A Tale of Two Vectors

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Supplementary Material

A Tale of Two Vectors: A Lanczos Algorithm For Calculating RPA Mean Excitation Energies

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S1 Proof for Preservation of Sum Rules

The RPA matrix, as seen in Eq. (4) in the main article, can be expanded in its (right and left) eigenvectors:

$$\mathbf{E} = \sum_n^{2N} \mathbf{E} \tilde{\mathbf{X}}_n^R (\tilde{\mathbf{X}}_n^L)^T = \sum_n^{2N} \omega_n \tilde{\mathbf{X}}_n^R (\tilde{\mathbf{X}}_n^L)^T \quad (\text{S1})$$

Here, the eigenvalue relation, $\mathbf{E} \tilde{\mathbf{X}}_n^R = \omega_n \tilde{\mathbf{X}}_n^R$, and the resolution of identity, $\mathbf{I}^{(2N)} = \sum_n^{2N} \tilde{\mathbf{X}}_n^R (\tilde{\mathbf{X}}_n^L)^T$, were applied, where the latter can be used since the full set of the RPA eigenvectors is a complete basis. Utilizing the RPA matrix block structure, Eq. (S1) can be re-expressed as:

$$\begin{pmatrix} \mathbf{A} & \mathbf{B} \\ -\mathbf{B} & -\mathbf{A} \end{pmatrix} = \sum_n^N \omega_n \left[\begin{pmatrix} e\tilde{\mathbf{X}}_n \\ d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} e\tilde{\mathbf{X}}_n & -d\tilde{\mathbf{X}}_n \end{pmatrix} - \begin{pmatrix} d\tilde{\mathbf{X}}_n \\ e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} -d\tilde{\mathbf{X}}_n & e\tilde{\mathbf{X}}_n \end{pmatrix} \right] \quad (\text{S2})$$

where the sum is over the positive eigenvalues only. Multiplying the RPA matrix in this expression with itself would change the sign in the parentheses:

$$\begin{aligned}
\begin{pmatrix} \mathbf{A} & \mathbf{B} \\ -\mathbf{B} & -\mathbf{A} \end{pmatrix}^2 &= \sum_n^N \omega_n \sum_m^N \omega_m \left[\begin{pmatrix} e\tilde{\mathbf{X}}_n \\ d\tilde{\mathbf{X}}_n \end{pmatrix} \underbrace{\begin{pmatrix} e\tilde{\mathbf{X}}_n & -d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} e\tilde{\mathbf{X}}_m \\ d\tilde{\mathbf{X}}_m \end{pmatrix}}_{\delta_{mn}} \begin{pmatrix} e\tilde{\mathbf{X}}_m & -d\tilde{\mathbf{X}}_m \end{pmatrix} \right. \\
&\quad - \underbrace{\begin{pmatrix} e\tilde{\mathbf{X}}_n \\ d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} e\tilde{\mathbf{X}}_n & -d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} d\tilde{\mathbf{X}}_m \\ e\tilde{\mathbf{X}}_m \end{pmatrix}}_0 \begin{pmatrix} -d\tilde{\mathbf{X}}_m & e\tilde{\mathbf{X}}_m \end{pmatrix} \\
&\quad - \underbrace{\begin{pmatrix} d\tilde{\mathbf{X}}_n \\ e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} -d\tilde{\mathbf{X}}_n & e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} e\tilde{\mathbf{X}}_m \\ d\tilde{\mathbf{X}}_m \end{pmatrix}}_0 \begin{pmatrix} e\tilde{\mathbf{X}}_m & -d\tilde{\mathbf{X}}_m \end{pmatrix} \\
&\quad \left. + \underbrace{\begin{pmatrix} d\tilde{\mathbf{X}}_n \\ e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} -d\tilde{\mathbf{X}}_n & e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} d\tilde{\mathbf{X}}_m \\ e\tilde{\mathbf{X}}_m \end{pmatrix}}_{\delta_{mn}} \begin{pmatrix} -d\tilde{\mathbf{X}}_m & e\tilde{\mathbf{X}}_m \end{pmatrix} \right] \\
&= \sum_n^N \omega_n^2 \left[\begin{pmatrix} e\tilde{\mathbf{X}}_n \\ d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} e\tilde{\mathbf{X}}_n & -d\tilde{\mathbf{X}}_n \end{pmatrix} + \begin{pmatrix} d\tilde{\mathbf{X}}_n \\ e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} -d\tilde{\mathbf{X}}_n & e\tilde{\mathbf{X}}_n \end{pmatrix} \right] \tag{S3}
\end{aligned}$$

Here, all the terms individually collapse to a single sum over n due to orthogonality of the full set of the RPA eigenvectors, and the second and third terms vanished due to the inner product inside these expressions then evaluating to zero. This can be easily generalized for the RPA matrix to some power m :

$$\begin{pmatrix} \mathbf{A} & \mathbf{B} \\ -\mathbf{B} & -\mathbf{A} \end{pmatrix}^m = \sum_n^N \omega_n^m \left[\begin{pmatrix} e\tilde{\mathbf{X}}_n \\ d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} e\tilde{\mathbf{X}}_n & -d\tilde{\mathbf{X}}_n \end{pmatrix} + (-1)^m \begin{pmatrix} d\tilde{\mathbf{X}}_n \\ e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} -d\tilde{\mathbf{X}}_n & e\tilde{\mathbf{X}}_n \end{pmatrix} \right] \tag{S4}$$

Note that for odd m , there is a minus sign in the square brackets. When computing some sum-over-states property, $M(m)$, using the appropriate property operator $\hat{O}$, the expression

usually takes such form:

$$M(m) = \sum_n \omega_n^m |\langle n | \hat{O} | 0 \rangle|^2 \quad (\text{S5})$$

For RPA, inserting Eq. (8) into Eq. (S5), yields:

$$\begin{aligned} M(m) &= \sum_n^N \omega_n^m (\mathbf{P}^L(\hat{O}))^T ({}^e\tilde{\mathbf{X}}_n^R) ({}^e\tilde{\mathbf{X}}_n^L) \mathbf{P}^R(\hat{O}) \\ &= \sum_n^N (\mathbf{P}^L(\hat{O}))^T \begin{pmatrix} {}^e\tilde{\mathbf{X}}_n \\ {}^d\tilde{\mathbf{X}}_n \end{pmatrix} \omega_n^m \begin{pmatrix} {}^e\tilde{\mathbf{X}}_n & -{}^d\tilde{\mathbf{X}}_n \end{pmatrix} \mathbf{P}^R(\hat{O}) \end{aligned} \quad (\text{S6})$$

The product of the RPA gradient property vector (see Eq. (4) in the main article) and the RPA de-excitation eigenvector can be expressed through the analogous product with the RPA excitation eigenvector:

$$\begin{pmatrix} {}^e\mathbf{P}^T(\hat{O}) & -{}^e\mathbf{P}^T(\hat{O}) \end{pmatrix} \begin{pmatrix} {}^e\tilde{\mathbf{X}}_n \\ {}^d\tilde{\mathbf{X}}_n \end{pmatrix} = - \begin{pmatrix} {}^e\mathbf{P}^T(\hat{O}) & -{}^e\mathbf{P}^T(\hat{O}) \end{pmatrix} \begin{pmatrix} {}^d\tilde{\mathbf{X}}_n \\ {}^e\tilde{\mathbf{X}}_n \end{pmatrix} \quad (\text{S7})$$

$$\begin{pmatrix} {}^e\tilde{\mathbf{X}}_n & -{}^d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} {}^e\mathbf{P}(\hat{O}) \\ {}^e\mathbf{P}(\hat{O}) \end{pmatrix} = \begin{pmatrix} -{}^d\tilde{\mathbf{X}}_n & {}^e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} {}^e\mathbf{P}(\hat{O}) \\ {}^e\mathbf{P}(\hat{O}) \end{pmatrix} \quad (\text{S8})$$

Knowing this, Eq. (S6) can be rewritten as an expansion in a linear combination of the RPA excitation and de-excitation eigenvectors:

$$\begin{aligned} M(m) &= \frac{1}{2} \sum_n^N \left[(\mathbf{P}^L(\hat{O}))^T \begin{pmatrix} {}^e\tilde{\mathbf{X}}_n \\ {}^d\tilde{\mathbf{X}}_n \end{pmatrix} \omega_n^m \begin{pmatrix} {}^e\tilde{\mathbf{X}}_n & -{}^d\tilde{\mathbf{X}}_n \end{pmatrix} \mathbf{P}^R(\hat{O}) \right. \\ &\quad \left. - (\mathbf{P}^L(\hat{O}))^T \begin{pmatrix} {}^d\tilde{\mathbf{X}}_n \\ {}^e\tilde{\mathbf{X}}_n \end{pmatrix} \omega_n^m \begin{pmatrix} -{}^d\tilde{\mathbf{X}}_n & {}^e\tilde{\mathbf{X}}_n \end{pmatrix} \mathbf{P}^R(\hat{O}) \right] \\ &= \frac{1}{2} (\mathbf{P}^L(\hat{O}))^T \sum_n^N \omega_n^m \left[\begin{pmatrix} {}^e\tilde{\mathbf{X}}_n \\ {}^d\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} {}^e\tilde{\mathbf{X}}_n & -{}^d\tilde{\mathbf{X}}_n \end{pmatrix} - \begin{pmatrix} {}^d\tilde{\mathbf{X}}_n \\ {}^e\tilde{\mathbf{X}}_n \end{pmatrix} \begin{pmatrix} -{}^d\tilde{\mathbf{X}}_n & {}^e\tilde{\mathbf{X}}_n \end{pmatrix} \right] \mathbf{P}^R(\hat{O}) \end{aligned} \quad (\text{S9})$$

Recognizing Eq. (S4) here, a final expression for the odd- m -power energy-weighted sum-over-states properties can be written as:

$$M(m) = \frac{1}{2}(\mathbf{P}^L(\hat{O}))^T \begin{pmatrix} \mathbf{A} & \mathbf{B} \\ -\mathbf{B} & -\mathbf{A} \end{pmatrix}^m \mathbf{P}^R(\hat{O}) = \frac{1}{2}(\mathbf{P}^L(\hat{O}))^T \mathbf{E}^m \mathbf{P}^R(\hat{O}) \quad (\text{S10})$$

The sum of the oscillator strengths, $S(0)$ (see Eq. (2) in the main article), which is of interest in this project, fulfills the odd- m -power requirement and can be written as $S(0) = \frac{2}{3}M(1)$ (see, e.g.,¹). It can thus be expressed as:

$$S(0) = \frac{1}{3}(\mathbf{P}^L(\hat{O}))^T \begin{pmatrix} \mathbf{A} & \mathbf{B} \\ -\mathbf{B} & -\mathbf{A} \end{pmatrix} \mathbf{P}^R(\hat{O}) = \frac{1}{3}(\mathbf{P}^L(\hat{O}))^T \mathbf{E} \mathbf{P}^R(\hat{O}) \quad (\text{S11})$$

The RPA matrix $\mathbf{E}$ can be expressed through a general projection into the (nonsymmetric) Lanczos subspace of size i (see Eq. (9) in the main article) in the following manner:

$$\begin{aligned} \mathbf{T}^{(i)} &= \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \\ \mathbf{E} &\approx \mathbf{Q}^{(i)} \mathbf{T}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \\ \implies \mathbf{E} &\approx \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \end{aligned} \quad (\text{S12})$$

If the Lanczos algorithm is initiated with (normalized) gradient property vectors (or a set of vectors that are linear combinations of them) as the first Lanczos vector or block, and the projection in Eq. (S12) is inserted in (S11):

$$S(0) \approx \frac{1}{3}(\mathbf{P}^L(\hat{O}))^T \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{P}^R(\hat{O}) \quad (\text{S13})$$

the projection then can be applied to the gradient property vectors on both sides:

$$\begin{aligned}
(\mathbf{P}^L(\hat{O}))^T \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} &= \sum_j (\mathbf{P}^L(\hat{O}))^T \mathbf{Q}_j \tilde{\mathbf{Q}}_j^T = (\mathbf{P}^L(\hat{O}))^T \mathbf{Q}_1 \tilde{\mathbf{Q}}_1^T \\
&= (\mathbf{P}^L(\hat{O}))^T \frac{\mathbf{P}^R(\hat{O})}{\|\mathbf{P}^R(\hat{O})\|_2} \frac{(\mathbf{P}^L(\hat{O}))^T}{\|\mathbf{P}^R(\hat{O})\|_2} = (\mathbf{P}^L(\hat{O}))^T
\end{aligned} \tag{S14}$$

$$\begin{aligned}
\mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{P}^R(\hat{O}) &= \sum_j \mathbf{Q}_j \tilde{\mathbf{Q}}_j^T \mathbf{P}^R(\hat{O}) = \mathbf{Q}_1 \tilde{\mathbf{Q}}_1^T \mathbf{P}^R(\hat{O}) \\
&= \frac{\mathbf{P}^R(\hat{O})}{\|\mathbf{P}^R(\hat{O})\|_2} \frac{(\mathbf{P}^L(\hat{O}))^T}{\|\mathbf{P}^R(\hat{O})\|_2} \mathbf{P}^R(\hat{O}) = \mathbf{P}^R(\hat{O})
\end{aligned} \tag{S15}$$

which leaves both gradient property vectors unchanged, and in turn, the exact RPA expression for $S(0)$ in (S11) is recovered exactly from Eq. (S13):

$$\begin{aligned}
S(0) &= \frac{1}{3} (\mathbf{P}^L(\hat{O}))^T \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{P}^R(\hat{O}) \\
&= \frac{1}{3} (\mathbf{P}^L(\hat{O}))^T \mathbf{E} \mathbf{P}^R(\hat{O})
\end{aligned} \tag{S16}$$

This means that as long a nonsymmetric or nonsymmetric block Lanczos is initiated with gradient property vectors (or some linear combination of them) as the first Lanczos vectors (or blocks of vectors), for $m = 1$, the sum-over-states properties will be preserved by the algorithm.

The Eq. (S12) can be generalized to all odd m powers of $\mathbf{E}$. Using (S12) to express $\mathbf{E}^m$ and writing it out for the next odd power, $m = 3$:

$$\mathbf{E}^m \approx (\mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T})^m \tag{S17}$$

$$\mathbf{E}^3 = (\mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T}) (\mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T}) (\mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T} \mathbf{E} \mathbf{Q}^{(i)} \tilde{\mathbf{Q}}^{(i)T}) \tag{S18}$$

One can now write the Eq. (S10) for $M(3)$ as:

$$\begin{aligned}
M(3) &= \frac{1}{2}(\mathbf{P}^L(\hat{O}))^T(\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T})(\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T})\dots \\
&\dots(\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T})\mathbf{P}^R(\hat{O}) = \\
&\frac{1}{2}(\mathbf{P}^L(\hat{O}))^T\mathbf{E}(\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T})\mathbf{E}\mathbf{P}^R(\hat{O}) = \\
&\frac{1}{2}(\mathbf{P}^L(\hat{O}))^T\mathbf{E}(\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T})\mathbf{E}\mathbf{P}^R(\hat{O})
\end{aligned} \tag{S19}$$

Here, (S14) and (S15) were used in the second equality. The only thing left to show here is that $\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{P}^R(\hat{O}) = \mathbf{E}\mathbf{P}^R(\hat{O})$ and $(\mathbf{P}^L(\hat{O}))^T\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T} = (\mathbf{P}^L(\hat{O}))^T\mathbf{E}$. This is the case for the full space, i.e. $i = N$, since :

$$\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{P}^R(\hat{O}) = \sum_j^N \mathbf{Q}_j\tilde{\mathbf{Q}}_j^T\mathbf{E}\mathbf{P}^R(\hat{O}) = \mathbf{I}\mathbf{E}\mathbf{P}^R(\hat{O}) = \mathbf{E}\mathbf{P}^R(\hat{O}) \tag{S20}$$

$$\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{P}^R(\hat{O}) = \sum_j^N \mathbf{Q}_j\tilde{\mathbf{Q}}_j^T\mathbf{E}\mathbf{P}^R(\hat{O}) = (\mathbf{Q}_1\tilde{\mathbf{Q}}_1^T + \mathbf{Q}_2\tilde{\mathbf{Q}}_2^T)\mathbf{E}\mathbf{P}^R(\hat{O}) \tag{S21}$$

The last equality in (S21) holds since $\text{span}\{\mathbf{Q}_1, \mathbf{Q}_2\} = \text{span}\{\mathbf{P}^R(\hat{O}), \mathbf{E}\mathbf{P}^R(\hat{O})\}$ as Lanczos algorithms build Krylov subspaces. This in turn means, that as long as $i \geq 2$, (S20) and (S21) are equal to each other (which is effectively true after the first Lanczos iteration when initial vectors are gradient property vectors or their linear combination):

$$\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{P}^R(\hat{O}) \stackrel{i \geq 1}{=} \mathbf{E}\mathbf{P}^R(\hat{O}) \tag{S22}$$

$(\mathbf{P}^L(\hat{O}))^T\mathbf{E}$ product can in the same way be proved to be left unchanged by the Lanczos basis, i.e. $(\mathbf{P}^L(\hat{O}))^T\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T} = (\mathbf{P}^L(\hat{O}))^T\mathbf{E}$. Inserting Eq. (S22) into Eq. (S19) yields:

$$\begin{aligned}
M(3) &= \frac{1}{2}(\mathbf{P}^L(\hat{O}))^T\mathbf{E}(\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T}\mathbf{E}\mathbf{Q}^{(i)}\tilde{\mathbf{Q}}^{(i)T})\mathbf{E}\mathbf{P}^R(\hat{O}) = \\
&\frac{1}{2}(\mathbf{P}^L(\hat{O}))^T\mathbf{E}^3\mathbf{P}^R(\hat{O})
\end{aligned} \tag{S23}$$

This then can be further generalized to all odd powers of m . Thus it has been shown that as long as a nonsymmetric or nonsymmetric block Lanczos is initiated with vectors that contain the gradient property vectors, the odd- m -power energy-weighted sum-over-states properties can be recovered exactly by the algorithm after only $\frac{m+1}{2}$ Lanczos vectors have been computed. Here, it is noted that in the notation in quantum chemistry the first power of excitation energy is usually absorbed in the oscillator strengths (see Eq. (3) in the main article), which in turn is contained in the sum rules expressions (see, e.g.,¹). Therefore, in quantum chemistry, the odd- m -power rule becomes an even- m -power rule.

S2 Reordering of the $\bar{\mathbf{T}}^{(k)}$ Matrix

If $k = N$, Lanczos block-tridiagonal matrix $\mathbf{T}^{(N)}$ can be reordered using the following scheme (equally valid for all k):

$$\begin{aligned}
 \mathbf{T}^{(N)} = & \begin{bmatrix} \overline{1} & \overline{N+1} & \overline{2} & \overline{N+2} & & & & \\ e_1 & d_1 & a_1 & b_1 & \cdots & & 0 & \\ -d_1 & -e_1 & -b_1 & -a_1 & & & \vdots & \\ a_1 & b_1 & e_2 & d_2 & \ddots & & & \\ -b_1 & -a_1 & -d_2 & -e_2 & & \ddots & & \\ & & \ddots & & \ddots & \ddots & & \\ & & & \ddots & & \ddots & \ddots & \\ \vdots & & & & \ddots & & \ddots & \\ 0 & \cdots & & & \ddots & & \ddots & \end{bmatrix} \begin{matrix} | 1 \\ | N+1 \\ | 2 \\ | N+2 \\ \\ \\ \\ \end{matrix} \iff \quad (\text{S24}) \\
 \bar{\mathbf{T}}^{(N)} = & \begin{bmatrix} \overline{1} & \overline{2} & & & \overline{N+1} & \overline{N+2} & & \\ e_1 & a_1 & \cdots & 0 & d_1 & b_1 & \cdots & 0 \\ a_1 & e_2 & & \vdots & b_2 & d_2 & & \vdots \\ \vdots & & \ddots & & \vdots & & \ddots & \\ 0 & \cdots & & \ddots & 0 & \cdots & & \ddots \\ -d_1 & -b_1 & \cdots & 0 & -e_1 & -a_1 & \cdots & 0 \\ -b_1 & -d_2 & & \vdots & -a_1 & -e_2 & & \vdots \\ \vdots & & \ddots & & \vdots & & \ddots & \\ 0 & \cdots & & \ddots & 0 & \cdots & & \ddots \end{bmatrix} \begin{matrix} | 1 \\ | 2 \\ \\ | N+1 \\ | N+2 \\ \\ \end{matrix} = \begin{bmatrix} \mathbf{A}'^{(N)} & \mathbf{B}'^{(N)} \\ -\mathbf{B}'^{(N)} & -\mathbf{A}'^{(N)} \end{bmatrix}
 \end{aligned}$$

S3 Supplementary Figures

S3.1 $I^{xx}(0)$ Convergence Analysis: All Molecules

Figure S1: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVDZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S2: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVTZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S3: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVQZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S4: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with aug-cc-pCV5Z basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S5: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with cc-pCVDZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S6: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with cc-pCVTZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S7: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with cc-pCVQZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S8: Normal distribution of the convergence of $I^{xx}(0)$ computed for 18 different molecules/atoms with cc-pCV5Z basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{xx}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

S3.2 $I^{yy}(0)$ Convergence Analysis: All Molecules

Figure S9: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVDZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S10: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVTZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S11: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVQZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S12: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with aug-cc-pCV5Z basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S13: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with cc-pCVDZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S14: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with cc-pCVTZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S15: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with cc-pCVQZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S16: Normal distribution of the convergence of $I^{yy}(0)$ computed for 18 different molecules/atoms with cc-pCV5Z basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{yy}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

S3.3 $I^{zz}(0)$ Convergence Analysis: All Molecules

Figure S17: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVDZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S18: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVTZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S19: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with aug-cc-pCVQZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S20: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with aug-cc-pCV5Z basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S21: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with cc-pCVDZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S22: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with cc-pCVTZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S23: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with cc-pCVQZ basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

Figure S24: Normal distribution of the convergence of $I^{zz}(0)$ computed for 18 different molecules/atoms with cc-pCV5Z basis set, using our block Lanczos RPA solver. The value compared to, $I_{diag}^{zz}(0)$, is computed after diagonalizing the full space electronic Hessian and the convergence threshold set to 0.5% of said value.

S3.4 $I^{xx}(0)$ and $I^{zz}(0)$ Convergence Analysis: BH and PH₃

Figure S25: BH. Convergence of $I^{xx}(0)$ and $I^{zz}(0)$ computed with different basis, using the block Lanczos RPA solver in Dalton. The $I^{yy}(0)$ results are identical to those of $I^{xx}(0)$, since x - and y -components transform as the same irreducible representation of the full point group, and are therefore not depicted here. The values compared to, $I_{2N}^{xx}(0)$, $I_{2N}^{yy}(0)$ and $I_{2N}^{zz}(0)$, are computed after diagonalizing the full space electronic Hessian ($2N = 378$, 378 and 570 for aug-cc-pCV5Z basis and x -, y - and z -components, respectively). Note that 1 Lanczos iteration implicitly generates 2 new Lanczos vectors.

Figure S26: PH_3 . Convergence of $I^{xx}(0)$ and $I^{zz}(0)$ computed with different basis, using the block Lanczos RPA solver in Dalton. The $I^{yy}(0)$ results are identical to those of $I^{xx}(0)$, since x - and y -components transform as the same irreducible representation of the full point group, and are therefore not depicted here. The values compared to, $I_{2N}^{xx}(0)$, $I_{2N}^{yy}(0)$ and $I_{2N}^{zz}(0)$, are computed after diagonalizing the full space electronic Hessian ($2N = 3722$, 4324 and 4324 for aug-cc-pCV5Z basis and x -, y - and z -components, respectively). Note that 1 Lanczos iteration implicitly creates 2 new Lanczos vectors.

S3.5 Convergence Analysis: Cumulative $I(0)$

S3.5.1 BH

Figure S27: Individual contributions the full space $I^{xx}(0)$ value for BH molecule, computed with the aug-cc-pCV5Z basis set. The cumulative $I^{xx}(0)$ is depicted as a dark red line.

Figure S28: Individual contributions the full space $I^{yy}(0)$ value for BH molecule, computed with the aug-cc-pCV5Z basis set. The cumulative $I^{yy}(0)$ is depicted as a dark red line.

Figure S29: Individual contributions the full space $I^{zz}(0)$ value for BH molecule, computed with the aug-cc-pCV5Z basis set. The cumulative $I^{zz}(0)$ is depicted as a dark red line.

S3.5.2 BH₃

Figure S30: Individual contributions the full space $I^{xx}(0)$ value for BH₃ molecule, computed with the aug-cc-pCV5Z basis set. The cumulative $I^{xx}(0)$ is depicted as a dark red line.

Figure S31: Individual contributions the full space $I^{yy}(0)$ value for BH_3 molecule, computed with the aug-cc-pCV5Z basis set. The cumulative $I^{yy}(0)$ is depicted as a dark red line.

Figure S32: Individual contributions the full space $I^{zz}(0)$ value for BH_3 molecule, computed with the aug-cc-pCV5Z basis set. The cumulative $I^{zz}(0)$ is depicted as a dark red line.

S4 Supplementary Tables

S4.1 Symmetry Information

Table S1: Point groups for the 18 molecules used to test the performance of the implicit block Lanczos subroutine in Dalton.

Molecule	Linear	Symmetry group	Represented in Dalton as
AlH	✓	$C_{\infty v}$	C_{2v}
BF	✓	$C_{\infty v}$	C_{2v}
BH	✓	$C_{\infty v}$	C_{2v}
CO	✓	$C_{\infty v}$	C_{2v}
HF	✓	$C_{\infty v}$	C_{2v}
HCl	✓	$C_{\infty v}$	C_{2v}
HCN	✓	$C_{\infty v}$	C_{2v}
LiH	✓	$C_{\infty v}$	C_{2v}
NaH	✓	$C_{\infty v}$	C_{2v}
N ₂ O	✓	$C_{\infty v}$	C_{2v}
C ₂ H ₂	✓	$D_{\infty h}$	D_{2h}
CO ₂	✓	$D_{\infty h}$	D_{2h}
F ₂	✓	$D_{\infty h}$	D_{2h}
N ₂	✓	$D_{\infty h}$	D_{2h}
BH ₃	✗	D_{3h}	D_{2h}
PH ₃	✗	C_{3v}	C_s
H ₂ CO	✗	C_{2v}	C_{2v}
H ₂ O	✗	C_{2v}	C_{2v}

S4.2 Componentwise Convergence of Lanczos $I(0)$

Table S2: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for AlH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	126.63 eV	86	10 (11.63 %)	40 (46.51 %)
cc-pCVTZ	109.93 eV	228	70 (30.70 %)	110 (48.25 %)
cc-pCVQZ	116.63 eV	456	100 (21.93 %)	160 (35.09 %)
cc-pCV5Z	120.25 eV	794	130 (16.37 %)	280 (35.26 %)
aug-cc-pCVDZ	103.15 eV	130	40 (30.77 %)	60 (46.15 %)
aug-cc-pCVTZ	110.12 eV	314	60 (19.11 %)	120 (38.22 %)
aug-cc-pCVQZ	116.73 eV	596	90 (15.10 %)	160 (26.85 %)
aug-cc-pCV5Z	120.50 eV	1006	120 (11.93 %)	250 (24.85 %)

Table S3: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for AlH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	126.63 eV	86	10 (11.63 %)	40 (46.51 %)
cc-pCVTZ	109.93 eV	228	70 (30.70 %)	110 (48.25 %)
cc-pCVQZ	116.63 eV	456	100 (21.93 %)	160 (35.09 %)
cc-pCV5Z	120.25 eV	794	130 (16.37 %)	280 (35.26 %)
aug-cc-pCVDZ	103.15 eV	130	40 (30.77 %)	60 (46.15 %)
aug-cc-pCVTZ	110.12 eV	314	60 (19.11 %)	120 (38.22 %)
aug-cc-pCVQZ	116.73 eV	596	90 (15.10 %)	160 (26.85 %)
aug-cc-pCV5Z	120.50 eV	1006	120 (11.93 %)	250 (24.85 %)

Table S4: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for AlH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	112.84 eV	134	20 (14.93 %)	20 (14.93 %)
cc-pCVTZ	103.99 eV	334	40 (11.98 %)	50 (14.97 %)
cc-pCVQZ	110.47 eV	638	60 (9.40 %)	80 (12.54 %)
cc-pCV5Z	114.10 eV	1064	100 (9.40 %)	140 (13.16 %)
aug-cc-pCVDZ	96.79 eV	206	10 (4.85 %)	20 (9.71 %)
aug-cc-pCVTZ	104.32 eV	458	40 (8.73 %)	50 (10.92 %)
aug-cc-pCVQZ	101.74 eV	818	60 (7.33 %)	100 (12.22 %)
aug-cc-pCV5Z	114.42 eV	1334	100 (7.50 %)	140 (10.49 %)

Table S5: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for BF molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	152.64 eV	100	40 (40.00 %)	60 (60.00 %)
cc-pCVTZ	99.87 eV	272	100 (36.76 %)	130 (47.79 %)
cc-pCVQZ	95.87 eV	556	200 (35.97 %)	260 (46.76 %)
cc-pCV5Z	95.07 eV	980	360 (36.73 %)	440 (44.90 %)
aug-cc-pCVDZ	124.70 eV	160	40 (25.00 %)	80 (50.00 %)
aug-cc-pCVTZ	95.81 eV	384	100 (26.04 %)	140 (36.46 %)
aug-cc-pCVQZ	94.69 eV	728	210 (28.85 %)	260 (35.71 %)
aug-cc-pCV5Z	94.60 eV	1232	370 (30.03 %)	440 (35.71 %)

Table S6: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for BF molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	152.64 eV	100	40 (40.00 %)	60 (60.00 %)
cc-pCVTZ	99.87 eV	272	100 (36.76 %)	130 (47.79 %)
cc-pCVQZ	95.87 eV	556	200 (35.97 %)	260 (46.76 %)
cc-pCV5Z	95.07 eV	980	360 (36.73 %)	440 (44.90 %)
aug-cc-pCVDZ	124.70 eV	160	40 (25.00 %)	80 (50.00 %)
aug-cc-pCVTZ	95.81 eV	384	100 (26.04 %)	140 (36.46 %)
aug-cc-pCVQZ	94.69 eV	728	210 (28.85 %)	260 (35.71 %)
aug-cc-pCV5Z	94.60 eV	1232	370 (30.03 %)	440 (35.71 %)

Table S7: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for BF molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	118.04 eV	158	40 (25.32 %)	50 (31.65 %)
cc-pCVTZ	87.85 eV	406	90 (22.17 %)	120 (29.56 %)
cc-pCVQZ	85.88 eV	786	160 (20.36 %)	220 (27.99 %)
cc-pCV5Z	85.47 eV	1326	300 (22.62 %)	350 (26.40 %)
aug-cc-pCVDZ	107.06 eV	254	40 (15.75 %)	40 (15.75 %)
aug-cc-pCVTZ	86.21 eV	558	90 (16.13 %)	120 (21.51 %)
aug-cc-pCVQZ	85.41 eV	1014	160 (15.78 %)	220 (21.70 %)
aug-cc-pCV5Z	85.30 eV	1638	300 (18.32 %)	380 (23.20 %)

Table S8: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for BH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	79.08 eV	30	30 (100.00 %)	30 (100.00 %)
cc-pCVTZ	50.81 eV	78	40 (51.28 %)	60 (76.92 %)
cc-pCVQZ	49.24 eV	162	90 (55.56 %)	110 (67.90 %)
cc-pCV5Z	49.04 eV	288	140 (48.61 %)	170 (59.03 %)
aug-cc-pCVDZ	71.15 eV	48	30 (62.50 %)	40 (83.33 %)
aug-cc-pCVTZ	49.84 eV	114	60 (52.63 %)	70 (61.40 %)
aug-cc-pCVQZ	48.80 eV	222	90 (40.54 %)	120 (54.05 %)
aug-cc-pCV5Z	48.69 eV	378	140 (37.04 %)	180 (47.62 %)

Table S9: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for BH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	79.08 eV	30	30 (100.00 %)	30 (100.00 %)
cc-pCVTZ	50.81 eV	78	40 (51.28 %)	60 (76.92 %)
cc-pCVQZ	49.24 eV	162	90 (55.56 %)	110 (67.90 %)
cc-pCV5Z	49.04 eV	288	140 (48.61 %)	170 (59.03 %)
aug-cc-pCVDZ	71.15 eV	48	30 (62.50 %)	40 (83.33 %)
aug-cc-pCVTZ	49.84 eV	114	60 (52.63 %)	70 (61.40 %)
aug-cc-pCVQZ	48.80 eV	222	90 (40.54 %)	120 (54.05 %)
aug-cc-pCV5Z	48.69 eV	378	140 (37.04 %)	180 (47.62 %)

Table S10: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for BH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	58.12 eV	54	10 (18.52 %)	10 (18.52 %)
cc-pCVTZ	42.77 eV	138	30 (21.74 %)	40 (28.99 %)
cc-pCVQZ	41.90 eV	264	20 (7.58 %)	60 (22.73 %)
cc-pCV5Z	41.78 eV	444	70 (15.77 %)	90 (20.27 %)
aug-cc-pCVDZ	53.68 eV	90	10 (11.11 %)	30 (33.33 %)
aug-cc-pCVTZ	42.49 eV	198	30 (15.15 %)	40 (20.20 %)
aug-cc-pCVQZ	41.85 eV	354	50 (14.12 %)	80 (22.60 %)
aug-cc-pCV5Z	41.75 eV	570	70 (12.28 %)	90 (15.79 %)

Table S11: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for BH_3 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	45.77 eV	94	10 (10.64 %)	10 (10.64 %)
cc-pCVTZ	38.35 eV	242	10 (4.13 %)	20 (8.26 %)
cc-pCVQZ	37.90 eV	476	20 (4.20 %)	20 (4.20 %)
cc-pCV5Z	37.86 eV	818	30 (3.67 %)	30 (3.67 %)
aug-cc-pCVDZ	43.58 eV	160	10 (6.25 %)	10 (6.25 %)
aug-cc-pCVTZ	38.15 eV	362	10 (2.76 %)	20 (5.52 %)
aug-cc-pCVQZ	37.86 eV	666	20 (3.00 %)	20 (3.00 %)
aug-cc-pCV5Z	37.82 eV	1094	20 (1.83 %)	30 (2.74 %)

Table S12: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for BH_3 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	45.77 eV	74	10 (13.51 %)	10 (13.51 %)
cc-pCVTZ	38.35 eV	198	10 (5.05 %)	20 (10.10 %)
cc-pCVQZ	37.90 eV	404	20 (4.95 %)	20 (4.95 %)
cc-pCV5Z	37.86 eV	710	30 (4.23 %)	30 (4.23 %)
aug-cc-pCVDZ	43.58 eV	128	10 (7.81 %)	10 (7.81 %)
aug-cc-pCVTZ	38.15 eV	302	10 (3.31 %)	20 (6.62 %)
aug-cc-pCVQZ	37.86 eV	574	20 (3.48 %)	20 (3.48 %)
aug-cc-pCV5Z	37.82 eV	962	20 (2.08 %)	30 (3.12 %)

Table S13: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for BH_3 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	63.07 eV	40	10 (25.00 %)	10 (25.00 %)
cc-pCVTZ	45.22 eV	120	20 (16.67 %)	30 (25.00 %)
cc-pCVQZ	44.26 eV	268	10 (3.73 %)	10 (3.73 %)
cc-pCV5Z	44.10 eV	502	20 (3.98 %)	20 (3.98 %)
aug-cc-pCVDZ	56.51 eV	68	10 (14.71 %)	10 (14.71 %)
aug-cc-pCVTZ	44.65 eV	186	20 (10.75 %)	20 (10.75 %)
aug-cc-pCVQZ	44.05 eV	388	20 (5.15 %)	20 (5.15 %)
aug-cc-pCV5Z	43.99 eV	692	20 (2.89 %)	40 (5.78 %)

Table S14: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for C_2H_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	89.77 eV	64	10 (15.62 %)	20 (31.25 %)
cc-pCVTZ	65.81 eV	180	20 (11.11 %)	50 (27.78 %)
cc-pCVQZ	63.97 eV	378	20 (5.29 %)	30 (7.94 %)
cc-pCV5Z	63.93 eV	676	30 (4.44 %)	50 (7.40 %)
aug-cc-pCVDZ	81.52 eV	108	10 (9.26 %)	10 (9.26 %)
aug-cc-pCVTZ	64.26 eV	266	20 (7.52 %)	40 (15.04 %)
aug-cc-pCVQZ	63.43 eV	520	30 (5.77 %)	80 (15.38 %)
aug-cc-pCV5Z	63.36 eV	888	50 (5.63 %)	120 (13.51 %)

Table S15: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for C_2H_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	89.77 eV	64	10 (15.62 %)	20 (31.25 %)
cc-pCVTZ	65.81 eV	180	20 (11.11 %)	50 (27.78 %)
cc-pCVQZ	63.97 eV	378	20 (5.29 %)	30 (7.94 %)
cc-pCV5Z	63.93 eV	676	30 (4.44 %)	50 (7.40 %)
aug-cc-pCVDZ	81.52 eV	108	10 (9.26 %)	10 (9.26 %)
aug-cc-pCVTZ	64.26 eV	266	20 (7.52 %)	40 (15.04 %)
aug-cc-pCVQZ	63.43 eV	520	30 (5.77 %)	80 (15.38 %)
aug-cc-pCV5Z	63.36 eV	888	50 (5.63 %)	120 (13.51 %)

Table S16: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for C_2H_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	56.98 eV	116	30 (25.86 %)	50 (43.10 %)
cc-pCVTZ	49.38 eV	288	70 (24.31 %)	80 (27.78 %)
cc-pCVQZ	48.57 eV	554	120 (21.66 %)	140 (25.27 %)
cc-pCV5Z	48.46 eV	938	190 (20.26 %)	260 (27.72 %)
aug-cc-pCVDZ	55.56 eV	188	30 (15.96 %)	50 (26.60 %)
aug-cc-pCVTZ	48.97 eV	412	70 (16.99 %)	80 (19.42 %)
aug-cc-pCVQZ	48.48 eV	744	120 (16.13 %)	140 (18.82 %)
aug-cc-pCV5Z	48.44 eV	1196	190 (15.89 %)	270 (22.58 %)

Table S17: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for CO molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	137.71 eV	100	40 (40.00 %)	40 (40.00 %)
cc-pCVTZ	94.50 eV	272	70 (25.74 %)	110 (40.44 %)
cc-pCVQZ	91.50 eV	556	140 (25.18 %)	190 (34.17 %)
cc-pCV5Z	91.19 eV	980	270 (27.55 %)	340 (34.69 %)
aug-cc-pCVDZ	120.32 eV	160	30 (18.75 %)	40 (25.00 %)
aug-cc-pCVTZ	91.95 eV	384	60 (15.62 %)	100 (26.04 %)
aug-cc-pCVQZ	90.77 eV	728	130 (17.86 %)	200 (27.47 %)
aug-cc-pCV5Z	90.69 eV	1232	240 (19.48 %)	330 (26.79 %)

Table S18: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for CO molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	137.71 eV	100	40 (40.00 %)	40 (40.00 %)
cc-pCVTZ	94.50 eV	272	70 (25.74 %)	110 (40.44 %)
cc-pCVQZ	91.50 eV	556	140 (25.18 %)	190 (34.17 %)
cc-pCV5Z	91.19 eV	980	270 (27.55 %)	340 (34.69 %)
aug-cc-pCVDZ	120.32 eV	160	30 (18.75 %)	40 (25.00 %)
aug-cc-pCVTZ	91.95 eV	384	60 (15.62 %)	100 (26.04 %)
aug-cc-pCVQZ	90.77 eV	728	130 (17.86 %)	200 (27.47 %)
aug-cc-pCV5Z	90.69 eV	1232	240 (19.48 %)	340 (27.60 %)

Table S19: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for CO molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	100.64 eV	158	40 (25.32 %)	50 (31.65 %)
cc-pCVTZ	78.83 eV	406	70 (17.24 %)	70 (17.24 %)
cc-pCVQZ	76.94 eV	786	130 (16.54 %)	150 (19.08 %)
cc-pCV5Z	76.68 eV	1326	210 (15.84 %)	260 (19.61 %)
aug-cc-pCVDZ	94.03 eV	254	20 (7.87 %)	40 (15.75 %)
aug-cc-pCVTZ	77.43 eV	558	70 (12.54 %)	70 (12.54 %)
aug-cc-pCVQZ	76.72 eV	1014	130 (12.82 %)	150 (14.79 %)
aug-cc-pCV5Z	76.64 eV	1638	220 (13.43 %)	250 (15.26 %)

Table S20: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for CO₂ molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	149.94 eV	118	10 (8.47 %)	10 (8.47 %)
cc-pCVTZ	102.55 eV	324	50 (15.43 %)	70 (21.60 %)
cc-pCVQZ	99.08 eV	662	50 (7.55 %)	150 (22.66 %)
cc-pCV5Z	98.60 eV	1166	70 (6.00 %)	90 (7.72 %)
aug-cc-pCVDZ	125.77 eV	190	10 (5.26 %)	40 (21.05 %)
aug-cc-pCVTZ	98.81 eV	458	20 (4.37 %)	50 (10.92 %)
aug-cc-pCVQZ	97.67 eV	866	50 (5.77 %)	120 (13.86 %)
aug-cc-pCV5Z	97.60 eV	1466	80 (5.46 %)	140 (9.55 %)

Table S21: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for CO₂ molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	149.94 eV	118	10 (8.47 %)	10 (8.47 %)
cc-pCVTZ	102.55 eV	324	50 (15.43 %)	70 (21.60 %)
cc-pCVQZ	99.08 eV	662	50 (7.55 %)	150 (22.66 %)
cc-pCV5Z	98.60 eV	1166	70 (6.00 %)	90 (7.72 %)
aug-cc-pCVDZ	125.77 eV	190	10 (5.26 %)	40 (21.05 %)
aug-cc-pCVTZ	98.81 eV	458	20 (4.37 %)	50 (10.92 %)
aug-cc-pCVQZ	97.67 eV	866	50 (5.77 %)	120 (13.86 %)
aug-cc-pCV5Z	97.60 eV	1466	80 (5.46 %)	140 (9.55 %)

Table S22: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for CO₂ molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	96.63 eV	178	60 (33.71 %)	60 (33.71 %)
cc-pCVTZ	78.49 eV	458	80 (17.47 %)	110 (24.02 %)
cc-pCVQZ	76.78 eV	890	170 (19.10 %)	210 (23.60 %)
cc-pCV5Z	76.47 eV	1508	300 (19.89 %)	390 (25.86 %)
aug-cc-pCVDZ	91.82 eV	284	40 (14.08 %)	50 (17.61 %)
aug-cc-pCVTZ	77.12 eV	632	80 (12.66 %)	100 (15.82 %)
aug-cc-pCVQZ	76.50 eV	1148	170 (14.81 %)	220 (19.16 %)
aug-cc-pCV5Z	76.42 eV	1868	300 (16.06 %)	380 (20.34 %)

Table S23: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for F_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	207.94 eV	60	10 (16.67 %)	10 (16.67 %)
cc-pCVTZ	135.98 eV	172	20 (11.63 %)	20 (11.63 %)
cc-pCVQZ	127.85 eV	356	20 (5.62 %)	30 (8.43 %)
cc-pCV5Z	125.19 eV	630	30 (4.76 %)	30 (4.76 %)
aug-cc-pCVDZ	152.43 eV	100	10 (10.00 %)	20 (20.00 %)
aug-cc-pCVTZ	120.39 eV	244	20 (8.20 %)	20 (8.20 %)
aug-cc-pCVQZ	119.75 eV	468	50 (10.68 %)	70 (14.96 %)
aug-cc-pCV5Z	119.82 eV	792	70 (8.84 %)	120 (15.15 %)

Table S24: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for F_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	207.94 eV	60	10 (16.67 %)	10 (16.67 %)
cc-pCVTZ	135.98 eV	172	20 (11.63 %)	20 (11.63 %)
cc-pCVQZ	127.85 eV	356	20 (5.62 %)	30 (8.43 %)
cc-pCV5Z	125.19 eV	630	30 (4.76 %)	30 (4.76 %)
aug-cc-pCVDZ	152.43 eV	100	10 (10.00 %)	20 (20.00 %)
aug-cc-pCVTZ	120.39 eV	244	20 (8.20 %)	20 (8.20 %)
aug-cc-pCVQZ	119.75 eV	468	50 (10.68 %)	70 (14.96 %)
aug-cc-pCV5Z	119.82 eV	792	70 (8.84 %)	120 (15.15 %)

Table S25: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for F_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	163.89 eV	90	40 (44.44 %)	50 (55.56 %)
cc-pCVTZ	109.03 eV	238	80 (33.61 %)	90 (37.82 %)
cc-pCVQZ	105.32 eV	468	160 (34.19 %)	190 (40.60 %)
cc-pCV5Z	104.27 eV	798	290 (36.34 %)	320 (40.10 %)
aug-cc-pCVDZ	132.44 eV	146	40 (27.40 %)	40 (27.40 %)
aug-cc-pCVTZ	103.22 eV	330	90 (27.27 %)	100 (30.30 %)
aug-cc-pCVQZ	102.32 eV	606	150 (24.75 %)	180 (29.70 %)
aug-cc-pCV5Z	102.20 eV	990	280 (28.28 %)	320 (32.32 %)

Table S26: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for H₂CO molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	118.66 eV	126	10 (7.94 %)	40 (31.75 %)
cc-pCVTZ	83.01 eV	364	50 (13.74 %)	60 (16.48 %)
cc-pCVQZ	80.29 eV	776	40 (5.15 %)	130 (16.75 %)
cc-pCV5Z	79.36 eV	1410	70 (4.96 %)	200 (14.18 %)
aug-cc-pCVDZ	100.92 eV	210	10 (4.76 %)	30 (14.29 %)
aug-cc-pCVTZ	79.71 eV	538	20 (3.72 %)	60 (11.15 %)
aug-cc-pCVQZ	78.81 eV	1068	40 (3.75 %)	120 (11.24 %)
aug-cc-pCV5Z	78.76 eV	1856	60 (3.23 %)	200 (10.78 %)

Table S27: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for H₂CO molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	102.87 eV	174	20 (11.49 %)	40 (22.99 %)
cc-pCVTZ	75.91 eV	476	20 (4.20 %)	70 (14.71 %)
cc-pCVQZ	73.12 eV	972	50 (5.14 %)	150 (15.43 %)
cc-pCV5Z	72.71 eV	1710	70 (4.09 %)	180 (10.53 %)
aug-cc-pCVDZ	88.32 eV	294	10 (3.40 %)	40 (13.61 %)
aug-cc-pCVTZ	72.87 eV	702	20 (2.85 %)	60 (8.55 %)
aug-cc-pCVQZ	72.14 eV	1332	40 (3.00 %)	100 (7.51 %)
aug-cc-pCV5Z	72.08 eV	2240	70 (3.12 %)	190 (8.48 %)

Table S28: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for H₂CO molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	82.58 eV	226	50 (22.12 %)	60 (26.55 %)
cc-pCVTZ	66.23 eV	588	80 (13.61 %)	160 (27.21 %)
cc-pCVQZ	64.78 eV	1160	160 (13.79 %)	230 (19.83 %)
cc-pCV5Z	64.55 eV	1990	250 (12.56 %)	420 (21.11 %)
aug-cc-pCVDZ	76.67 eV	374	40 (10.70 %)	50 (13.37 %)
aug-cc-pCVTZ	65.10 eV	846	70 (8.27 %)	110 (13.00 %)
aug-cc-pCVQZ	64.54 eV	1564	150 (9.59 %)	230 (14.71 %)
aug-cc-pCV5Z	64.48 eV	2568	250 (9.74 %)	410 (15.97 %)

Table S29: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for H₂O molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	122.12 eV	48	10 (20.83 %)	20 (41.67 %)
cc-pCVTZ	85.59 eV	146	20 (13.70 %)	40 (27.40 %)
cc-pCVQZ	81.40 eV	316	30 (9.49 %)	80 (25.32 %)
cc-pCV5Z	79.03 eV	578	40 (6.92 %)	130 (22.49 %)
aug-cc-pCVDZ	95.55 eV	84	20 (23.81 %)	30 (35.71 %)
aug-cc-pCVTZ	77.61 eV	222	20 (9.01 %)	30 (13.51 %)
aug-cc-pCVQZ	76.98 eV	446	50 (11.21 %)	80 (17.94 %)
aug-cc-pCV5Z	76.96 eV	778	80 (10.28 %)	160 (20.57 %)

Table S30: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for H₂O molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	93.22 eV	66	20 (30.30 %)	30 (45.45 %)
cc-pCVTZ	71.89 eV	182	20 (10.99 %)	30 (16.48 %)
cc-pCVQZ	70.25 eV	376	30 (7.98 %)	30 (7.98 %)
cc-pCV5Z	69.87 eV	668	40 (5.99 %)	50 (7.49 %)
aug-cc-pCVDZ	82.52 eV	114	10 (8.77 %)	10 (8.77 %)
aug-cc-pCVTZ	69.96 eV	276	20 (7.25 %)	20 (7.25 %)
aug-cc-pCVQZ	69.43 eV	530	30 (5.66 %)	30 (5.66 %)
aug-cc-pCV5Z	69.41 eV	898	40 (4.45 %)	60 (6.68 %)

Table S31: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for H₂O molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	104.11 eV	82	30 (36.59 %)	30 (36.59 %)
cc-pCVTZ	76.54 eV	220	40 (18.18 %)	50 (22.73 %)
cc-pCVQZ	74.16 eV	442	40 (9.05 %)	100 (22.62 %)
cc-pCV5Z	73.58 eV	768	60 (7.81 %)	160 (20.83 %)
aug-cc-pCVDZ	88.10 eV	140	20 (14.29 %)	30 (21.43 %)
aug-cc-pCVTZ	73.35 eV	326	20 (6.13 %)	50 (15.34 %)
aug-cc-pCVQZ	72.78 eV	612	40 (6.54 %)	100 (16.34 %)
aug-cc-pCV5Z	72.74 eV	1016	70 (6.89 %)	160 (15.75 %)

Table S32: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for HCN molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	109.73 eV	116	10 (8.62 %)	40 (34.48 %)
cc-pCVTZ	78.08 eV	318	20 (6.29 %)	70 (22.01 %)
cc-pCVQZ	75.82 eV	658	40 (6.08 %)	130 (19.76 %)
cc-pCV5Z	75.67 eV	1168	60 (5.14 %)	110 (9.42 %)
aug-cc-pCVDZ	96.95 eV	190	10 (5.26 %)	10 (5.26 %)
aug-cc-pCVTZ	76.29 eV	460	20 (4.35 %)	60 (13.04 %)
aug-cc-pCVQZ	75.28 eV	886	40 (4.51 %)	130 (14.67 %)
aug-cc-pCV5Z	75.20 eV	1506	60 (3.98 %)	200 (13.28 %)

Table S33: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for HCN molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	109.73 eV	116	10 (8.62 %)	40 (34.48 %)
cc-pCVTZ	78.08 eV	318	20 (6.29 %)	70 (22.01 %)
cc-pCVQZ	75.82 eV	658	40 (6.08 %)	130 (19.76 %)
cc-pCV5Z	75.67 eV	1168	60 (5.14 %)	110 (9.42 %)
aug-cc-pCVDZ	96.95 eV	190	10 (5.26 %)	10 (5.26 %)
aug-cc-pCVTZ	76.29 eV	460	20 (4.35 %)	60 (13.04 %)
aug-cc-pCVQZ	75.28 eV	886	40 (4.51 %)	130 (14.67 %)
aug-cc-pCV5Z	75.20 eV	1506	60 (3.98 %)	200 (13.28 %)

Table S34: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for HCN molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	72.02 eV	192	40 (20.83 %)	60 (31.25 %)
cc-pCVTZ	60.87 eV	488	70 (14.34 %)	110 (22.54 %)
cc-pCVQZ	59.81 eV	944	160 (16.95 %)	180 (19.07 %)
cc-pCV5Z	59.70 eV	1598	240 (15.02 %)	300 (18.77 %)
aug-cc-pCVDZ	70.63 eV	312	40 (12.82 %)	40 (12.82 %)
aug-cc-pCVTZ	60.39 eV	688	70 (10.17 %)	100 (14.53 %)
aug-cc-pCVQZ	59.76 eV	1248	150 (12.02 %)	180 (14.42 %)
aug-cc-pCV5Z	59.70 eV	2024	230 (11.36 %)	300 (14.82 %)

Table S35: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for HCl molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	184.16 eV	102	10 (9.80 %)	10 (9.80 %)
cc-pCVTZ	144.66 eV	286	50 (17.48 %)	60 (20.98 %)
cc-pCVQZ	158.12 eV	582	40 (6.87 %)	60 (10.31 %)
cc-pCV5Z	159.24 eV	1018	80 (7.86 %)	110 (10.81 %)
aug-cc-pCVDZ	160.01 eV	160	10 (6.25 %)	10 (6.25 %)
aug-cc-pCVTZ	144.51 eV	398	30 (7.54 %)	40 (10.05 %)
aug-cc-pCVQZ	154.77 eV	766	40 (5.22 %)	50 (6.53 %)
aug-cc-pCV5Z	157.68 eV	1292	60 (4.64 %)	100 (7.74 %)

Table S36: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for HCl molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	184.16 eV	102	10 (9.80 %)	10 (9.80 %)
cc-pCVTZ	144.66 eV	286	50 (17.48 %)	60 (20.98 %)
cc-pCVQZ	158.12 eV	582	40 (6.87 %)	60 (10.31 %)
cc-pCV5Z	159.24 eV	1018	80 (7.86 %)	110 (10.81 %)
aug-cc-pCVDZ	160.01 eV	160	10 (6.25 %)	10 (6.25 %)
aug-cc-pCVTZ	144.51 eV	398	30 (7.54 %)	40 (10.05 %)
aug-cc-pCVQZ	154.77 eV	766	40 (5.22 %)	50 (6.53 %)
aug-cc-pCV5Z	157.68 eV	1292	60 (4.64 %)	100 (7.74 %)

Table S37: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for HCl molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	169.81 eV	150	20 (13.33 %)	20 (13.33 %)
cc-pCVTZ	135.91 eV	390	60 (15.38 %)	80 (20.51 %)
cc-pCVQZ	147.64 eV	758	50 (6.60 %)	90 (11.87 %)
cc-pCV5Z	150.34 eV	1280	80 (6.25 %)	120 (9.38 %)
aug-cc-pCVDZ	153.19 eV	234	20 (8.55 %)	20 (8.55 %)
aug-cc-pCVTZ	137.76 eV	538	30 (5.58 %)	40 (7.43 %)
aug-cc-pCVQZ	147.36 eV	988	50 (5.06 %)	60 (6.07 %)
aug-cc-pCV5Z	150.15 eV	1610	70 (4.35 %)	110 (6.83 %)

Table S38: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for HF molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	177.45 eV	44	10 (22.73 %)	20 (45.45 %)
cc-pCVTZ	117.69 eV	128	30 (23.44 %)	40 (31.25 %)
cc-pCVQZ	109.89 eV	270	40 (14.81 %)	80 (29.63 %)
cc-pCV5Z	106.65 eV	484	40 (8.26 %)	70 (14.46 %)
aug-cc-pCVDZ	128.03 eV	76	20 (26.32 %)	30 (39.47 %)
aug-cc-pCVTZ	103.02 eV	190	20 (10.53 %)	30 (15.79 %)
aug-cc-pCVQZ	102.36 eV	372	50 (13.44 %)	80 (21.51 %)
aug-cc-pCV5Z	102.37 eV	636	70 (11.01 %)	140 (22.01 %)

Table S39: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for HF molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	177.45 eV	44	10 (22.73 %)	20 (45.45 %)
cc-pCVTZ	117.69 eV	128	30 (23.44 %)	40 (31.25 %)
cc-pCVQZ	109.89 eV	270	40 (14.81 %)	80 (29.63 %)
cc-pCV5Z	106.65 eV	484	40 (8.26 %)	70 (14.46 %)
aug-cc-pCVDZ	128.03 eV	76	20 (26.32 %)	30 (39.47 %)
aug-cc-pCVTZ	103.02 eV	190	20 (10.53 %)	30 (15.79 %)
aug-cc-pCVQZ	102.36 eV	372	50 (13.44 %)	80 (21.51 %)
aug-cc-pCV5Z	102.37 eV	636	70 (11.01 %)	140 (22.01 %)

Table S40: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for HF molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	135.72 eV	70	30 (42.86 %)	30 (42.86 %)
cc-pCVTZ	97.47 eV	186	40 (21.51 %)	50 (26.88 %)
cc-pCVQZ	94.81 eV	368	80 (21.74 %)	100 (27.17 %)
cc-pCV5Z	94.39 eV	632	80 (12.66 %)	180 (28.48 %)
aug-cc-pCVDZ	114.00 eV	118	30 (25.42 %)	30 (25.42 %)
aug-cc-pCVTZ	94.36 eV	270	30 (11.11 %)	50 (18.52 %)
aug-cc-pCVQZ	93.70 eV	498	50 (10.04 %)	90 (18.07 %)
aug-cc-pCV5Z	93.67 eV	818	90 (11.00 %)	150 (18.34 %)

Table S41: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for N_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	131.20 eV	48	20 (41.67 %)	30 (62.50 %)
cc-pCVTZ	94.09 eV	134	40 (29.85 %)	50 (37.31 %)
cc-pCVQZ	90.52 eV	276	30 (10.87 %)	90 (32.61 %)
cc-pCV5Z	90.11 eV	488	40 (8.20 %)	120 (24.59 %)
aug-cc-pCVDZ	117.95 eV	78	10 (12.82 %)	30 (38.46 %)
aug-cc-pCVTZ	90.20 eV	190	30 (15.79 %)	50 (26.32 %)
aug-cc-pCVQZ	89.17 eV	362	50 (13.81 %)	100 (27.62 %)
aug-cc-pCV5Z	89.11 eV	614	50 (8.14 %)	140 (22.80 %)

Table S42: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for N_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	131.20 eV	48	20 (41.67 %)	30 (62.50 %)
cc-pCVTZ	94.09 eV	134	40 (29.85 %)	50 (37.31 %)
cc-pCVQZ	90.52 eV	276	30 (10.87 %)	90 (32.61 %)
cc-pCV5Z	90.11 eV	488	40 (8.20 %)	120 (24.59 %)
aug-cc-pCVDZ	117.95 eV	78	10 (12.82 %)	30 (38.46 %)
aug-cc-pCVTZ	90.20 eV	190	30 (15.79 %)	50 (26.32 %)
aug-cc-pCVQZ	89.17 eV	362	50 (13.81 %)	100 (27.62 %)
aug-cc-pCV5Z	89.11 eV	614	50 (8.14 %)	140 (22.80 %)

Table S43: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for N_2 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	92.44 eV	82	30 (36.59 %)	50 (60.98 %)
cc-pCVTZ	74.41 eV	206	50 (24.27 %)	50 (24.27 %)
cc-pCVQZ	73.46 eV	396	100 (25.25 %)	110 (27.78 %)
cc-pCV5Z	73.39 eV	666	150 (22.52 %)	170 (25.53 %)
aug-cc-pCVDZ	91.76 eV	130	30 (23.08 %)	50 (38.46 %)
aug-cc-pCVTZ	74.20 eV	282	40 (14.18 %)	60 (21.28 %)
aug-cc-pCVQZ	73.39 eV	510	100 (19.61 %)	110 (21.57 %)
aug-cc-pCV5Z	73.33 eV	822	160 (19.46 %)	180 (21.90 %)

Table S44: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for N₂O molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	144.24 eV	232	20 (8.62 %)	40 (17.24 %)
cc-pCVTZ	101.37 eV	640	60 (9.38 %)	90 (14.06 %)
cc-pCVQZ	97.37 eV	1312	50 (3.81 %)	200 (15.24 %)
cc-pCV5Z	96.64 eV	2314	80 (3.46 %)	140 (6.05 %)
aug-cc-pCVDZ	124.50 eV	376	10 (2.66 %)	40 (10.64 %)
aug-cc-pCVTZ	96.89 eV	904	20 (2.21 %)	70 (7.74 %)
aug-cc-pCVQZ	95.85 eV	1720	40 (2.33 %)	170 (9.88 %)
aug-cc-pCV5Z	95.80 eV	2908	70 (2.41 %)	270 (9.28 %)

Table S45: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for N₂O molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	144.24 eV	232	20 (8.62 %)	40 (17.24 %)
cc-pCVTZ	101.37 eV	640	60 (9.38 %)	90 (14.06 %)
cc-pCVQZ	97.37 eV	1312	50 (3.81 %)	200 (15.24 %)
cc-pCV5Z	96.64 eV	2314	80 (3.46 %)	140 (6.05 %)
aug-cc-pCVDZ	124.50 eV	376	10 (2.66 %)	40 (10.64 %)
aug-cc-pCVTZ	96.89 eV	904	20 (2.21 %)	70 (7.74 %)
aug-cc-pCVQZ	95.85 eV	1720	40 (2.33 %)	170 (9.88 %)
aug-cc-pCV5Z	95.80 eV	2908	70 (2.41 %)	280 (9.63 %)

Table S46: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for N₂O molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 %, No. Lanczos vectors, (% of full space)
cc-pCVDZ	89.13 eV	360	70 (19.44 %)	100 (27.78 %)
cc-pCVTZ	72.00 eV	924	140 (15.15 %)	200 (21.65 %)
cc-pCVQZ	71.02 eV	1794	290 (16.16 %)	360 (20.07 %)
cc-pCV5Z	70.90 eV	3036	500 (16.47 %)	650 (21.41 %)
aug-cc-pCVDZ	86.99 eV	576	60 (10.42 %)	70 (12.15 %)
aug-cc-pCVTZ	71.60 eV	1272	120 (9.43 %)	180 (14.15 %)
aug-cc-pCVQZ	70.95 eV	2316	290 (12.52 %)	360 (15.54 %)
aug-cc-pCV5Z	70.90 eV	3756	500 (13.31 %)	620 (16.51 %)

Table S47: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for Ne atom with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	257.76 eV	20	10 (50.00 %)	10 (50.00 %)
cc-pCVTZ	160.05 eV	56	20 (35.71 %)	20 (35.71 %)
cc-pCVQZ	149.74 eV	114	20 (17.54 %)	30 (26.32 %)
cc-pCV5Z	147.34 eV	198	30 (15.15 %)	30 (15.15 %)
aug-cc-pCVDZ	170.98 eV	34	10 (29.41 %)	20 (58.82 %)
aug-cc-pCVTZ	138.20 eV	78	20 (25.64 %)	20 (25.64 %)
aug-cc-pCVQZ	137.34 eV	150	30 (20.00 %)	40 (26.67 %)
aug-cc-pCV5Z	137.24 eV	246	40 (16.26 %)	70 (28.46 %)

Table S48: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for Ne atom with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	257.76 eV	20	10 (50.00 %)	10 (50.00 %)
cc-pCVTZ	160.05 eV	56	20 (35.71 %)	20 (35.71 %)
cc-pCVQZ	149.74 eV	114	20 (17.54 %)	30 (26.32 %)
cc-pCV5Z	147.34 eV	198	30 (15.15 %)	30 (15.15 %)
aug-cc-pCVDZ	170.98 eV	34	10 (29.41 %)	20 (58.82 %)
aug-cc-pCVTZ	138.20 eV	78	20 (25.64 %)	20 (25.64 %)
aug-cc-pCVQZ	137.34 eV	150	30 (20.00 %)	40 (26.67 %)
aug-cc-pCV5Z	137.24 eV	246	40 (16.26 %)	70 (28.46 %)

Table S49: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for Ne atom with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	257.76 eV	20	10 (50.00 %)	10 (50.00 %)
cc-pCVTZ	160.05 eV	56	20 (35.71 %)	20 (35.71 %)
cc-pCVQZ	149.74 eV	114	20 (17.54 %)	30 (26.32 %)
cc-pCV5Z	147.34 eV	198	30 (15.15 %)	30 (15.15 %)
aug-cc-pCVDZ	170.98 eV	34	10 (29.41 %)	20 (58.82 %)
aug-cc-pCVTZ	138.20 eV	78	20 (25.64 %)	20 (25.64 %)
aug-cc-pCVQZ	137.34 eV	150	30 (20.00 %)	40 (26.67 %)
aug-cc-pCV5Z	137.24 eV	246	40 (16.26 %)	70 (28.46 %)

Table S50: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for LiH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	52.99 eV	20	20 (100.00 %)	20 (100.00 %)
cc-pCVTZ	36.20 eV	52	20 (38.46 %)	20 (38.46 %)
cc-pCVQZ	34.44 eV	108	10 (9.26 %)	20 (18.52 %)
aug-cc-pCVDZ	48.97 eV	32	10 (31.25 %)	20 (62.50 %)
aug-cc-pCVTZ	35.43 eV	76	10 (13.16 %)	20 (26.32 %)
aug-cc-pCVQZ	34.31 eV	148	10 (6.76 %)	20 (13.51 %)

Table S51: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for LiH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	52.99 eV	20	20 (100.00 %)	20 (100.00 %)
cc-pCVTZ	36.20 eV	52	20 (38.46 %)	20 (38.46 %)
cc-pCVQZ	34.44 eV	108	10 (9.26 %)	20 (18.52 %)
aug-cc-pCVDZ	48.97 eV	32	10 (31.25 %)	20 (62.50 %)
aug-cc-pCVTZ	35.43 eV	76	10 (13.16 %)	20 (26.32 %)
aug-cc-pCVQZ	34.31 eV	148	10 (6.76 %)	20 (13.51 %)

Table S52: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for LiH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space size, $2N$	Convergence within 1.0 %, No. Lanczos vectors, (% of full space)	Convergence within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	51.43 eV	40	10 (25.00 %)	20 (50.00 %)
cc-pCVTZ	35.24 eV	96	20 (20.83 %)	40 (41.67 %)
cc-pCVQZ	34.21 eV	180	10 (5.56 %)	70 (38.89 %)
aug-cc-pCVDZ	47.87 eV	64	10 (15.62 %)	10 (15.62 %)
aug-cc-pCVTZ	34.48 eV	136	10 (7.35 %)	10 (7.35 %)
aug-cc-pCVQZ	34.11 eV	240	10 (4.17 %)	60 (25.00 %)

Table S53: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for NaH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	79.74 eV	76	30 (39.47 %)	30 (39.47 %)
cc-pCVTZ	89.85 eV	198	30 (15.15 %)	30 (15.15 %)
cc-pCVQZ	106.81 eV	394	50 (12.69 %)	60 (15.23 %)
cc-pCV5Z	112.10 eV	680	70 (10.29 %)	80 (11.76 %)
aug-cc-pCVDZ	111.06 eV	114	10 (8.77 %)	20 (17.54 %)
aug-cc-pCVTZ	89.98 eV	272	30 (11.03 %)	30 (11.03 %)
aug-cc-pCVQZ	106.84 eV	516	50 (9.69 %)	50 (9.69 %)
aug-cc-pCV5Z	112.39 eV	862	70 (8.12 %)	80 (9.28 %)

Table S54: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for NaH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	79.74 eV	76	30 (39.47 %)	30 (39.47 %)
cc-pCVTZ	89.85 eV	198	30 (15.15 %)	30 (15.15 %)
cc-pCVQZ	106.81 eV	394	50 (12.69 %)	60 (15.23 %)
cc-pCV5Z	112.10 eV	680	70 (10.29 %)	80 (11.76 %)
aug-cc-pCVDZ	111.06 eV	114	10 (8.77 %)	20 (17.54 %)
aug-cc-pCVTZ	89.98 eV	272	30 (11.03 %)	30 (11.03 %)
aug-cc-pCVQZ	106.84 eV	516	50 (9.69 %)	50 (9.69 %)
aug-cc-pCV5Z	112.39 eV	862	70 (8.12 %)	80 (9.28 %)

Table S55: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for NaH molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	72.54 eV	120	30 (25.00 %)	50 (41.67 %)
cc-pCVTZ	88.67 eV	288	40 (13.89 %)	40 (13.89 %)
cc-pCVQZ	104.52 eV	544	60 (11.03 %)	80 (14.71 %)
cc-pCV5Z	107.34 eV	888	90 (10.14 %)	140 (15.77 %)
aug-cc-pCVDZ	110.23 eV	180	10 (5.56 %)	20 (11.11 %)
aug-cc-pCVTZ	87.53 eV	392	40 (10.20 %)	40 (10.20 %)
aug-cc-pCVQZ	104.10 eV	704	70 (9.94 %)	90 (12.78 %)
aug-cc-pCV5Z	108.23 eV	1116	90 (8.06 %)	140 (12.54 %)

Table S56: The convergence of Lanczos iteration, when computing the x-component of $I(0)$ for PH_3 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{xx}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	116.68 eV	252	20 (7.94 %)	30 (11.90 %)
cc-pCVTZ	99.31 eV	728	50 (6.87 %)	80 (10.99 %)
cc-pCVQZ	107.35 eV	1540	50 (3.25 %)	90 (5.84 %)
cc-pCV5Z	109.45 eV	2778	100 (3.60 %)	150 (5.40 %)
aug-cc-pCVDZ	110.86 eV	416	10 (2.40 %)	20 (4.81 %)
aug-cc-pCVTZ	99.98 eV	1080	40 (3.70 %)	40 (3.70 %)
aug-cc-pCVQZ	107.26 eV	2152	50 (2.32 %)	80 (3.72 %)
aug-cc-pCV5Z	109.49 eV	3722	90 (2.42 %)	110 (2.96 %)

Table S57: The convergence of Lanczos iteration, when computing the y-component of $I(0)$ for PH_3 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{yy}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	116.68 eV	342	20 (5.85 %)	30 (8.77 %)
cc-pCVTZ	99.31 eV	928	50 (5.39 %)	80 (8.62 %)
cc-pCVQZ	107.35 eV	1880	60 (3.19 %)	100 (5.32 %)
cc-pCV5Z	109.45 eV	3288	120 (3.65 %)	170 (5.17 %)
aug-cc-pCVDZ	110.86 eV	556	10 (1.80 %)	20 (3.60 %)
aug-cc-pCVTZ	99.98 eV	1350	40 (2.96 %)	40 (2.96 %)
aug-cc-pCVQZ	107.26 eV	2582	50 (1.94 %)	90 (3.49 %)
aug-cc-pCV5Z	109.49 eV	4342	80 (1.84 %)	140 (3.22 %)

Table S58: The convergence of Lanczos iteration, when computing the z-component of $I(0)$ for PH_3 molecule with different basis sets. The reference value is the $I(0)$ computed after diagonalizing the full RPA electronic Hessian. Note that the algorithm used here creates 2 Lanczos vectors in one iteration.

Basis	$I_{diag}^{zz}(0)$	Full space	Convergence	Convergence
		size, $2N$	within 1.0 %, No. Lanczos vectors, (% of full space)	within 0.5 % No. Lanczos vectors, (% of full space)
cc-pCVDZ	118.09 eV	342	20 (5.85 %)	20 (5.85 %)
cc-pCVTZ	99.71 eV	928	40 (4.31 %)	60 (6.47 %)
cc-pCVQZ	108.19 eV	1880	50 (2.66 %)	60 (3.19 %)
cc-pCV5Z	110.07 eV	3288	80 (2.43 %)	130 (3.95 %)
aug-cc-pCVDZ	111.59 eV	556	20 (3.60 %)	20 (3.60 %)
aug-cc-pCVTZ	100.46 eV	1350	30 (2.22 %)	40 (2.96 %)
aug-cc-pCVQZ	107.83 eV	2582	50 (1.94 %)	60 (2.32 %)
aug-cc-pCV5Z	110.06 eV	4342	80 (1.84 %)	110 (2.53 %)

References

- (1) Sauer, S. P. A. *Molecular Electromagnetism: A Computational Chemistry Approach*; Oxford University Press, Oxford, 2011.