Relativistic full-configuration-interaction calculations of magic wavelengths for the $2^3S_1 \rightarrow 2^1S_0$ transition of helium isotopes

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A large-scale full-configuration-interaction calculation based on Dirac-Coulomb-Breit (DCB) Hamiltonian is performed for the $2^1S_0$ and $2^3S_1$ states of helium. The operators of the normal and specific mass shifts are directly included in the DCB framework to take the finite nuclear mass correction into account. Accurate energies and matrix elements involved (the main quantum number) up to 13 are tabulated. Specially, the accuracy of energies and matrix elements can be remained even for the Rydberg states from one diagonalization of Hamiltonian. The dynamic dipole polarizabilities are calculated by using the sum rule of intermediate states. And a series of magic wavelengths around 320 nm for the $2^3S_1 \rightarrow 2^1S_0$ transition of helium are identified with 5-7 significant digits. The high-order ac Stark shift determined by the dynamic hyperpolarizabilities at the magic wavelengths are also evaluated. Since the most promising magic wavelength for application in experiment is at 319.8 nm, the high-accuracy magic wavelengths of 319.816 07(9) nm and 319.831 31(9) nm for $2^3S_1(M_J = \pm 1) \rightarrow 2^1S_0$ transition of $^4$He and $^3$He, respectively, are reported in present work, which provides theoretical support for experimental designing an optical dipole trap to precisely determine the nuclear charge radius of helium.

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The long-term outstanding proton radius puzzle causes great interest in recent years [1, 5]. So far there has not been a satisfying explanation for the discrepancy of 5.6σ in the proton size derived from muonic hydrogen Lamb shift measurements [1, 2] and the accepted CODATA value [3]. Research in this field has expanded to measurements of the 2S-4P transition energy in hydrogen [4], transition energies between circular Rydberg states in heavy-H-like ions [5], and the 1S-2S transition energy in muonic helium ions [6]. In order to help solve the proton size puzzle, the measurement of high-precision spectroscopy in helium isotopes has become an additional contribution to this field [7–12]. However, the nuclear charge radius difference determined from the $2^3S \rightarrow 2^1S$ and $2^3S \rightarrow 2^3P$ transitions of helium disagrees by 4σ [7–12]. Even combined with the recent theoretical investigations [13, 15], where the higher-order recoil corrections are taken into account, the 4σ discrepancy does still exist and remains unexplained by any missed corrections in existing theoretical predictions. So this discrepancy calls for the verification of the experimental transition frequencies by independent measurements.

For the $2^3S \rightarrow 2^3P$ transition frequency of helium, recently, the frequency measurement of $^4$He is achieved to $5.1 \times 10^{-12}$ [16], which is more accurate than the early result of Refs. [11, 12, 17]. But it’s interesting that when the $2^3S \rightarrow 2^3P$ transition frequency of $^4$He of Ref. [12] is replaced with the latest value of Ref. [16], the resulting nuclear charge radii difference agrees well with the value derived from the $2^3S \rightarrow 2^1S$ transition [7] but differs with the value from $2^3S \rightarrow 2^3P$ transition [9]. This deviation indicates the need for further independent measurements of He isotope shifts.

For the $2^3S \rightarrow 2^1S$ transition frequency of $^4$He, the measurement accuracy is $9.4 \times 10^{-12}$ [7], which is less accurate than the frequency measurement of $2^3S \rightarrow 2^3P$ transition [16]. One of the main systematic uncertainty of Ref. [7] comes from ac Stark shift. Implementation of a magic wavelength trap can solve this problem in many high-precision measurement [18, 19]. Recently, Notermans et. al. obtain the magic wavelengths of He($2^3S \rightarrow 2^1S$) with use of available energies and Einstein A coefficients [20]. The accuracy of their values are limited by extrapolated contributions from continuums. Since the dynamic dipole polarizability at the 319.8 nm magic wavelength is large enough to provide sufficient trap depth at reasonable laser powers while the scattering lifetime is accepted, the 319.8 nm magic wavelength is proposed to design a optical dipole trap (ODT) to eliminate the ac Stark shift [20]. In order to determine the nuclear charge radius difference with a precision comparable to the muonic helium ion, Vassen et. al. aim to measure the $2^3S \rightarrow 2^1S$ transition with sub-kHz precision. At this level of precision, the ab-initio calculation for the accurate magic wavelengths of helium isotopes are requisite.

In this paper, we add the mass shift operators directly into the Dirac-Coulomb-Breit (DCB) Hamiltonian to improve the previous relativistic configuration interaction (RCI) method [21]. Then we perform a larger-scale full-configuration-interaction calculation of the dynamic dipole polarizabilities for the $2^3S$ and $2^1S$ states.
of $^4\text{He}$ and $^3\text{He}$. A series of magic wavelengths for the $2^3S_1 \rightarrow 2^1S_0$ transition are accurately identified according to the dynamic polarizabilities. In order to do a cross test of the magic wavelengths, we also carry out a non-relativistic calculations of dynamic polarizabilities and hyperpolarizabilities of helium by using the newly developed Hylleraas-B-spline method [22]. Our results of all the magic wavelengths from two different theoretical methods are in good agreement. Specially, we report our recommended magic wavelengths of 319.816079(9) nm and 319.83131(9) nm for the $2^3S_1 (M_J = \pm 1) \rightarrow 2^1S_0$ transition of $^4\text{He}$ and $^3\text{He}$, respectively, which provides a theoretical guidance to design ODT for developments of high-precision spectroscopy measurements in helium.

The DCB Hamiltonian with mass shift (MS) operator included for the two-electron atomic system is written as

$$H = \sum_{i=1}^{2} \left[ \alpha_i \cdot p_i + \beta m_e c^2 \cdot \frac{Z}{r_i} \right] + \frac{1}{r_{12}} - \frac{1}{2r_{12}} \left[ \alpha_1 \cdot \alpha_2 + (\alpha_1 \cdot \hat{r}_{12}) (\alpha_2 \cdot \hat{r}_{12}) \right] + H_{MS},$$

where $c = 137.035999074$ is the speed of light [22], $Z$ is the nuclear charge, $\beta$ is the $4 \times 4$ Dirac matrix, $m_e = 1$ is the electron mass, $\alpha_i$ and $p_i$ are respectively the Dirac matrix and the momentum operator for the $i$-th electron, $\hat{r}_{12}$ is the unit vector of the electron-electron distance $r_{12}$, and the MS operator $H_{MS}$ includes the leading term of normal and special mass shift (NMS, SMS) operators,

$$H_{MS} = H_{NMS} + H_{SMS} = \sum_{i=1}^{2} \frac{p_i^2}{2m_0} + \frac{p_1 \cdot p_2}{m_0},$$

with $m_0$ being the nuclear mass. For $^4\text{He}$, $m_0 = 7294.2995361 m_e$, and for $^3\text{He}$, $m_0 = 5495.8852754 m_e$ [23]. The wave function $\psi_{ij}(JM_J)$ of helium for a state with angular momentum $(J, M_J)$ is expanded as a linear combination of the configuration-state wave functions $\phi_{ij}(JM_J)$, which are constructed by the single-electron wave functions. Using the Notre Dame basis sets [24, 25] of $N$ number of B-spline functions, the single-electron wave functions are obtained by solving the single-electron Dirac equation.

The non-relativistic Hamiltonian of two-electron atomic system with the infinite nuclear mass is solved

$$\alpha_1(\omega) = \alpha_1^S(\omega) + \frac{3M_e^2 - J_g(J_g + 1)}{J_g(2J_g - 1)} \alpha_1^T(\omega),$$

where $\alpha_1^S(\omega)$ and $\alpha_1^T(\omega)$ are the scalar and tensor dipole polarizabilities, respectively, which can be expressed as the summation over all intermediate states,

$$\alpha_1^S(\omega) = \sum_{n \neq g} \frac{f_{gn}^{(1)}}{(\Delta E_{gn})^2 - \omega^2},$$

with $f_{gn}^{(1)}$ is the dipole oscillator strength,

$$f_{gn}^{(1)} = \frac{2|\langle N_g J_g | T_1 | N_n J_n \rangle|^2 \Delta E_{gn}}{3(2J_g + 1)},$$

where $\Delta E_{gn} = E_n - E_g$ is transition energy between the initial state $| N_g J_g \rangle$ and the intermediate state $| N_n J_n \rangle$, and $T_1$ is the dipole transition operator. The nonrelativistic polarizabilities are obtained by replacing $J$ number with $L$ number in Eqs. [4] - [7].

The nonrelativistic dynamic hyperpolarizability for $S$ state is

$$\gamma_0(\omega) = \frac{128\pi^2}{3} \frac{1}{9} T(1,0,1,\omega) + \frac{2}{45} T(1,2,1,\omega),$$

where $T(1,0,1,\omega)$ and $T(1,2,1,\omega)$ are the dipole transition operator.
where $\mathcal{T}(L_a, L_b, L_c, \omega)$ is written as

\[
\begin{align*}
\mathcal{T}(L_a, L_b, L_c, \omega) &= \sum_{mnk} \frac{\langle 0L|T_1|mL_a\rangle\langle mL_a|T_3|nL_b\rangle\langle nL_b|T_1|kL_c\rangle\langle kL_c|T_1|0L\rangle}{(\Delta E_{m0} - \omega)(\Delta E_{k0} - \omega)} \\
&\quad - \delta_{L_b, L} \sum_m \frac{|\langle 0L|T_1|mL_a\rangle|^2}{(\Delta E_{m0} - \omega)} \sum_k \frac{|\langle 0L|T_1|kL_c\rangle|^2}{(\Delta E_{k0} - \omega)^2}.
\end{align*}
\]

The accurate calculations of the dynamic hyperpolarizabilities is very difficult, since the formula involves three summations over different intermediated states, which cannot be properly evaluated by summing over low-lying contributions.

The magic wavelengths of the $2^1S \rightarrow 2^1S$ transition are determined from the calculations of the dynamic dipole polarizabilities of the $2^1S_0$ and $2^1S_1$ states to find their crossing points. The accuracy of magic wavelengths depends on accurate energies and wavefunctions of initial and intermediate states. The high-precision B spline RCI method was very successful in accurate calculation of atomic polarizabilities for the $2^3S_1$ state of helium [21]. However, for the $2^1S_0$ state, it’s clearly seen from Figs. 1, 2, 3, the magic wavelengths in the range of 300 nm to 400 nm involve a series of $2^1S_0 \rightarrow n^1P_1$ ($n \leq 13$) resonance transitions. Especially for the magic wavelengths around 320 nm that we are interested in the present work, they are located at the edge of the $2^1S_0 \rightarrow 10^1P_1$, $2^1S_0 \rightarrow 11^1P_1$, and $2^1S_0 \rightarrow 12^1P_1$ transitions. The accurate determination of these magic wavelengths near these resonance frequencies requires construction of sufficient configurations in a sufficient box size to make sure that all the transition energies from the $2^1S_0$ state transit to the $n^1P_1$ intermediate states, especially to the $10^1P_1$, $11^1P_1$, and $12^1P_1$ Rydberg states, are accurate. This is
a biggest challenge for our RCI calculation.

In order to get accurate energy for the \( ^2 \! P_\text{o} \) state, we chose the box size \( R = 200 \text{ a.u.} \) and increase the partial wave \( l_{\text{max}} \) to 20, then we can get the final convergent energy of \(-2.145786 \text{ a.u.} \) for the \( ^2 \! S_\text{o} \) state. Our result has six same digits with the best value of \(-2.145787 \text{ a.u.} \), which is derived by adding the finite nuclear mass, relativistic, and anomalous magnetic moment corrections on the non-relativistic Hylleraas energy \([26]\). In order to get accurate energies for the intermediate \( n \) \( ^1 \! P_1 \) states, we adjust the box size \( R = 600 \text{ a.u.} \) to make sure the energies of the \( n \) \( ^1 \! P_1 \) states are negative in the present calculation. Then we fix the box size \( R=600 \text{ a.u.} \) and increase the partial wave \( l_{\text{max}} \) to 7 to test the convergence of magic wavelengths. In this work, the maximal number of configurations is nearly 90000, the time consuming and large memory occupy for calculating the electron-electron Coulomb and Breit interaction integrals are the biggest problems. We improve and optimize the previous RCI program by using OpenMP parallel and block calculations to overcome the resulting problems, which significantly expands the applicability of the previous RCI method. Extensive tests of the numerical stability for the energies, matrix elements, dipole polarizabilities, and magic wavelengths of helium are carried out.

**TABLE I:** Comparison of present RCI energies (in a.u.) for some selective \( n \) \( ^1 \! P_1 \) \((n \leq 13)\) states of \(^4\text{He}\). The numbers in parentheses are computational uncertainties.

| State  | RCI     | Hylleraas \([26]\) |
|--------|---------|-------------------|
| \( ^2 \! P_1 \) | -2.123 650 17(2) | -2.123 654 51 |
| \( ^3 \! P_1 \) | -2.054 968 56(2) | -2.054 970 17 |
| \( ^4 \! P_1 \) | -2.030 896 59(2) | -2.030 897 47 |
| \( ^5 \! P_1 \) | -2.019 734 98(2) | -2.019 735 59 |
| \( ^6 \! P_1 \) | -2.013 664 02(2) | -2.013 664 52 |
| \( ^7 \! P_1 \) | -2.009 999 98(2) | -2.010 000 41 |
| \( ^8 \! P_1 \) | -2.007 620 18(2) | -2.007 620 57 |
| \( ^9 \! P_1 \) | -2.005 987 70(2) | -2.005 988 07 |
| \( ^{10} \! P_1 \) | -2.004 819 48(2) | -2.004 819 84 |
| \( ^{11} \! P_1 \) | -2.003 954 83(2) | |
| \( ^{12} \! P_1 \) | -2.003 954 99(4) | |
| \( ^{13} \! P_1 \) | -2.002 784 91(4) | |

Table I presents the RCI energies for the singlet \( P \) states of \(^4\text{He}\). All the energies for \( n \) \( ^1 \! P_1 \) \((n \leq 13)\) intermediate states have eight significant digits. That means the energy accuracy for all the states, even for the Rydberg states, can be guaranteed to the same level of precision just from one diagonalization in present RCI calculations. The Hylleraas energies, which include the finite nuclear mass, relativistic, and anomalous magnetic moment corrections, are derived by combining the values in the Tables 11.7 and 11.8 of Drake’s work \([26]\) and the ground-state energy of \(-1.999832 \text{ a.u.} \) of He \(^4\). The comparison of the energies shows that our energies are in good agreement with the accurate Hylleraas energies \([26]\).

**TABLE II:** Comparison of some reduced matrix elements between present RCI calculations and Hylleraas calculations \([27]\) of the \( ^2 \! S_\text{o} \rightarrow n \) \( ^1 \! P_1 \) \((n \leq 13)\) transitions for \(^4\text{He}\). The numbers in parentheses are computational uncertainties.

| State | RCI        | Hylleraas \([27]\) |
|-------|------------|-------------------|
| \( ^2 \! S_\text{o} \rightarrow n \) \( ^1 \! P_1 \) | | |
| \( ^2 \) | 5.052 06(8) | 5.050 977 |
| \( ^3 \) | 1.580 81(2) | 1.581 082 |
| \( ^4 \) | 0.801 03(2) | 0.801 106 |
| \( ^5 \) | 0.515 53(2) | 0.515 578 |
| \( ^6 \) | 0.371 14(2) | 0.371 159 |
| \( ^7 \) | 0.285 12(2) | 0.285 131 |
| \( ^8 \) | 0.228 58(2) | 0.228 590 |
| \( ^9 \) | 0.188 89(2) | 0.188 899 |
| 10      | 0.159 67(2) | 0.159 686 |
| 11      | 0.137 39(2) | |
| 12      | 0.119 92(2) | |
| 13      | 0.105 89(2) | |

**TABLE III:** Convergence test of 319.8 nm magic wavelength for the \( ^2 \! S_\text{o} \rightarrow 2 \) \( ^4 \! P_1 \) \((M_J = \pm 1)\) transition of \(^4\text{He}\) and \(^3\text{He}\) as the number of B-splines, \( N \), increased by fixing the number of the partial wave as \( l_{\text{max}} = 7 \) and choosing the box size as \( R_0 = 600 \text{ a.u.} \). The dynamic dipole polarizabilities at the corresponding magic wavelengths are also listed.

| \( N \) | \( ^4\text{He} \) | \( ^3\text{He} \) | \( ^3\text{He} \) | \( ^4\text{He} \) |
|--------|----------------|----------------|----------------|----------------|
| 40     | 319.830 167 319.845 384 | 183.479 869 183.691 003 | | |
| 45     | 319.817 599 319.832 836 | 186.225 612 186.440 744 | | |
| 50     | 319.816 205 319.831 441 | 186.531 280 186.743 237 | | |
| 55     | 319.816 090 319.831 324 | 186.555 980 186.772 052 | | |
| 60     | 319.816 078 319.831 312 | 186.559 001 186.774 338 | | |
| Extrap. | 319.816 07(2) 319.831 31(2) | 186.560(2) 186.775(2) | | |

Table II gives a comparison of the reduced matrix elements for the dipole allowed \( ^2 \! S_\text{o} \rightarrow n \) \( ^1 \! P_1 \) \((n \leq 13)\) transitions. The Hylleraas values are derived by using available oscillator strengths and the transition energies of Ref. \([27]\), which includes the finite nuclear mass and the leading-order of relativistic corrections. It shows that our matrix elements have at least four significant digits. Compared with the converted values from the work of Drake and Morton \([27]\), present RCI results have four same digits for the \( ^2 \! S_\text{o} \rightarrow n \) \( ^1 \! P_1 \) \((n \geq 5)\) transitions. Other energies and reduced matrix elements \((n \leq 13)\) for \(^4\text{He}\) and \(^3\text{He}\) are presented in Supplemental Material.

Since the 319.8 nm magic wavelength was proposed to trap helium for high-precision measurement. Table III lists a convergent test for the 319.8 nm magic wavelength of the \( ^2 \! S_\text{o} \rightarrow 2 \) \( ^4 \! P_1 \) \((M_J = \pm 1)\) transition of helium isotopes as the number of B-splines increased, and the corresponding dynamic polarizabilities at the magic wavelengths are also listed. As \( N \) increased to 60, the magic wavelengths converge to the seventh significant digit, and the corresponding dynamic dipole polarizabilities converge to the fifth significant digit. The extrapolated val-
ues of the magic wavelength are 319.816 07(2) nm and 319.831 31(2) nm for $^4$He and $^3$He, respectively. They are more accurate than the semi-empirical results of 319.815 nm and 319.830 nm. Since the maximal difference of the matrix elements between ours and Drake’s in Table III is about 0.022%, we indicate a 0.05% difference for all the reduced matrix elements of $2^3S_i \rightarrow n^1P_j$ transitions for conversation. We found this way produces 0.05 picometer (pm) difference on the extrapolated values of Table III. In order to estimate the correction from the incomplete configuration space, we double the uncertainties of the extrapolated values and take the 0.05 pm as another part of the uncertainties. Then we can give the recommended values of the magic wavelength of 319.816 07(9) nm for $^4$He and 319.831 31(9) nm for $^3$He.

### Table IV: Contributions of individual transitions to the dynamic dipole polarizabilities (in a.u.) of the $2^3S_0$ and $2^3S_i(M_J = \pm 1)$ states of $^4$He at the magic wavelengths 319.816 nm, 321.411 nm, and 323.589 nm.

| Contribution | 319.816 nm | 321.411 nm | 323.589 nm |
|--------------|------------|------------|------------|
| $2^3S_0$     |            |            |            |
| $2^1P_1$     | $-19.016$   | $-19.211$  | $-19.479$  |
| $3^1P_1$     | $-12.556$   | $-12.769$  | $-13.067$  |
| $4^1P_1$     | $-6.924$    | $-7.126$   | $-7.416$   |
| $5^1P_1$     | $-5.066$    | $-5.308$   | $-5.672$   |
| $6^1P_1$     | $-4.271$    | $-4.596$   | $-5.119$   |
| $7^1P_1$     | $-3.958$    | $-4.348$   | $-5.300$   |
| $8^1P_1$     | $-3.986$    | $-4.783$   | $-6.534$   |
| $9^1P_1$     | $-4.413$    | $-6.020$   | $-11.762$  |
| $10^1P_1$    | $-5.633$    | $-10.679$  | $52.919$   |
| $11^1P_1$    | $-9.870$    | $88.632$   | $6.159$    |
| $12^1P_1$    | $220.585$   | $6.593$    | $2.864$    |
| $13^1P_1$    | $7.015$     | $3.025$    | $1.715$    |
| Remainder    | $34.657$    | $29.624$   | $25.899$   |
| Total        | $186.559$   | $52.942$   | $15.207$   |

Except the important application of 319.816 nm magic wavelength of $^4$He, both of the magic wavelengths 321.411 nm and 323.589 nm can also be used to design experiments once high-power laser can be realized. Table IV gives a breakdown of the various contributions from different intermediate states to the dynamic dipole polarizability of the $2^3S_0$ and $2^3S_i(M_J = \pm 1)$ states of $^4$He at the magic wavelengths 319.816 nm, 321.411 nm and 323.589 nm. The contributions from the first 13 $n^1P_j$ states are separately listed for the $2^1S_0$ state. The dominant contribution at the three magic wavelengths 319.816 nm, 321.321.411 nm, and 323.589 nm comes from the $2^1S_0 \rightarrow 2^1P_1$, $2^1S_0 \rightarrow 1^1P_2$, and $2^3S_0 \rightarrow 1^3P_1$ resonance transitions, respectively. The contributions from $n^1P_j(n \leq 11)$ at the 319.816 nm magic wavelength are negative, which result in cancellations in the oscillator strength sum. For the $2^3S_1(M_J = \pm 1)$ state, the dominate contributions to the polarizabilities at the three magic wavelengths are from the $2^3S_1 \rightarrow 4^3P_1$ and $2^3S_1 \rightarrow 4^3P_2$ transitions.

Table V summarizes the first nine magic wavelengths in the range of 318 - 413 nm from the calculations of both Hylleraas-B-splines and RCI methods. All the magic wavelengths are extrapolated directly from their convergence test. All the values from two different theoretical methods are consistent. The relativistic and finite nuclear mass corrections on all the magic wavelengths are less than 62.3 pm. For the RCI calculation, the difference of all the magic wavelengths between $^4$He and $^3$He are less than 16.5 pm.

For the dynamic hyperpolarizabilities, using the Hylleraas-B-splines method, we get the values of $-1.0 \times 10^{10}$ a.u. and $-3.3 \times 10^{9}$ a.u. at the 319.8 nm magic wavelengths for the $2^3S$ and $2^3S$ states of $^\infty$He, respectively. The difference of the dynamic hyperpolarizabilities at this magic wavelength for the $2^3S \rightarrow 2^1S$ transition is $\Delta \gamma_0(\omega) = 6.7 \times 10^9$ a.u. If the power of the incident trapping laser beam is $P = 0.2$ W with beam waist $w_0 = 85 \mu m$, then we can get the electric field intensity $F^2 \approx 6.3 \times 10^{-28}$ a.u. Accordingly, the higher-order ac Stark shift is evaluated as $\frac{1}{2} \Delta \gamma_0(\omega) F^4 \approx 1.7 \times 10^{-19}$ a.u.$\approx 1.2$ mHz, it is smaller by six orders of magnitude than the 1.8 kHz uncertainty of the absolute frequency for the $2^3S_1 \rightarrow 2^1S_0$ transition of $^3$He $\frac{\Delta \omega}{\Delta \omega}$, which indicates the high-order ac Stark shift can be neglected for the precision spectroscopy measurement of the $2^3S_1 \rightarrow 2^1S_0$ transition of helium by implementation of a magic wavelength trap.

In summary, the improved RCI method enables us to calculate the dynamic dipole polarizabilities in wide range of laser frequency for both $2^3S_1$ and $2^1S_0$ states of helium. A series of magic wavelengths for $2^3S_1 \rightarrow 2^1S_0$ forbidden transition of $^4$He and $^3$He are accurately determined. The non-relativistic calculations of magic wavelength for $^\infty$He are also carried out by using the Hylleraas-B-spline method. The high-order ac Stark shift related with the dynamic hyperpolarizabilities are estimated. All the magic wavelengths from two different theoretical methods are consistent. Present work provides solid theoretical supports for experimental design of a magic wavelength trap to eliminate the ac Stark shift for the precision spectroscopy of the $2^3S_1 \rightarrow 2^1S_0$ transition of helium.

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TABLE V: The first nine magic wavelengths (in nm) of $^2\!\!S_0 \rightarrow ^2\!\!S_1$ transition of helium. The numbers in parentheses are computational uncertainties.

| Hylleraass-B-splines | RCI | Ref. [20] |
|----------------------|-----|-----------|
| $^\infty$He $^2\!\!S_0 \rightarrow ^2\!\!S_1$ | $^3$He $^2\!\!S_0 \rightarrow ^2\!\!S_1$ | $^3$He $^2\!\!S_0 \rightarrow ^2\!\!S_1$ |
| No. | | $^3$He | $^3$He |
| 1 | 412.16(2) | 412.167(1) | 412.168(1) |
| 2 | 352.399(4) | 352.335 7(1) | 352.335 4(1) |
| 3 | 338.641(2) | 338.683 7(1) | 338.699 2(1) |
| 4 | 331.240(1) | 331.300 07(2) | 331.300 20(2) |
| 5 | 326.633(1) | 326.789 49(2) | 326.789 31(2) |
| 6 | 323.544(1) | 323.589 72(2) | 323.589 84(2) |
| 7 | 321.366(1) | 321.411 36(2) | 321.411 40(2) |
| 8 | 319.771 1(1) | 319.816 07(2) | 319.816 09(2) |
| 9 | 318.566 8(1) | 318.611 63(5) | 318.611 63(5) |

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Supplemental Material for
Relativistic full-configuration-interaction calculations of magic wavelengths for the $2^3S_1 \rightarrow 2^1S_0$ transition of helium isotopes

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Table I - IV present the comparison of the energies of helium between present RCI calculations and Hylleraas calculations [1]. Table V - VI present the comparison of the reduced matrix elements between present RCI calculations and Hylleraas calculations [1]. All the values in these tables are obtained from one diagonalization of Hamiltonian. These results are presented as a benchmark for comparison with experiment and theory.

### TABLE I: Comparison of the energies (in a.u.) for $n^1S_0$ ($n \leq 13$) states of $^4$He and $^3$He. The numbers in parentheses are computational uncertainties.

| State  | $^4$He                  | $^3$He                  |
|--------|-------------------------|-------------------------|
|        | RCI                     | Hylleraas [1]           | RCI                     | Hylleraas [1]           |
| $1^1S_0$ | -2.903 35(2)           | -2.903 408 72           | -2.903 22(2)           | -2.903 271 37           |
|        | -2.903 398 137$^a$     |                         | -2.903 260 650$^a$     |                         |
| $2^1S_0$ | -2.145 783 5(2)        | -2.145 787 13           | -2.145 686 8(2)        | -2.145 690 46           |
|        | -2.145 786 084$^a$     |                         | -2.145 689 379$^a$     |                         |
| $3^1S_0$ | -2.061 095 2(2)        | -2.061 096 44           | -2.061 002 6(2)        | -2.061 003 88           |
| $4^1S_0$ | -2.033 414 13(2)       | -2.033 414 83           | -2.033 322 84(2)       | -2.033 323 58           |
| $5^1S_0$ | -2.021 006 09(2)       | -2.021 006 60           | -2.020 915 38(2)       | -2.020 915 93           |
| $6^1S_0$ | -2.014 393 29(2)       | -2.014 393 72           | -2.014 302 89(2)       | -2.014 303 36           |
| $7^1S_0$ | -2.010 456 53(2)       | -2.010 456 92           | -2.010 366 32(2)       | -2.010 366 75           |
| $8^1S_0$ | -2.007 924 74(2)       | -2.007 925 10           | -2.007 834 64(2)       | -2.007 835 05           |
| $9^1S_0$ | -2.006 200 91(2)       | -2.006 201 27           | -2.006 110 89(2)       | -2.006 111 29           |
| $10^1S_0$ | -2.004 974 52(2)      | -2.004 974 87           | -2.004 884 56(2)       | -2.004 884 94           |
| $11^1S_0$ | -2.004 071 07(2)      | -2.004 981 15(2)       | -2.003 981 15(2)       |                         |
| $12^1S_0$ | -2.003 386 38(4)      | -2.003 296 49(4)       | -2.003 296 49(4)       |                         |
| $13^1S_0$ | -2.002 855 12(4)      | -2.002 765 25(4)       | -2.002 765 25(4)       |                         |

$^a$ Both values are obtained by increasing the partial wave of $l_{max}$ to 20.

### TABLE II: The same as Table I but for $n^1P_1$ ($n \leq 13$) states of $^3$He.

| State  | RCI                     | Hylleraas [1]           |
|--------|-------------------------|-------------------------|
| $2^1P_1$ | -2.123 552 83(2)       | -2.123 557 20           |
| $3^1P_1$ | -2.054 875 71(2)       | -2.054 877 36           |
| $4^1P_1$ | -2.030 805 19(2)       | -2.030 806 11           |
| $5^1P_1$ | -2.019 644 21(2)       | -2.019 644 86           |
| $6^1P_1$ | -2.013 573 59(2)       | -2.013 574 20           |
| $7^1P_1$ | -2.009 909 74(2)       | -2.009 910 20           |
| $8^1P_1$ | -2.007 530 06(2)       | -2.007 530 50           |
| $9^1P_1$ | -2.005 897 67(2)       | -2.005 898 08           |
| $10^1P_1$ | -2.004 729 51(2)      | -2.004 729 91           |
| $11^1P_1$ | -2.003 864 90(2)      | -2.003 864 90(2)       |
| $12^1P_1$ | -2.003 207 09(4)      | -2.003 207 09(4)       |
| $13^1P_1$ | -2.002 695 04(4)      | -2.002 695 04(4)       |
TABLE III: The same as Table I but for \( n^3S_1 \) \((n \leq 13)\) states of \(^4\text{He}\) and \(^3\text{He}\).

| State  | \(^4\text{He}\) | \(^3\text{He}\) |
|--------|----------------|----------------|
|        | RCI            | Hylleraas [1]  | RCI            | Hylleraas [1]  |
| \(^2\text{He}\) | -2.175 045 25(2) | -2.175 045 67  | -2.174 947 30(2) | -2.174 947 79 |
| \(^3\text{He}\) | -2.068 514 02(2) | -2.068 514 37  | -2.068 421 12(2) | -2.068 421 51 |
| \(^4\text{He}\) | -2.036 340 19(2) | -2.036 340 52  | -2.036 248 79(2) | -2.036 249 16 |
| \(^5\text{He}\) | -2.022 448 45(2) | -2.022 448 78  | -2.022 357 69(2) | -2.022 358 06 |
| \(^6\text{He}\) | -2.015 207 84(2) | -2.015 208 17  | -2.015 117 41(2) | -2.015 117 78 |
| \(^7\text{He}\) | -2.010 960 80(2) | -2.010 961 12  | -2.010 870 56(2) | -2.010 870 92 |
| \(^8\text{He}\) | -2.008 258 32(2) | -2.008 258 64  | -2.008 168 21(2) | -2.008 168 57 |
| \(^9\text{He}\) | -2.006 432 93(2) | -2.006 433 26  | -2.006 342 90(2) | -2.006 343 26 |
| \(^10\text{He}\) | -2.005 142 37(2) | -2.005 142 69  | -2.005 052 40(2) | -2.005 052 76 |
| \(^11\text{He}\) | -2.004 196 39(2) |                    | -2.004 106 46(2) |                    |
| \(^12\text{He}\) | -2.003 482 41(4) |                    | -2.003 392 51(4) |                    |
| \(^13\text{He}\) | -2.002 930 32(4) |                    | -2.002 840 45(4) |                    |
TABLE IV: The same as Table I, for $n^3P_f$ ($n \leq 13$) states of $^4$He and $^3$He. The numbers in parentheses are computational uncertainties.

| State $| RCI | 4^He | Hylleraas \[1\] | RCI | 3^He | Hylleraas \[1\] |
|------|-----|------|--------------|-----|------|--------------|
| $2^3P_0$ | −2.132 980 94(2) | −2.132 981 43 | −2.132 888 13(2) | −2.132 888 66 |
| $3^3P_0$ | −2.057 906 35(2) | −2.057 906 72 | −2.057 814 84(2) | −2.057 815 25 |
| $4^3P_0$ | −2.032 152 61(2) | −2.032 152 95 | −2.032 061 77(2) | −2.032 062 15 |
| $5^3P_0$ | −2.020 380 85(2) | −2.020 381 18 | −2.020 290 37(2) | −2.020 290 74 |
| $6^3P_0$ | −2.014 038 40(2) | −2.014 038 73 | −2.013 948 13(2) | −2.013 948 41 |
| $7^3P_0$ | −2.010 235 87(2) | −2.010 236 20 | −2.010 145 73(2) | −2.010 146 10 |
| $8^3P_0$ | −2.007 778 23(2) | −2.007 778 56 | −2.007 688 19(2) | −2.007 688 55 |
| $9^3P_0$ | −2.006 098 70(2) | −2.006 099 02 | −2.006 008 72(2) | −2.006 009 08 |
| $10^3P_0$ | −2.004 900 39(2) | −2.004 900 71 | −2.004 810 45(2) | −2.004 810 82 |
| $11^3P_0$ | −2.004 015 60(2) | | −2.003 925 70(2) | | |
| $12^3P_0$ | −2.003 343 79(4) | | −2.003 253 92(4) | | |
| $13^3P_0$ | −2.002 821 71(4) | | −2.002 731 86(4) | | |
| | | | | | |
| $2^3P_1$ | −2.132 985 44(2) | −2.132 985 93 | −2.132 892 63(2) | −2.132 893 16 |
| $3^3P_1$ | −2.057 907 58(2) | −2.057 907 95 | −2.057 816 07(2) | −2.057 816 48 |
| $4^3P_1$ | −2.032 153 12(2) | −2.032 153 46 | −2.032 062 27(2) | −2.032 062 65 |
| $5^3P_1$ | −2.020 381 10(2) | −2.020 381 44 | −2.020 290 62(2) | −2.020 290 99 |
| $6^3P_1$ | −2.014 038 54(2) | −2.014 038 87 | −2.013 948 27(2) | −2.013 948 55 |
| $7^3P_1$ | −2.010 235 96(2) | −2.010 236 29 | −2.010 145 82(2) | −2.010 146 19 |
| $8^3P_1$ | −2.007 778 29(2) | −2.007 778 62 | −2.007 688 25(2) | −2.007 688 61 |
| $9^3P_1$ | −2.006 098 74(2) | −2.006 099 07 | −2.006 008 76(2) | −2.006 009 12 |
| $10^3P_1$ | −2.004 900 42(2) | −2.004 900 75 | −2.004 810 48(2) | −2.004 810 85 |
| $11^3P_1$ | −2.004 015 63(2) | | −2.003 925 72(2) | | |
| $12^3P_1$ | −2.003 343 81(4) | | −2.003 253 93(4) | | |
| $13^3P_1$ | −2.002 821 73(4) | | −2.002 731 87(4) | | |
TABLE V: Comparison of reduced matrix elements between present RCI calculations and Hylleraas calculations [2] of the $2^3S_1 \rightarrow n^3P_J (n \leq 13)$ transitions for helium. The numbers in parentheses are computational uncertainties.

| $n^3P_J$ | $^4He$ RCI | $^4He$ Hylleraas [2] | $n^3P_J$ | $^4He$ RCI | $^4He$ Hylleraas [2] |
|----------|-------------|---------------------|----------|-------------|---------------------|
| $2^3P_2$ | 5.660 70(2) | 5.660 906(3) & 5.660 605 & $8^3P_2$ | 0.205 91(2) & 0.205 938(2) & 0.205 885 |
| $2^3P_1$ | 4.384 77(2) | 4.384 920(2) & 4.384 692 & $8^3P_1$ | 0.159 50(2) & 0.159 517(2) & 0.159 476 |
| $2^3P_0$ | 2.531 58(2) | 2.531 673(2) & 2.531 375 & $8^3P_0$ | 0.092 08(2) & 0.092 087(2) & 0.092 073 |
| $3^3P_2$ | 1.173 17(2) | 1.173 398(2) & 1.172 934 & $9^3P_2$ | 0.170 76(2) & 0.170 785(2) & 0.170 742 |
| $3^3P_1$ | 0.908 71(2) | 0.908 893(2) & 0.908 546 & $9^3P_1$ | 0.132 27(2) & 0.132 288(2) & 0.132 254 |
| $3^3P_0$ | 0.524 54(2) | 0.524 644(2) & 0.524 549 & $9^3P_0$ | 0.076 36(2) & 0.076 368(2) & 0.076 356 |
| $4^3P_2$ | 0.671 56(2) | 0.671 667(2) & 0.671 450 & $10^3P_2$ | 0.144 70(2) & 0.144 713(2) & 0.144 674 |
| $4^3P_1$ | 0.520 18(2) | 0.520 264(2) & 0.520 103 & $10^3P_1$ | 0.112 08(2) & 0.112 093(2) & 0.112 064 |
| $4^3P_0$ | 0.300 34(2) | 0.300 331(2) & 0.300 281 & $10^3P_0$ | 0.064 70(2) & 0.064 710(2) & 0.064 701 |
| $5^3P_2$ | 0.449 40(2) | 0.449 465(2) & 0.449 332 & $11^3P_2$ | 0.124 71(2) & 0.124 720(2) |
| $5^3P_1$ | 0.348 10(2) | 0.348 149(2) & 0.348 050 & $11^3P_1$ | 0.096 59(2) & 0.096 607(2) |
| $5^3P_0$ | 0.200 99(2) | 0.200 979(2) & 0.200 944 & $11^3P_0$ | 0.055 76(2) & 0.055 770(2) |
| $6^3P_2$ | 0.329 31(2) | 0.329 356(2) & 0.329 269 & $12^3P_2$ | 0.108 97(2) & 0.108 978(2) |
| $6^3P_1$ | 0.255 08(2) | 0.255 115(2) & 0.255 047 & $12^3P_1$ | 0.084 40(2) & 0.084 413(2) |
| $6^3P_0$ | 0.147 28(2) | 0.147 273(2) & 0.147 252 & $12^3P_0$ | 0.048 72(2) & 0.048 731(2) |
| $7^3P_2$ | 0.255 40(2) | 0.255 437(2) & 0.255 365 & $13^3P_2$ | 0.096 30(2) & 0.096 313(2) |
| $7^3P_1$ | 0.197 83(2) | 0.197 858(2) & 0.197 806 & $13^3P_1$ | 0.074 59(2) & 0.074 603(2) |
| $7^3P_0$ | 0.114 23(2) | 0.114 220(2) & 0.114 201 & $13^3P_0$ | 0.043 06(2) & 0.043 067(2) |

TABLE VI: The same as Table V but for the $2^1S_0 \rightarrow n^3P_1$ and $2^3S_1 \rightarrow 1^1P_1 (n \leq 10)$ forbidden transitions of helium. The numbers in the square brackets denote powers of ten.

| $n^3P_1$ | $^4He$ RCI | $^4He$ Hylleraas [3] | $n^1P_1$ | $^4He$ RCI | $^4He$ Hylleraas [3] |
|----------|-------------|---------------------|----------|-------------|---------------------|
| $2^3P_1$ | 1.34(2)[-3] | 1.34(1)[-3] & 1.337[-3] & $2^1P_1$ | 1.266(3)[-3] & 1.268(2)[-3] & 1.262[-3] |
| $3^3P_1$ | 5.41(4)[-4] | 5.44(1)[-4] & 5.48[-4] & $3^1P_1$ | 1.21(2)[-4] & 1.22(2)[-4] & 1.21[-4] |
| $4^3P_1$ | 2.43(4)[-4] | 2.46(1)[-4] & $4^1P_1$ | 8.12(3)[-5] & 8.14(2)[-5] |
| $5^3P_1$ | 1.50(2)[-4] | 1.52(1)[-4] & $5^1P_1$ | 5.72(3)[-5] & 5.70(2)[-5] |
| $6^3P_1$ | 1.06(2)[-4] | 1.07(1)[-4] & $6^1P_1$ | 4.28(2)[-5] & 4.26(2)[-5] |
| $7^3P_1$ | 8.0(2)[-5] | 8.12(2)[-5] & $7^1P_1$ | 3.35(2)[-5] & 3.34(2)[-5] |
| $8^3P_1$ | 6.3(2)[-5] | 6.46(2)[-5] & $8^1P_1$ | 2.71(2)[-5] & 2.72(2)[-5] |
| $9^3P_1$ | 5.2(2)[-5] | 5.31(2)[-5] & $9^1P_1$ | 2.27(2)[-5] & 2.26(2)[-5] |
| $10^3P_1$ | 4.3(2)[-5] | 4.47(2)[-5] & $10^1P_1$ | 1.94(3)[-5] & 1.92(2)[-5] |
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