314255
H	5.892573278	2.571053079	2.554702550
O	4.821944762	-0.720470121	0.861937163
C	5.704241210	-1.073133321	1.865655057
H	5.245470277	-1.042731321	2.740277401
H	6.456217832	-0.428610921	1.889860472
C	6.218787289	-2.416901721	1.625214597
H	5.900283125	-3.039129321	2.323944256
H	7.209753868	-2.414874921	1.631669374
C	5.711011897	-2.826315321	0.295530442
H	5.520967433	-3.795125721	0.253574389
H	6.334242354	-2.577018921	-0.432245715
C	4.432235891	-1.987220121	0.266483944
H	4.115279302	-1.859531721	-0.663004009
H	3.714274481	-2.420955321	0.794162000
B	-2.030486161	1.772493879	-1.137430152
B	-1.563180403	-2.200034121	0.542425680
Li	3.537761758	0.696263079	0.955531436

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