I. INTRODUCTION

Atomic systems offer a means to test fundamental physics at a high level of precision in the search for phenomena beyond the Standard Model of elementary particles \[1\]. This may be undertaken by examining King plots that are generated through isotope shift spectroscopy of at least two transitions in an atomic species \[2\]. Nonlinearities in such plots may arise due to higher-order effects within the Standard Model (SM), such as higher-order mass shifts \[8\], nuclear deformation \[10\], or nuclear polarizability \[12\], or due to phenomena beyond the Standard Model \[5\]. Accurate atomic structure calculations are needed to explore possible causes for such nonlinearities, as is done by investigating additional contributions to isotope shifts beyond the simple mass shift and field shift \[4\] — this can be done by analysing the residuals of a linear fit to a King plot, whereby different nonlinearities are expected to have different signatures in the residuals \[4\]. Such calculations can also be used in the extraction of information about the nuclear structure \[15\].

The recent work of Counts et al. \[4\], using narrow optical quadrupole transitions in Yb\(^{+}\), is the only King plot to date which demonstrates nonlinearity beyond the level of experimental uncertainty. This \(3\sigma\) nonlinearity is consistent with interpretations as either higher-order SM effects or physics beyond the SM. Linearity of the Ca\(^{+}\) King plot in Ref. \[20\] suggests that its interpretation as higher-order SM effects should be favoured \[17\]. Recent work by Allehabi et al. \[11\] suggests that nuclear deformation in Yb nuclei can produce a King plot nonlinearity at a level consistent with that found in \[4\]. A means of exploring the dominant cause of King plot nonlinearity is by combining prior Yb\(^{+}\) data with isotope shift measurements of the \(1S_0 - 3P_{0,1}\) transition in neutral Yb. In this work we provide estimates of these clock transition frequencies for all the bosonic isotopes of Yb \(1\), aiding the experimental search for these lines.

Advents in modern computing allow for relativistic atomic structure calculations to be performed with results consistent with experimentally determined values to a few parts in \(10^5\) \[21\]. Such computations are also used to determine mass and field shift parameters of isotope shifts for King plot analyses \[4\], \[10\], \[20\], \[25\], \[28\]. Low-lying energy levels in ytterbium have been explored through computational means \[29\], however they usually do not compute isotopes separately and often avoid the \(3P_0\) state. In this paper, the isotope shifts of the clock and intercombination line (ICL) transitions are computed \textit{ab initio} and the mass and field shift parameters that aid King plot analysis are calculated.

We describe our computational procedure in Sect. \[II\] where the atomic state function is refined through a restricted active set approach using MCDHF-CI computations from a multireference set of configuration state functions. Sect. \[III\] summarises the energy level differences and isotope shifts resulting from the MCDHF-CI computations. Sect. \[IV\] gives a detailed account of the mass and field shift parameters that are evaluated with Riss4 and the calculated atomic state functions. The second order hyperfine interaction is discussed in Sect. \[V\] which is necessary to account for the shift in centroid frequencies for the fermionic isotopes. The nuclear charge parameter is evaluated in Sect. \[VI\] followed by a King plot analysis and estimates of the clock line isotope shifts in Sect. \[VII\]. Additional information, including the predictions of the absolute clock line frequencies, is given in the Appendices.
II. COMPUTATIONAL METHOD

Most *ab initio* isotope shift computations perform computations for a single isotope and then calculate the mass and field shift parameters, using nuclear charge parameter \((\lambda^{A,A'})\) values derived from experiment to arrive at isotope shifts. In contrast, the computations presented here are similar to the “exact” method of [19] and to those of [44] and [45], in that energies and wavefunctions are computed for each isotope of interest, and the isotope shifts are taken as the differences between these energies. It is suggested that this approach can be strongly model-dependent [46], so the more common method of calculating isotope shifts via computed mass and field shift parameters is also pursued in Sections IV and VII.

A two-step approach is used to estimate the isotope shifts, mass shift and field shift parameters for the clock and intercombination transitions using computational methods. First, a MCDHF-CI approach is used to compute the atomic state functions (ASFs) for the \(^1S_0\) ground state and \(^3P_{0,1}\) excited states using the Fortran 95 package GRASP2018 (General Relativistic Atomic Structure Package) [47]. Isotope shifts are calculated as the differences in energy between the ground and excited states for different isotopes. Mass and field shift parameters are then extracted using the Fortran 90 program RIS4 (Relativistic Isotope Shift) [48]. RIS4 was written as an addition to the GRASP2K package [18]; however, we have been able to use it in conjunction with GRASP2018 [49]. The computational process is outlined in Figure 1 with further explanation below. A MCDHF-CI approach is used in favour of other approaches, e.g. configuration interaction with many-body perturbation theory (CI+MBPT) [50], all-order methods [51], and relativistic coupled cluster (RCC) calculations [52], due to the recent updates of the GRASP and RIS packages and their cross-compatibility allowing for ease of extraction of isotope shift parameters.

A restricted active space approach is used to construct the atomic state functions, whereby a multi-reference set (MR) is chosen, and additional configuration state functions (CSFs) are systematically included according to allowed substitution rules. The ground state electron configuration for ytterbium is \([Xe]4f^{14}6s^2\). The multi-reference (MR) set for the \(^1S_0\) ground state is thus taken to be \([Xe]4f^{14}(6s^2, 5d^2, 6p^2)\) as these are the configurations with two valence electrons which can form \(^1S_0\) terms and are near in energy to the \(6s^2\) ground state. This is the same MR set as that of the ‘MCDF IV’ approach used in [53]. The excited states \(^3P_{0,1}\) have electron configuration \([Xe]4f^{14}6s6p\). Conveniently, these can be computed simultaneously using the extended optimal level (EOL) mode of the RMCDHF MPI program [47]. Computing the \(^3P_{0}\) and \(^3P_{1}\) excited states together with the EOL mode is found to have negligible effect on the clock transition frequency compared with computation of the \(^3P_{0}\) state on its own (~0.3% difference). The MR set for \(^3P_{0,1}\) is taken to be \([Xe]4f^{14}(6s6p, 5d6p)\). The MR sets are summarised in Table I where \(N_{CSFs}\) is the number of configuration state functions for the MR set when using relativistic orbital labelling.

Correlation orbitals are added layer by layer, where a layer includes orbitals for each angular momentum value (e.g. \(7s, 7p\ldots, 6d\ldots, 6f\ldots, 5f\ldots, 5f\ldots\), with the subscript \(\pm\) indicating \(j = l \pm 1/2\)). Correlation layers are truncated at a principal quantum number of 12 [54]. The new correlation orbitals are optimized using the self consistent

![FIG. 1. Procedure for performing MCDHF-CI computations (GRASP2018) and extracting isotope shift information (GRASP2K). Correlation layers are added until \(n = 12\) for ground and excited states. Mass and field shift parameters are evaluated with the RIS4 package. MR - multireference; RHFS - relativistic hyperfine structure program [47].](image)

| Level \(J^H\) | MR configurations | \(N_{CSFs}\) |
|---|---|---|
| \(6s^2\) \(^1S_0\) | \([Xe]4d^{10}4f^{14}6s^25p^0\ldots\{6s^2, 5d^2, 6p^2\}\) | 5 |
| \(6s6p\) \(^3P_0\) | \([Xe]4d^{10}4f^{14}6s^25p^0\ldots\{6s6p, 5d6p\}\) | 2 |
| \(6s6p\) \(^3P_1\) | \([Xe]4d^{10}4f^{14}6s^25p^0\ldots\{6s6p, 5d6p\}\) | 5 |
field procedure [55] whilst leaving the previously computed orbitals invariant. The correlation layers are built using single and restricted double substitutions from configurations in the MR set. The closed core is taken to be [Kr]. Substitutions from the core are restricted to a single electron from either the 4f, 5s or 5p, together with unrestricted substitutions from the valence orbitals (6s, 6p, 5d). This keeps the computations tractable whilst allowing a considerable degree of valence-valence and core-valence correlation. The number of CSFs grows to 30256, 30668 and 84519 for 1S0, 3P0 and 3P1, respectively [56]. The dominant CSFs by percentage contribution to the total ASF for each state are tabulated in Appendix A.

The atomic state function computed with all the desired correlation layers is corrected for higher-order QED effects through the rclmpi program. The transverse photon interaction is reduced to the Breit interaction by scaling all transverse photon frequencies by a factor of 10−6 [47]. Vacuum polarisation effects are accounted for, and self-energy is estimated for orbitals up to n = 6. The normal and specific mass shifts due to the nuclear recoil are also included in the CI computations. Ytterbium nuclei are known to be deformed [11, 57, 58]; however, the nuclear model used for these computations — see Appendix A — does not account for nuclear deformation.

The wavefunction arising from a single CSF is an antisymmetric product of one electron wavefunctions [59] in the form of a Slater determinant [60]. The radial functions for the 6p− and 6p+ orbitals resulting from our MCDHF-CI computations for the 1S0, 3P0, and 3P1 states are represented in Fig. 2 where the large-component, P(r), and small-component, Q(r), radial functions are presented separately. Less significant deviations between the ground state and excited state radial functions were found for the 6s orbital.

Where possible, uncertainties in the presented computational results are estimated by direct comparison with experimental results [61]. In other cases, uncertainties are estimated by systematically adding correlation layers or increasing the size of the core available for correlation, and analysing the convergence of the desired properties [55]. The latter approach may not include systematic uncertainties arising from the MCDHF-CI method, and so it is desirable to compare against other computational approaches [62]. We use a combination of these methods, with quoted uncertainties corresponding to 1σ unless otherwise stated.

The efficacy of MCDHF-CI computations has been demonstrated recently; for example, Zhang et al. calculate energies for sulfur-like tungsten with near-spectroscopic accuracy [63]; Silwal et al. compute isotope shifts within the uncertainty bounds of experimental results in Mg-like and Al-like systems [44]; and Palmeri et al. produce isotope shifts in reasonable agreement with experimental results for osmium [64]. Froese Fischer and Senchuk note that good accuracy is generally achieved for light elements or highly-ionised heavy elements, but suggest neutral heavy elements with open core sub-shells can be subject to problems with the accuracy of the calculations or the energies of states not being resolved [65]. These problems are not expected to strongly influence the results of this paper, due to the simple closed-shell electron configuration of neutral ytterbium, in particular the closed 4f14 sub-shell. Further, neglecting core-core correlations here is justified: The inclusion of core-core correlation in MCDHF computations of neutral lithium and sodium, which have closed core sub-shells, was found to decrease the agreement with experiment compared to computations with only valence and core-valence correlation [66]. The agreement between computational and experimental values for oscillator strength in singly-ionised thallium, a heavier system than ytterbium with electron configuration [Xe]4f145d106s2, was also found to be better in the absence of core-core correlation [87].

The bulk of the computations were performed at the University of Western Australia High Performance Computing Centre on Kaya [68], one of their high-performance computing machines [69]. Kaya is comprised of two Dell PowerEdge R740 nodes, each with 2 Intel Xeon Gold 6254 processors with 18 cores, 768 GB of RAM and dual 1.6 TB NVMe devices. 34 cores were utilised for the computations.
III. RESULTS: ISOTOPE SHIFTS

The computed energy level differences for the clock transition are presented in Table II and in Table III for the intercombination line. Energies computed in atomic units (E₀) are converted into frequencies in Hz via multiplication by \(2\pi R_\infty = 6.579683920502(13) \times 10^{15} \text{ Hz E}_0^{-1}\) [70]. The computed energy level differences are 0.8% larger than the experimental values for the clock transition, and 0.7% larger for the intercombination line.

\[
\begin{array}{ccc}
\text{Isotope} & \text{Energy separation (MHz)} & \text{Isotope shift (MHz)} \\
168 & 522,679,368 & -5073 \\
170 & 522,677,872 & -3577 \\
171 & 522,677,352 & -3057 \\
172 & 522,676,461 & -2166 \\
173 & 522,675,963 & -1668 \\
174 & 522,675,348 & -1053 \\
176 & 522,674,295 & 0 \\
\end{array}
\]

\[
\begin{array}{ccc}
\text{Isotope} & \text{Energy separation (MHz)} & \text{Isotope shift (MHz)} \\
168 & 543,180,934 & -5127 \\
170 & 543,179,422 & -3615 \\
171 & 543,178,897 & -3090 \\
172 & 543,177,997 & -2190 \\
173 & 543,177,493 & -1686 \\
174 & 543,176,872 & -1065 \\
176 & 543,175,807 & 0 \\
\end{array}
\]

\[
\begin{array}{ccc}
\text{Isotope} & \text{Energy separation (MHz)} & \text{Isotope shift (MHz)} \\
168 & 543,180,934 & -5127 \\
170 & 543,179,422 & -3615 \\
171 & 543,178,897 & -3090 \\
172 & 543,177,997 & -2190 \\
173 & 543,177,493 & -1686 \\
174 & 543,176,872 & -1065 \\
176 & 543,175,807 & 0 \\
\end{array}
\]

For the clock transition, the isotope shift between \(^{173}\text{Yb}\) and \(^{174}\text{Yb}\) is calculated as \(-615\) MHz and between \(^{171}\text{Yb}\) and \(^{173}\text{Yb}\) is calculated as \(-1389\) MHz. Experimentally these values are found to be \(-551,536,050(11)\) MHz and \(-1259,745,595(11)\) MHz, corresponding to percentage differences of 11.5% and 10.3%, respectively. Both computed isotope shifts are larger in magnitude than the experimental values. We expect that the isotope shifts presented in Table II all have an error of approximately 11%. The computed isotope shifts for the intercombination line are presented in Table III. These values differ on average by 11.5% when compared with measured values from [71]. This difference may reduce with the inclusion of deeper core-valence correlations [27, 17, 67]. Variation in nuclear deformation between the isotopes, not accounted for in these computations, may also contribute to the differences between the experimental and computational isotope shifts. While these differences are a concern, they do not prevent us from making viable predictions for clock transition frequencies in the bosonic isotopes (discussed below). The mass and field shift factors determined from these calculations (see Section IV) lead to nuclear charge parameters consistent with previous results (see Section VI) and isotope shift estimates consistent with estimates using a method based on experimental results (see Section VII).

IV. MASS AND FIELD SHIFT PARAMETERS

The differences in nuclear mass and charge distributions between isotopes of the same element give rise to small variations in the energy eigenvalues for the atomic system, i.e., isotope shifts. By convention, the isotope shift for a pair of isotopes is calculated by subtracting the energy of the lighter isotope from that of the heavier isotope [72], so for isotopes \(A\) and \(A'\) with \(m_A > m_{A'}\), the isotope shift is given by

\[
\delta \nu^{A,A'} = \nu^A - \nu^{A'}.
\]

To a very good approximation, an isotope shift may be split into a mass shift and a field shift, arising from differences in the nuclear recoil and nuclear charge distribution, respectively, between the isotopes [72]. Under the approximation that the electronic wavefunction for a particular state is invariant between isotopes, the mass and field shifts for an atomic state \(i\) factor into electronic and nuclear components

\[
\delta \nu_i^{A,A'} = K_i \mu^{A,A'} + F_i \lambda^{A,A'},
\]

where \(K_i (F_i)\) is the electronic mass (field) shift factor,

\[
\mu^{A,A'} = \frac{1}{m_A} - \frac{1}{m_{A'}} = \frac{m_{A'} - m_A}{m_A m_{A'}}
\]

is the nuclear mass parameter, and

\[
\lambda^{A,A'} = \lambda^A - \lambda^{A'} = \sum_{n \geq 1} C_n \delta(n^{2n})^{A,A'}
\]

is the nuclear charge parameter, where \(C_n\) are Seltzer’s coefficients [22, 73, 74]. For a transition between an upper state \(j\) and a lower state \(i\), the isotope shift is given by

\[
\delta \nu_j^{A,A'} = \delta \nu_j^{A,A'} - \delta \nu_i^{A,A'} = K \mu^{A,A'} + F \lambda^{A,A'} ,
\]

where \(K = K_j - K_i\) and \(F = F_j - F_i\).

The field shift factor, \(F\), has been evaluated for each isotope for both the clock and intercombination transitions with the RIS program following GRASP2018. We present the values in Table IV where we see some
isotope-dependence. The mean values across all seven stable isotopes are $F_{\text{clock}} = -10.848(21)$ GHz fm$^{-2}$ and $F_{\text{ICL}} = -10.951(21)$ GHz fm$^{-2}$; we comment on the uncertainties below. For the clock transition, a previously reported value of $F_{\text{clock}}$ was calculated via AMBID [50], using configuration interaction only (without MBPT) and a very similar correlation model to this work [17]. For the intercombination line, the mean value is compared with previous evaluations of $F_{\text{ICL}}$ at the base of the table. Our value lies approximately central to the range of previous estimations, but with higher precision.

| Isotope | $F_{\text{clock}}$ (GHz fm$^{-2}$) | $F_{\text{ICL}}$ (GHz fm$^{-2}$) |
|---------|------------------------------------|----------------------------------|
| 168     | $-10.865(18)$                     | $-10.969(18)$                   |
| 170     | $-10.855(18)$                     | $-10.959(18)$                   |
| 171     | $-10.852(18)$                     | $-10.955(18)$                   |
| 172     | $-10.846(18)$                     | $-10.950(18)$                   |
| 173     | $-10.843(18)$                     | $-10.947(18)$                   |
| 174     | $-10.839(18)$                     | $-10.943(18)$                   |
| 176     | $-10.833(18)$                     | $-10.936(18)$                   |
| Mean    | $-10.848(21)$                     | $-10.951(21)$                   |
| Ref. 17 | $-9.7192$                         | —                               |
| Ref. 75 | —                                 | $-9.3(2.1)$                     |
| Ref. 58 | —                                 | $-10.5(1)$                      |
| Ref. 70 | —                                 | $-12.3(0.2)$                    |
| Ref. 77 | —                                 | $-12.2(0.7)$                    |

* Value is positive in reference (assumed to be absolute value)

The mass shift factors ($K$) experience little change with isotope; the mean values are $K_{\text{clock}} = -288(75)$ GHz u and $K_{\text{ICL}} = -280(72)$ GHz u. Note the values are negative. A negative specific mass shift for $^3P$ states for two-electron spectra is suggested to arise from angular correlation (private communication in [39]). Whilst these negative mass shifts appear to be at odds with the positive value of $K_{\text{ICL}} = 1.5(5)$ THz u found by [76], a review of their formulae reveals a difference in sign for the nuclear mass parameter. The same convention ($\mu_{\text{A,A}} > 0$ for $m_A > m_A$) is used in [58] [78]. The convention used in this work (equation (3)) is consistent with that of [46] [75]. Berengut et al. [17] determine $K_{\text{clock}} = -655$ GHz u using a CI+MBPT method, implemented via AMBID [50], demonstrating the dependence of the calculation on the method and supporting its approximate magnitude and sign.

Uncertainties in $K$ and $F$ for each isotope are estimated by systematically increasing the size of the computations. The convergence of the parameters as correlation layers are added for isotope $^{174}$Yb, and as the set of core orbitals available for core-valence correlation is extended for isotope $^{176}$Yb, are presented in Appendix C. The uncertainties for the mean values (over isotopes) are taken to be the sum by quadrature of (i) the standard deviation of the isotopic data and (ii) the estimated uncertainty for each isotope. The calculated mass shift factor for the intercombination line is consistent with that of [76], and the field shift factor is consistent with [58] [78].

### V. SECOND-ORDER HYPERFINE STRUCTURE

The off-diagonal second-order hyperfine interaction for isotopes with nuclear spin results in a shift of the centroid (center of gravity) of the hyperfine manifold relative to that of an isotope with no nuclear spin [41] [79]. Correcting the experimentally-determined centers of gravity for these shifts provides a means of comparison between the bosonic and fermionic isotopes (e.g. for King plot analysis). The shift for a state denoted $\gamma(J'I FM_F)$ is given by

$$\Delta E_F^{(2)} = \sum_{\gamma', J' \neq \gamma, J} \frac{\left| \langle \gamma'I FM_F | \hat{H}_{\text{hfs}} | \gamma J' FM_F \rangle \right|^2}{E_{\gamma', J'} - E_{\gamma, J}'}. \quad (6)$$

The matrix element in (6) can be written in terms of the off-diagonal hyperfine structure constants, $A(J, J')$ and $B(J, J')$, as

$$\langle \gamma' (J - 1) IF m_F | \hat{H}_{\text{hfs}} | \gamma J IF m_F \rangle = B(J, J - 1) \quad (8)$$

where $K = I + J + F$.
vanishing. The hyperfine constants calculated using the RHFS program in the GRASP2018 package [42] and are presented in Table V. Uncertainties are taken to be 4% by comparison of the diagonal hyperfine constants with the experimental values from Atkinson et al. [71].

| Isotope | \(A(3P_1)\) (GHz) | \(A(3P_1, 3P_0)\) (GHz) | \(B(3P_1)\) (GHz) |
|---------|-------------------|-------------------|-------------------|
| 171     | 4.07(17)          | 3.89(16)          | 0                 |
| 173     | -1.12(5)          | -1.07(5)          | -0.794(32)        |

Calculation of the centroid shift using Eq. 2 makes use of the energy difference between the fine-structure levels, \(3P_0^o\) and \(3P_1^o\); i.e., the value of 21 092 574.882(93) MHz for \(^{172}\)Yb, based on measurements presented in [71] and [80]. The centroid shifts for the clock transition, to second-order in perturbation theory, for the mixing of the \(3P_0\) and \(3P_1\) states we calculate to be \(-0.537(44)\) MHz and \(-0.476(39)\) MHz for \(^{171}\)Yb and \(^{173}\)Yb, respectively. For the ICL, the \(F = I\) hyperfine level is the only one influenced by mixing with the \(3P_0^o\) state, and so the shift to its centroid is smaller. The new centroids for the ICL isotope shifts relative to \(^{170}\)Yb are \(-1510.948(42)\) MHz for \(^{173}\)Yb, and \(-2781.182(54)\) MHz for \(^{171}\)Yb (c.f. Ref. [71]).

The centers of gravity for the intercombination line isotope shifts presented in [71] are correct to first-order in perturbation theory; however, the second-order corrections due to mixing with the \(3P_0^o\) state are greater than the experimental uncertainty and so are accounted for here. The effects of mixing with other nearby states \((1P_1, 3P_2)\) are estimated to be less than experimental uncertainty. The centers of gravity determined from the measured clock transition frequencies for \(^{171}\)Yb [81, 82] and \(^{173}\)Yb [83] must also take into account the higher-order perturbations in order to make comparison with that of \(^{174}\)Yb [81] in a King plot analysis. The resultant isotope shifts (between centers of gravity for the fermions) are presented in Table VI. The values are used later in Sect. VII.

### VI. NUCLEAR CHARGE PARAMETER

The nuclear charge parameter can be calculated by rearranging equation (5) to find,

\[
\lambda^{A,A'} = \frac{1}{F} \left( \delta\nu^{A,A'} - K\mu^{A,A'} \right). \tag{9}
\]

By use of the isotope shifts presented in Table VI, the mass shift and field shift parameters calculated in Table IV and the isotope masses presented in Appendix A the nuclear charge parameter \(\lambda^{A,A'}\) can be determined from Eq. 9 as presented in Table VII. The uncertainties are dominated by the uncertainty in \(K\), but they are lower than those of previous estimates by at least a factor of four. King [72] notes that the values from Clark et al. [58] give excessive weight to the muonic and x-ray data in their combined analysis, which leads to larger values than our own. Column 5 shows \(\lambda^{A,A'}\) values from Clark et al. based on optical data alone, showing better agreement with our values. Jin et al. [77] assume a specific mass shift of zero and use a larger value for the field shift parameter (12.2 GHz fm\(^{-2}\)), leading to their lower values for \(\lambda^{A,A'}\).

The nuclear charge parameter, \(\lambda^{A,A'}\) can be converted into the difference in mean-square nuclear charge radii, \(\delta(r^2)^{A,A'}\), through rescaling [75, 76] or using an iterative procedure [78, 84]. Fricke and Heilig [75] determine the higher-order moments to contribute \(-5.9\%\) to \(\lambda^{A,A'}\) based on experimental data from muonic atoms, so the differences in mean-square charge radii are recovered in this work and by [77], leading to the tabulated values being larger than those determined in this work. Allehabi et al. [11] also suggest that the tabulated \(\delta(r^2)^{A,A'}\) values are too large based on their own nuclear and electronic structure calculations.
TABLE VII. Nuclear charge parameters $\lambda_{A,A'}$ determined from the Yb $i$ intercombination line measurements and calculated $F$ parameters – in units of $10^{-3}$ fm$^2$ (column 3). Data from prior works are presented for comparison.

| $A$ | $A'$ | This work | Ref. [55] | Ref. [55] | Ref. [76] | Ref. [76] |
|-----|-----|-----------|------------|------------|------------|------------|
| 176 | 174 | 88.86(47) | 109(8)     | 87(13)     | 79.4(4.0)  | 86(2)      |
| 174 | 172 | 93.10(48) | 114(8)     | 92(15)     | 83.3(4.2)  | 90(2)      |
| 172 | 170 | 119.17(51)| 139(8)     | 116(16)    | 106.6(5.3) | 113(3)     |
| 170 | 168 | 126.86(53)| 147(8)     | 128(19)    | 113.6(5.7) | 120(14)    |
| 173 | 172 | 41.46(24) | 53(4)      | 41(10)     | 37.1(1.9)  | 40(1)      |
| 172 | 171 | 76.27(27) | 85(4)      | —          | 68.3(3.4)  | 71(1)      |
| 171 | 170 | 42.90(25) | 54(4)      | 41(10)     | 38.3(1.9)  | 42(1)      |
| 174 | 173 | 51.64(25) | 61(4)      | —          | 46.2(2.3)  | 49(1)      |
| 173 | 171 | 117.72(50)| —          | —          | —          | —          |

$a$ Combined analysis of optical, x-ray & muonic isotope shifts  
$b$ Optical isotope shifts only  
$c$ Value calculated using results from Ref. [76]

TABLE VIII. Differences in mean-square nuclear charge radii $\delta(r^2)^{A,A'}$, determined from the $\lambda_{A,A'}$ values in Table VII via $\delta(r^2)^{A,A'} = \lambda^{A,A'}/0.941$ [75], in units of $10^{-3}$ fm$^2$ (column 3). Data from other works are presented for comparison.

| $A$ | $A'$ | This work | Ref. [76] | Ref. [75] | Ref. [75] | Ref. [75] |
|-----|-----|-----------|------------|------------|------------|------------|
| 176 | 174 | 94.4(0.5)| 84.8(4.6)  | 90(2)      | 115.9(0.1)| 114(30)    | 97         |
| 174 | 172 | 98.9(0.6)| 88.8(4.6)  | 95(3)      | 120.7(0.2)| 118(28)    | 102        |
| 172 | 170 | 126.6(0.6)|113.5(6.6)| 119(4)    | 147.9(0.2)| 151(36)    | 130        |
| 170 | 168 | 154.8(0.6)|121.0(7.2)| 125(15)   | 156.1(0.4)| 160(126)   | 138        |
| 173 | 172 | 44.1(0.3)| —         | 42(2)     | 55.6(0.2)| 52(19)     | —          |
| 172 | 171 | 81.1(0.3)| —         | 75(2)     | 90.7(0.2)| —          | —          |
| 171 | 170 | 45.6(0.3)| —         | 44(2)     | 57.2(0.2)| 55(80)     | —          |
| 174 | 173 | 54.9(0.3)| —         | 51(2)     | 65.1(0.2)| —          | —          |
| 173 | 171 | 125.1(0.6)|—         | 116(3)   | 146.3(0.2)| —          | —          |

$a$ Ref. [78] presents only statistical errors in the uncertainty — the large uncertainty in the mass shift parameter used in calculation is not propagated through. Propagating the uncertainty from the mass shift parameter leads to an uncertainty of $\sim 9 \times 10^{-3}$ fm$^2$ for the first row.  
$b$ Purely computational values (presented without uncertainty)
VII. KING PLOT AND CLOCK TRANSITION ISOTOPE SHIFTS

A King plot compares the isotopic shifts of one transition, \( i \), against that of another, \( j \). By scaling the isotope shift with the reciprocal of the nuclear mass parameter, one defines the modified isotope shift,

\[
\xi_i^{A,A'} = \delta \nu_i^{A,A'}/\mu^{A,A'}.
\]

From Eq. 5 and assuming the nuclear parameters \( \lambda^{A,A'} \) and \( \mu^{A,A'} \) are the same for both (all) transitions, one finds,

\[
\xi_i^{A,A'} = (F_i/F_j)\xi_j^{A,A'} + (K_i - K_jF_i/F_j).
\]

A plot of \( \xi_i^{AA'} \) versus \( \xi_j^{AA'} \) should thus, to first order, form a straight line with slope \( F_i/F_j \) and intercept \( K_i - K_jF_i/F_j \), known as a King plot. A King plot constructed from the measured isotope shifts for the clock and intercombination lines in Yb t is presented in Figure 3. With only three isotopes having frequency measurements for the clock transition, only two independent data points can be used to create the King plot (the 171–174 pairing makes it overdetermined). The gradient and intercept for the linear ‘fit’ are 1.0138(12) and −0.102(22) THz u, respectively, where the uncertainties are derived from the square roots of the diagonal entries to the covariance matrix calculated using an orthogonal distance regression [86].

The gradient is given as \( F_{\text{ICL}}/F_{\text{clock}} \) and the intercept as \( K_{\text{ICL}} - K_{\text{clock}} F_{\text{ICL}}/F_{\text{clock}} \). The calculated values in Table IX produce a gradient of 1.0095(28), and an intercept of 0.01(11) THz u. This gradient and intercept values are not inconsistent with those obtained from the King plot in Fig. 3 (experimental).

The unknown isotope shifts for the \( ^1S_0 - ^3P_0 \) clock transition can be estimated in three ways (further information follows),

1. Energy level differences for each isotope are found through MCDHF-CI computations. From these, the isotope shifts are evaluated, and because there is a consistent offset from measured values in \( ^1S_0 - ^3P_1 \), can be scaled to match the three known experimental isotope shifts.

2. The mass shift and field shift parameters calculated using Mtx4 are used with the ICL isotope shifts presented in Table IX to estimate the clock transition isotope shifts. This estimate is based predominantly on theoretical calculation.

3. The modified frequency shifts are extrapolated from a King plot constructed using the clock and intercombination lines, and converted back into isotope shifts. This estimate is based predominantly on experimental measurement.

The estimated isotope shifts for the clock transition for each method are presented in Table IX.

| \( A \) | \( A' \) | Method-1 | Method-2 | Method-3 |
|---|---|---|---|---|
| 176 | 174 | −949(10) | −945.1(7.3) | −949.5(2.8) |
| 174 | 172 | −1002(11) | −990.7(7.5) | −995.0(2.9) |
| 172 | 170 | −1272(13) | −1273.0(8.0) | −1275.3(3.3) |
| 170 | 168 | −1347(14) | −1356.0(8.2) | −1357.9(3.4) |
| 173 | 172 | −448(5) | −440.0(3.7) | −443.0(1.4) |
| 172 | 171 | −803(9) | −817.5(4.2) | −816.7(2.0) |
| 171 | 170 | −469(5) | −455.5(3.8) | −458.5(1.5) |
| 174 | 173 | −554(6) | −550.6(3.8) | −552.0(1.5) |
| 173 | 171 | −1251(13) | −1257.6(7.9) | −1259.8(3.3) |

(Method-1): The \( \text{ab initio} \) isotope shifts calculated for the clock transition using MCDHF-CI computations, presented in Table IX, are larger than experimental values by \( \sim 11\% \) (for all the isotopes). This difference we attribute to a systematic effect in the calculations, which we can account for by a scaling factor. Accounting for the difference leads to the estimates given in the ‘Method-1’ column of Table IX. The adjusted isotope shift between \( ^{173}\text{Yb} \) and \( ^{174}\text{Yb} \) is −554 MHz and between \( ^{173}\text{Yb} \) and \( ^{173}\text{Yb} \) is −1251 MHz, at differences from experiment of 0.5% and −0.7%, respectively. In line with these values we place uncertainties of 1% on the remaining shifts in...
Table IX (Method-1). We regard this as the least reliable estimate of the unmeasured clock line isotope shifts.

(Method-2): Equation 5 applies for both the clock and ICL transitions, with the nuclear parameters taken to be independent of the electronic states. Substituting for $\lambda^{A,A'}$ between these two equations leads to

$$\delta\nu_{\text{clock}}^{A,A'} = \left( K_{\text{clock}} - F_{\text{clock}}^K K_{\text{ICL}}^{A,A'} \right) \mu^{A,A'} + F_{\text{clock}}^K \delta\nu_{\text{ICL}}^{A,A'}.$$  

The ICL isotope shifts presented in Table VII can be used with the calculated mass shift and field shift parameters to arrive at the clock transition isotope shifts. This is equivalent to constructing a King plot using the theoretical mass and field shifts computed using an Ab initio method, similar to those for Table VII.

(METHOD-3): Assuming King linearity holds, the King plot in Figure 3 can be extrapolated to arrive at the clock transition isotope shifts for other pairings. These estimates are presented in the final column of Table IX.

VIII. CONCLUSIONS

*Ab initio* computations of the isotope shifts for the clock transition and its partnering intercombination line ($^1S_0 - ^3P_1$) have been performed separately for each stable isotope using a MCDHF-CI method implemented by the GRASP2018 [47] package. Absolute transition frequency measurements agree with experimental results to less than 1% error, with isotope shifts differing from experimental values by 11%. Using these same computations, the hyperfine interaction constants for the $^3P_1$ state have been calculated to within 4% of corresponding experimental values. Corrections of the centroids of the hyperfine manifolds for the second-order hyperfine interaction in the fermionic isotopes have also been made.

The electronic mass shift and field shift parameters are computed with the program RIS4 [46] using the results of the MCDHF-CI computations. The corrected isotope shifts for the intercombination line together with these electronic mass shift and field shift parameters enable computation of the nuclear charge parameters, $\lambda^{A,A'}$, consistent with previous results, but with an estimated order of magnitude reduction in uncertainties. The differences in mean-square charge radii, $\delta\langle r^2\rangle^{A,A'}$, are calculated and found to be significantly smaller than tabulated values in Angeli and Marinova [75].

Experimental isotope shifts for the clock and intercombination lines, corrected for the second-order hyperfine interaction, have been used to construct a King plot with two data points. This King plot is used to estimate the isotope shifts for the clock transition for the undiscovered bosonic isotopes. These estimates are found to be reasonably consistent with estimates based on the calculated mass shift and field shift parameters.

The computations may be increased in size by including deeper core-valence correlation, and by extending the active set of orbitals beyond a principal quantum number of 12, given sufficient computational resources. The inclusion of deeper core-valence correlation is expected to reduce the 11% discrepancy between the computed and experimental isotope shifts [27, 47]. Different nuclear models, including models accounting for the known deformation of Yb nuclei, may also be explored to investigate their potential systematic effects on the computed results.

With suggestions to combine the results of Counts et al. [4] with isotope shift measurements of a clock transition in neutral ytterbium [41, 17], the undiscovered bosonic-isotope clock transitions should be sought using the isotope shift estimates presented in this work (e.g. with cold Yb atoms in an optical lattice trap and a DC magnetic field applied [87]). Once the clock isotope shifts are identified, King plots can be constructed with other high-precision isotope shift measurements in neutral and ionised ytterbium in order to investigate King nonlinearity and identify or constrain physics beyond the Standard Model.

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APPENDIX A: NUCLEAR MODEL

The nuclear charge distribution is modelled as a two-component Fermi distribution \[ \rho(r) = \frac{\rho_0}{1 + e^{(r-c)/a}}, \] (A1)

where \( c \) is the half-density radius, \( a \) is related to the nuclear skin thickness \( t \) by \( t = (4\ln 3)a \), and \( \rho_0 \) is a normalisation factor such that

\[ \int_0^\infty 4\pi r^2 \rho(r) dr = Z. \] (A2)

For all isotopes the atomic number is \( Z = 70 \) and the nuclear skin thickness is taken to be \( t = 2.18(2) \text{ fm} \) \[76\]. This value for the nuclear skin thickness is less than the typical value of \( t = 2.3 \text{ fm} \) assumed for most nuclei \[88\]; however, it is the only value found for Yb which includes an explicit uncertainty. Other authors have used \( t = 2.3 \text{ fm} \) \[78\] or \( t = 2.4 \text{ fm} \) \[90\]. The dependence of the results upon the skin thickness was investigated and found to be insignificant. The nuclear parameters used in the MCDHF-CI computations are presented in Table X. In addition to these, the only isotopes with non-zero nuclear spin and magnetic dipole moment are \( ^{171}\text{Yb} \) and \( ^{173}\text{Yb} \). \( ^{171}\text{Yb} \) has a nuclear spin of \( I = 1/2 \) and a magnetic dipole moment of \( \mu = 0.49367(1) \mu_N \) \[91\]. \( ^{173}\text{Yb} \) has a nuclear spin of \( I = 5/2 \) and a magnetic dipole moment of \( \mu = -0.67989(3) \mu_N \) \[91\], and nuclear electric quadrupole moment of \( Q = 2.80(4) \text{ b} \) \[57\].

APPENDIX B: STATE COMPOSITIONS

The atomic state functions determined using the MCDHF-CI method consist of weighted combinations of many configuration state functions (CSFs). The percentage contributions of the most significant CSFs are listed for the \( ^1S_0 \) ground state and the \( ^3P^0_{0,1} \) excited states in Table XI. Our values are consistent with those reported by Migdalek and Baylis \[38\], where their calculation extended only to our MR set.

### Table XI. The highest contributing CSFs in the compositions of three Yb \( ^1 \) atomic states.

| CSF          | Percentage |
|--------------|------------|
| \( ^1S_0 \)   | 91.56%     |
| \( 6s^2 \)    | 1.87%      |
| \( 6p^2 \)    | 1.31%      |
| \( 5d^2 \)    | 0.74%      |
| \( 5d^2 \)    | 0.56%      |
| \( 5d^2 \)    | 0.35%      |
| \( ^3P^0_{0} \)  | 95.60%     |
| \( 6s6p_- \)  | 22.02%     |
| \( 5d_{-6p}^+ \) | 0.68%      |
| \( 5d_{-6p}^- \) | 0.46%      |
| \( 6p_{-7s} \) | 0.38%      |

APPENDIX C: UNCERTAINTY ESTIMATES FOR ISOTOPE SHIFT PARAMETERS

Systematic expansions of the active space and correlation model have been undertaken in order to estimate the uncertainties for the isotope shift parameters, \( K \) and \( F \). The error introduced by truncating the active space at \( 12spdf \) is estimated by analysing the \( K \) and \( F \) values after adding each new correlation layer. This analysis was performed using \( ^{174}\text{Yb} \) with core-valence correlations restricted to single excitations from \( 5s, 5p, 4f \) and unrestricted valence-valence correlations. The results are presented in Table XII. Based on these results, the uncertainty in the final \( K \) and \( F \) values due to the truncated active space is estimated to be the absolute difference between the \( 12sp11d10f \) and \( 11sp10d9f \) layers, as these were the largest two correlation layers added with an orbital of each symmetry.

The error introduced by restricting the core-valence correlation to single excitations from \( 5s, 5p, 4f \) is esti-
TABLE XII. Sequences of isotope shift parameters upon addition of correlation layers. The final row constitutes estimates for the uncertainty in each of the parameters for each isotope. The units for $K$ are GHz u and the units for $F$ are GHz fm$^{-2}$.

| Layer   | $K_{\text{clock}}$ | $F_{\text{clock}}$ | $K_{\text{ICL}}$ | $F_{\text{ICL}}$ |
|---------|---------------------|---------------------|-------------------|-------------------|
| 5sp$^5$d5f | -192.67             | -10.4960            | -176.48           | -10.5151          |
| 8sp$^7$d6f | -204.07             | -10.0544            | -189.54           | -10.1697          |
| 9sp$^8$d7f | -282.05             | -10.9553            | -273.16           | -11.0695          |
| 10sp$^9$d8f | -268.73             | -10.9639            | -257.58           | -11.0710          |
| 11sp$^{10}$d9f | -290.47           | -10.8480            | -281.57           | -10.9548          |
| 12sp$^{11}$d10f | -288.15           | -10.8386            | -279.59           | -10.9418          |
| 12spdf | -288.07             | -10.8393            | -279.55           | -10.9425          |

Uncertainty estimate 2.4 0.0094 2.0 0.013

TABLE XIII. Sequences of isotope shift parameters upon inclusion of deeper core-valence correlation. The final row constitutes estimates for the uncertainty in each of the parameters for each isotope. The units for $K$ are GHz u and the units for $F$ are GHz fm$^{-2}$.

| Available core | $K_{\text{clock}}$ | $F_{\text{clock}}$ | $K_{\text{ICL}}$ | $F_{\text{ICL}}$ |
|----------------|---------------------|---------------------|-------------------|-------------------|
| 4f             | 54.90               | -9.4283             | 56.36             | -9.5023           |
| 5p, 4f         | -265.41             | -10.5830            | -255.65           | -10.6681          |
| 5s, 5p, 4f     | -288.05             | -10.8326            | -279.53           | -10.9358          |
| 5s, 4d, 5p, 4f | -325.22             | -10.8255            | -315.26           | -10.9297          |

Uncertainty estimate 75 0.015 72 0.013

APPENDIX D: ALTERNATIVE PRESENTATION OF DIFFERENCES IN MEAN-SQUARE CHARGE RADII

The differences in mean-square charge radii are presented in Table VIII for pairs of isotopes. Alternatively, a single reference isotope may be chosen and differences in mean-square charge radii given relative to this reference isotope. For ytterbium, this reference isotope is commonly chosen to be $^{176}$Yb. Differences in mean-square nuclear charge radii of this type are presented in Table XIV with the reference isotope of $^{176}$Yb.

Table XIV. Differences in mean-square nuclear charge radii relative to $^{176}$Yb, $\delta(\langle r^2 \rangle)$, in units of $10^{-3}$ fm$^2$ (column 2). Data from other works are presented for comparison.

| $A'$          | This work | Ref. [76] | Ref. [78] |
|---------------|-----------|-----------|-----------|
| 174           | 94.4(0.5) | 90(2)     | 115.9(0.1)|
| 173           | 149.3(0.8)| 142(3)    | 181.0(0.1)|
| 172           | 193.4(1.0)| 184(5)    | 236.6(0.1)|
| 171           | 274.4(1.3)| 259(6)    | 327.3(0.1)|
| 170           | 320.0(1.6)| 303(7)    | 384.5(0.1)|
| 168           | 454.8(2.1)| 428(13)   | 540.6(0.3)|

APPENDIX E: CLOCK TRANSITION FREQUENCIES

Table XV lists our estimates for the absolute $^1S_0 - ^3P_0$ transition frequencies in neutral ytterbium for isotopes where it is yet to be measured, together with the known frequencies. Our estimates and their uncertainties are based on the weighted mean of the isotope shift values presented in Table IX using Methods 2 and 3, and the existing absolute transition frequency measurements.

Table XV. Estimated (this work) and previously measured clock transition frequencies in Yb 1.

| Isotope | Transition frequency (MHz) |
|---------|---------------------------|
| 168     | 518 297 652.3(3.5)         |
| 170     | 518 296 294.7(1.4)         |
| 171     | 518 295 836.59086361(13)   | S2 |
| 172     | 518 295 836.59086371(11)   | S1 |
| 173     | 518 295 019.7(1.9)         |
| 174     | 518 294 857.845268(10)     | S3 |
| 176     | 518 293 076.4(2.7)         |

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