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Theoretical calculations on lifetimes of the low-lying excited states in Lu^+

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The lifetime of the 5d6s $^3D_1$ clock state in Lu^+ exhibits a large discrepancy between experimental and theoretical values. To resolve this discrepancy, we perform calculations of the magnetic dipole transition rate between the 5d6s $^3D_1$ and $6s^2$ $^1S_0$ states using the multi-configuration Dirac–Hartree–Fock method. The effects of electron correlations, Breit interaction, and quantum electrodynamics (QED) corrections on the transition parameters are analyzed systematically. The calculated 5d6s $^3D_1$–$6s^2$ $^1S_0$ magnetic dipole transition rate, $1.69(7) \times 10^{-6}$ s^{-1}, shows excellent agreement with the experimental measurement. To accurately determine the lifetime of the $^3D_1$ clock state, the hyperfine-induced electric quadrupole transition rate between the $^3D_1$ and ground states is also calculated. Furthermore, the rates of various transitions between states in the 5d6s configuration are obtained. The lifetimes of the $^3D_{2,3}$ and $^1D_2$ states are consistent with previous theoretical calculations.

Keywords: Lu^+ ion, magnetic dipole transition, electric quadrupole transition, hyperfine-induced interaction, lifetime

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1. Introduction

Optical atomic clocks have advanced significantly over the past decade, with several systems now achieving fractional frequency uncertainties approaching 10^{-18} [1–6] or better.[7–9] Among these optical clock candidates, singly ionized lutetium is unique since it has many interesting properties. Firstly, it offers three possible clock transitions in a single atomic system.[10] The large atomic mass of lutetium and additional clock transitions allow micromotion shifts to be controlled below 10^{-19}.[7] Secondly, the 5d6s $^3D_1$–6s6p $^3P_0$ cooling transition provides a fractional second-order Doppler shift below 10^{-19}.[11] Thirdly, the blackbody radiation shift of the $6s^2$ $^1S_0$–5d6s $^3D_1$ transition is $-1.36(10) \times 10^{-19}$ at 300 K, which is the lowest of any optical clock system[10] and can be controlled to less than 10^{-19} with modest technical effort. Moreover, Tan *et al.*[12] and Kaewuam *et al.*[13] have demonstrated the potential for Lu^+ clock operation with multiple ions, which can improve the stability of the ionic optical clock. Owing to these advantageous properties, Lu^+ has emerged as a leading candidate for next-generation optical clocks with potential applications in tests of relativity,[14,15] dark matter searches,[16,17] and quantum information processing.[18,19]

However, the lifetime of the 5d6s $^3D_1$ metastable state in Lu^+ remains ambiguous. Experimentally, Arnold *et al.*[20] measured the coupling rate of the 5d6s $^3D_1$–$6s^2$ $^1S_0$ M1 transition and obtained a lifetime of 174^{+23}_{-32} hours for the $^3D_1$ state. In theory, Paez *et al.*[21] calculated the M1 transition rate using the configuration interaction combined with the linearized coupled-cluster (CI+all-order) approach and obtained a lifetime of 54.04 hours. The theoretical value was about one-third of the experimentally determined lifetime. Furthermore, the hyperfine interaction induces mixing of the $^3D_1$ state with the $^3D_2$ and $^1D_2$ states and opens an additional transition channel between the $^3D_1$ and $^1S_0$ states. However, the effect of the hyperfine-induced transition on the lifetime of the $^3D_1$ state remains undetermined. In addition, Paez *et al.*[21] measured the lifetimes of the 5d6s $^{1,3}D_2$ states to be 0.19(0.4) s and 17.33(98) s, respectively. Theoretically, they also calculated the lifetimes of the 5d6s $^{1,3}D_2$ and $^3D_3$ states to be 0.23 s, 20.90 s, and 10.8 s using the CI+all-order method. There was more than a 20% discrepancy between the experimental and theoretical lifetimes for the 5d6s $^{1,3}D_2$ states in Lu^+. To resolve these large discrepancies, it is imperative to determine the lifetimes of states in the 5d6s configuration using another theoretical method.

In this work, we calculate the lifetime of the 5d6s $^3D_1$ state using the multi-configuration Dirac–Hartree–Fock (MCDHF) method. The effects of electron correlations, Breit

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interaction, and quantum electrodynamics (QED) corrections on the transition parameters are taken into account systematically using the active space approach. The rate of the 5d6s ^{3}D$_1$–6s^2 ^{1}S$_0$ M1 transition is determined, which shows good agreement with the experimental result. The hyperfine-induced 5d6s ^{3}D$_1$–6s^2 ^{1}S$_0$ E2 transition rate in ^{175}Lu$^+$ is also calculated. The influence of electron correlations on the hyperfine-induced transition (HIT) parameters is also analyzed. In addition, the rates of the M1 and E2 transitions between states in the 5d6s configuration are calculated to determine the lifetimes of the ^{3}D$_{2,3}$ and ^{1}D$_2$ states.

2. Theory

In the presence of hyperfine interactions, the electronic angular momentum J couples with the nuclear angular momentum I to form the total angular momentum F of the atomic system, and only F and the corresponding magnetic quantum number M_F are good quantum numbers. The wave function of an atomic state can be written as

$$|\Gamma F M_F\rangle = \sum_i d_i \,|\gamma_i J I F M_F\rangle. \tag{1}$$

The zeroth-order functions $|\gamma_i JIFM_F\rangle$ in the expansion are the coupled products of the electronic $|\gamma JM_J\rangle$ and the nuclear $|IM_I\rangle$ wave functions.

The hyperfine interaction induces mixing between atomic states with the same parity but different J and opens up new transition channels.[22–24] In the case of Lu$^+$, the hyperfine interaction induces mixing between the ^{3}D$_1$ state and other states, thereby opening an additional transition channel between the ^{3}D$_1$ and ^{1}S$_0$ states, referred to as the hyperfine-induced 5d6s ^{3}D$_1$–6s^2 ^{1}S$_0$ E2 transition. In this study, we only consider the effect of the adjacent ^{1,3}D$_2$ perturbing states, and we neglect interactions between the ^{3}D$_1$ state and other excited states due to the large energy separations and relatively weak hyperfine couplings. The ^{1}S$_0$ ground state is separated from other states by a large energy gap. The wave function of the ^{3}D$_1$ state can be represented by

$$\left|5\mathrm{d6s}\ {}^3\mathrm{D}_1 IF\right\rangle = d_0\left|5\mathrm{d6s}\ {}^3\mathrm{D}_1 IF\right\rangle + \sum_{S=1,3} d_S\left|5\mathrm{d6s}\ {}^S\mathrm{D}_2 IF\right\rangle. \tag{2}$$

The mixing coefficients d_S are obtained in the first-order perturbation theory as ratios between the hyperfine matrix elements and the unperturbed energy differences

$$d_S = \frac{\left\langle 5\mathrm{d6s}\ {}^S\mathrm{D}_2 IF\right|H_{\mathrm{hfs}}\left|5\mathrm{d6s}\ {}^3\mathrm{D}_1 IF\right\rangle}{E\left({}^3\mathrm{D}_1\right) - E\left({}^S\mathrm{D}_2\right)}. \tag{3}$$

According to Racah theory, the off-diagonal matrix elements are[25]

$$\begin{aligned}&\left\langle \gamma\Gamma IJFM_F\right|\boldsymbol{T}^{(1)}\cdot\boldsymbol{M}^{(1)}\left|\gamma\Gamma' IJ'FM_F\right\rangle\\ &= (-1)^{I+J+F}\begin{Bmatrix} I & J & F\\ J' & I & 1\end{Bmatrix}\sqrt{2J+1}\sqrt{2I+1}\\ &\times\left\langle \Gamma J\right\|\boldsymbol{T}^{(1)}\left\|\Gamma' J'\right\rangle\left\langle \gamma I\right\|\boldsymbol{M}^{(1)}\left\|\gamma I\right\rangle,\end{aligned} \tag{4}$$

where $J' = J-1$, J, or $J+1$, and

$$\begin{aligned}&\left\langle \gamma\Gamma IJFM_F\right|\boldsymbol{T}^{(2)}\cdot\boldsymbol{M}^{(2)}\left|\gamma\Gamma' IJ'FM_F\right\rangle\\ &= (-1)^{I+J+F}\begin{Bmatrix} I & J & F\\ J' & I & 2\end{Bmatrix}\sqrt{2J+1}\sqrt{2I+1}\\ &\times\left\langle \Gamma J\right\|\boldsymbol{T}^{(2)}\left\|\Gamma' J'\right\rangle\left\langle \gamma I\right\|\boldsymbol{M}^{(2)}\left\|\gamma I\right\rangle,\end{aligned} \tag{5}$$

where $J' = J-2$, $J-1$, J, or $J+1$ for $J = 2$.

The HIT rate can be estimated by[26]

$$\begin{aligned}A\left({}^3\mathrm{D}_1\mathrm{F} - {}^1\mathrm{S}_0\right) &= \frac{1.120\times10^{18}}{3\lambda^5}\times\left|d_1\left\langle {}^1\mathrm{D}_2\right\|\mathrm{E2}\left\|\ {}^1\mathrm{S}_0\right\rangle\right.\\ &\left.+d_3\left\langle {}^3\mathrm{D}_2\right\|\mathrm{E2}\left\|\ {}^1\mathrm{S}_0\right\rangle\right|^2,\end{aligned} \tag{6}$$

where λ is the HIT transition wavelength in Å, $\left\langle {}^1\mathrm{D}_2\right\|\mathrm{E2}\left\|\ {}^1\mathrm{S}_0\right\rangle$ and $\left\langle {}^3\mathrm{D}_2\right\|\mathrm{E2}\left\|\ {}^1\mathrm{S}_0\right\rangle$ are the E2 reduced transition matrix elements between the ^{1,3}D$_2$ perturbing states and the ground state.

According to the MCDHF method,[27–29] the atomic state wave function $\Psi(\Gamma PJM_J)$ is a linear combination of configuration state functions (CSFs) $\Phi_j(\gamma_j PJM_J)$ with the same parity P, total angular momentum J, and magnetic quantum number M_J

$$\Psi\left(\Gamma PJM_J\right) = \sum_i^{N_{\mathrm{CSFs}}} c_i\Phi_i(\gamma_i PJM_J), \tag{7}$$

where c_i are the configuration mixing coefficients, γ_i represents the other quantum numbers that define the state uniquely, Γ is the identifying label containing all additional information necessary to uniquely describe the state function, and N_{CSFs} is the number of CSFs. The CSFs are built from sums of products of one-electron Dirac orbitals. In the self-consistent field (SCF) procedure, the CSFs are systematically enlarged[28] to yield increasingly accurate wave functions. This involves optimizing the coefficients c_i and the radial functions of the one-electron Dirac orbitals to minimize the energies concerned. Following the SCF optimization step, a relativistic configuration interaction (RCI) calculation is conducted to include core-core correlation, higher-order correlation, Breit interaction, and QED correction, with only the expansion coefficients being optimized. This approach ensures a rigorous and comprehensive treatment of electron correlations and relativistic effects.

Once the wave functions of the atomic states are obtained, the transition rate can be evaluated using the reduced matrix element $\left\langle \Psi\left(\gamma J\right)\right\|\mathrm{O}^{(\lambda)}\left\|\Psi\left(\gamma' J'\right)\right\rangle$, where the tensor operator $\mathrm{O}^{(\lambda)}$ is a multipole radiation field operator. The biorthogonal transformation technique[30,31] is applied to compute the

reduced matrix elements when the ground and excited state wave functions are built from independently optimized orbital sets.

3. Computational model

The electron correlations are systematically considered within the framework of the MCDHF method by employing the active space approach.[28,32] According to perturbation theory, electron correlations can be divided into first-order and higher-order contributions.[33,34] The first-order electron correlation effects, composed of valence–valence (VV), core–valence (CV), and core–core (CC) correlations, are captured by the CSFs generated through single (S) and double (D) excitations from the occupied orbitals in the single-reference configuration set. The higher-order correlations can be described by incorporating the CSFs resulting from SD replacements of multi-reference (MR) configurations.[29,35] In this approach, the CSFs with large mixing coefficients in the first-order correlation are selected to form the MR configuration set.

The ground-state configuration [Xe]$4f^{14}6s^2$ and the excited-state configuration [Xe]$4f^{14}5d6s$ are adopted as the reference configurations and optimized independently in our calculations. Electrons from the outermost 5d and 6s shells are treated as valence electrons, while the remaining electrons are treated as core electrons. Our calculations start from the Dirac–Hartree–Fock (DHF) approximation to optimize the spectroscopic orbitals occupied in the reference configurations. The VV and CV correlations between valence and core electrons are considered by SD excitations from the reference configurations to an active set of orbitals in the SCF computational procedure. The SD excitations are restricted such that at most one electron in a core subshell can be replaced with an active orbital. The occupied shells are opened successively for substitutions into the virtual set, beginning with 5sp, followed by 4spdf. Meanwhile, the virtual orbitals (VOs) are added layer by layer, and only newly added virtual orbitals are varied. We augment eight layers of VOs to fully account for VV and CV correlations and ensure the convergence of the parameters under investigation. Each computational model is labeled C*m*V-*l*, where '*m*' denotes the latest opened core subshell and '*l*' is the number of VO layers. The CSFs that do not interact with the reference configurations are removed to improve computational efficiency.[36] The orbital set obtained in the C4V-8 model is used for subsequent RCI calculations.

The CC electron correlation is estimated with the CSFs generated by SD excitations from the $n = 5$ core shell to seven layers of virtual orbitals; this model is denoted CC5-7. The SD replacements of the MR method are applied to estimate the higher-order electron correlation effects among the $n = 5$ and 6 shells. The MR configurations of the ground state include $5s^25p^66p^2$; $5s^25p^45d^26s^2$; $5s^25p^55d6s6p$; $5s^25p^66s7s$; $5s^25p^65d^2$; $5s^25p^65d6d$; $5s^25p^66p7p$; and $5s^25p^45d6s^26d$, while those of the excited state are $5s^25p^65d7s$; $5s^25p^65d^2$; and $5s^25p^66p^2$. SD excitations are allowed from the MR configurations to four layers of virtual orbitals for the ground state and three layers of virtual orbitals for the excited states. Finally, the Breit interaction and QED correction are evaluated based on the MR model in the RCI method. The GRASP2018 package[37] is employed for all calculations. The `rhfszeeman` program[23] is used to calculate the off-diagonal hyperfine interaction matrix elements, and the `transition-phase` program[23] is used to compute the electric quadrupole transition matrix element.

4. Results and discussion

4.1. Lifetime of the 5d6s $^3D_1$ clock state

4.1.1. The 5d6s $^3D_1$–6s^2 $^1S_0$ M1 transition

Table 1 displays the transition energy (ΔE), reduced transition matrix element (RME), and transition rate (A) of the 5d6s $^3D_1$–6s^2 $^1S_0$ M1 transition as functions of the computational models. It is found that VV and CV correlations have converged when eight layers of virtual orbitals are added in the SCF calculations. These correlations are the main factors influencing the transition parameters, contributing approximately 18.0% to the transition energy and 96.3% to the reduced transition matrix element. The CC correlation increases the transition energy by 15.4% but decreases the reduced matrix element by 25.3%. The higher-order correlation contributes 27.4% to the transition energy and 12.9% to the matrix element. The combined contributions of the Breit interaction and QED correction are less than 3%, but they are essential for obtaining reliable transition parameters. Generally, the uncertainty of the calculations mainly arises from electron correlations neglected in the computational models. In this work, we systematically include the contributions from VV correlation, CV correlation with $n \geq 4$, CC correlation with $n \geq 5$, and the main part of the higher-order correlation. The uncertainties during the convergence process are smaller than 0.1%. It is worth noting that the effect of the $n = 5$ CC correlation on the reduced matrix element counteracts the higher-order correlation. We speculate that a similar cancellation occurs between the CC4 correlation and its associated higher-order effects. Consequently, the uncertainty due to neglecting CC4 and the corresponding higher-order correlations is smaller than the isolated CC4 correlation contribution, which is estimated to be about 3.9%.

In Table 1, previous theoretical and experimental values are also listed for comparison. For the transition energy, the Fock-space coupled cluster (FSCC) results of Dzuba *et al.*[38] and Kahl *et al.*[39] exceed the National Institute of Standards

and Technology (NIST) value[40] by 1.3% and 4.7%, respectively. The configuration interaction plus many-body perturbation theory (CI + MBPT) calculation by Kozlov *et al.*[41] yields a value 1.7% higher than the NIST value, while the result of Kahl *et al.*[39] using the same method is 1.1% below the NIST value. However, none of these studies include higher-order electron correlations. Our result is 5.7% lower than the NIST value. This larger deviation may arise from CC and higher-order electron correlations related to the 4f core shell, which dramatically increase the number of CSFs and exceed our computational capacity. Regarding the matrix element, our value is 43.7% smaller than the theoretical result of Paez *et al.*,[21] who used the CI + all-order method. For the transition rate, the experimental measurement[20] is about one-third of the theoretical prediction.[21] To obtain a more accurate result, we revise the transition rate by employing the NIST excitation energy. Our recalculated transition rate shows excellent agreement with the experimental value of Arnold *et al.*,[20] who observed the coupling rate of the ^{3}D$_1$, $F = 9/2$, $M_F = 9/2$ to ^{1}S$_0$, $F' = 7/2$, $M'_F = 7/2$ clock transition and measured the intensity of the clock laser operating on the ion. This resolves the large discrepancy mentioned above and demonstrates the reliability and precision of our computational approach.

Table 1. The transition energy ΔE (in cm^{-1}), reduced matrix element RME (in a.u.), and transition rate A (in s^{-1}) for the ^{3}D$_1$–^{1}S$_0$ M1 transition. Row 14, labeled as 'Resived', represents the transition rate calculated using the transition energy of the NIST.[40] Numbers in square brackets stand for the power of 10 and in parentheses for error bars.

Model	ΔE	RME	A
DHF	8288.41	1.37[−5]	9.67[−10]
VV-1	13264.10	6.83[−5]	9.79[−8]
C5V-2	9404.28	3.93[−4]	1.16[−6]
C4V-3	7231.64	3.74[−4]	4.76[−7]
C4V-4	7125.12	3.76[−4]	4.60[−7]
C4V-5	7055.25	3.75[−4]	4.43[−7]
C4V-6	7028.89	3.74[−4]	4.36[−7]
C4V-7	7025.06	3.74[−4]	4.36[−7]
C4V-8	7025.34	3.74[−4]	4.36[−7]
CC5-7	8306.81	2.98[−4]	4.59[−7]
MR	11434.22	3.43[−4]	1.58[−6]
MR + B	11258.50	3.38[−4]	1.47[−6]
MR + BQ	11119.16	3.38[−4]	1.42[−6]
Revised			1.69(7)[−6]
Theories			
Paez *et al.*[21]		6.0[−4]	5.14[−6]
Dzuba *et al.*[38]	11948		
Kozlov *et al.*[41]	11995		
[a]Kahl *et al.*[39]	12354		
[b]Kahl *et al.*[39]	11664		
Experiments			
NIST[40]	11796.24		
Arnold *et al.*[20]			1.60(15)[−6]

[a] Fock space coupled cluster (FSCC) method. [b] Configuration interaction with many-body perturbation theory (CI + MBPT).

5. Hyperfine-induced 5d6s^{3}D$_1$6s^2 ^{1}S$_0$ E2 transition

The excitation energies, reduced transition matrix elements, and the rates of the 5d6s ^{1,3}D$_2$–6s^2 ^{1}S$_0$ E2 transitions are listed in Table 2. It is found from Table 2 that the VV and CV correlations make dominant contributions to all physical quantities under investigation. The higher-order correlation effect also counteracts the CC correlation effect. The total contributions of the Breit interaction and QED corrections are within 2.8% for the transition energies and 1.5% for the transition matrix elements. For the ^{1}D$_2$–^{1}S$_0$ transition energy, the previous theoretical calculations[38,39] are 3% larger than the NIST data, while our result is 2.6% less than the NIST data. For the ^{3}D$_2$–^{1}S$_0$ transition energy, Kahl's[39] result using the CI + MBPT method is below the NIST values by 0.4%, whereas the FSCC theoretical predictions[38,39] exceed the NIST data by over 2%. Our calculation is below the NIST values by 5%. To obtain more precise transition rates, we use the energy gaps from NIST and revise the rates of ^{1,3}D$_2$–^{1}S$_0$ transition, as detailed in the "Revised" row of Table 2. For the reduced matrix elements and rates of ^{1,3}D$_2$–^{1}S$_0$ transition, our results closely align with Paez *et al.*'s calculations.[21] However, there are over 20% differences between the theoretical transition rates and the corresponding experimental values[21] obtained using the optical pumping and fluorescence monitoring method.

The hyperfine mixing coefficient (d_1 and d_3) can be calculated from the ratio of the off-diagonal hyperfine interaction matrix element to the energy gap between the ^{3}D$_1$ state and perturbing states. The off-diagonal hyperfine interaction matrix elements include magnetic dipole and electric quadrupole matrix elements, as shown in Table 3. It is found from Table 3 that VV and CV correlations have the main influence on both the energy gaps and matrix elements. The effect of CC correlation on the off-diagonal matrix element is also offset by the higher-order correlation. The Breit interaction and QED correction contribute less than 1.7%. The off-diagonal matrix elements we calculated are roughly consistent with the results of Paez *et al.*,[21] especially for the magnetic dipole hyperfine interaction matrix element of the ^{3}D$_2$ and ^{3}D$_1$ states. The F-dependent HIT rates are calculated and presented in columns 8–10 of Table 3. The total HIT rate is determined to be 1.40×10^{-8} s^{-1} by means of a weighted average ($A_{\rm HIT} = \frac{\sum_{F_i}(2F_i+1)A_{F_i}}{\sum_{F_i}(2F_i+1)}$), which is two orders of magnitude smaller than the ^{3}D$_1$–^{1}S$_0$ M1 transition rate. Therefore, the hyperfine interactions can be neglected to determine the lifetime of the ^{3}D$_1$ clock state.

Table 2. The transition energies ΔE (in cm^{-1}), reduced matrix elements RME (in a.u.), and transition rates A (in s^{-1}) under different gauges for the $^3D_2$–$^1S_0$ E2 transitions. Here, 'C' in parentheses denotes Coulomb gauges while 'B' represents Babushkin gauges. Numbers in square brackets stand for the power of 10.

Model	$^1D_2$–$^1S_0$					$^3D_2$–$^1S_0$				
	ΔE (C)	RME (C)	RME (B)	A (C)	A (B)	ΔE (B)	RME (C)	RME (B)	A (C)	A (B)
DHF	17993.43	10.72	12.72	4.85	6.84	8874.63	0.20	1.06	5.14[−5]	1.37[−3]
VV-1	17720.69	11.71	11.68	5.37	5.34	13708.95	2.33	2.57	5.91[−2]	7.14[−2]
C5V-2	14826.61	10.46	10.70	1.76	1.84	9970.30	2.16	2.35	1.03[−2]	1.22[−2]
C4V-3	12947.73	8.69	9.93	0.61	0.80	7863.68	1.70	2.43	1.96[−3]	3.96[−3]
C4V-4	12854.15	8.51	9.88	0.57	0.77	7760.03	1.65	2.44	1.72[−3]	3.74[−3]
C4V-5	12781.86	8.42	9.85	0.54	0.74	7690.01	1.62	2.44	1.58[−3]	3.58[−3]
C4V-6	12755.31	8.37	9.84	0.53	0.73	7663.65	1.60	2.44	1.51[−3]	3.51[−3]
C4V-7	12750.35	8.35	9.82	0.53	0.73	7659.55	1.59	2.44	1.49[−3]	3.50[−3]
C4V-8	12750.30	8.32	9.82	0.52	0.73	7659.78	1.57	2.43	1.46[−3]	3.50[−3]
CC5-7	15752.67	9.45	11.09	1.94	2.67	8970.79	1.41	2.01	2.57[−3]	5.26[−3]
MR	17238.30	9.03	10.33	2.78	3.64	12107.00	1.96	2.49	2.25[−2]	3.62[−2]
MR + B	17015.92	9.01	10.32	2.59	3.40	11922.18	1.92	2.45	1.99[−2]	3.25[−2]
MR + BQ	16871.78	9.11	10.31	2.54	3.26	11783.18	1.96	2.46	1.95[−2]	3.08[−2]
Revised					3.73					4.01[−2]
					Theories					
Paez *et al.*[21]		10.59		3.96			2.51		4.19[−2]	
Dzuba *et al.*[38]	17892					12695				
[a]Kahl *et al.*[39]	17892					12985				
[b]Kahl *et al.*[39]	17875					12380				
					Experiments					
NIST[40]	17332.58					12435.32				
Paez *et al.*[21]				5.20(12)					5.19(33)[−2]	

[a]Fock space coupled cluster (FSCC) method. [b]Configuration interaction with many-body perturbation theory (CI + MBPT).

Table 3. The energy gaps (in a.u.), off-diagonal hyperfine interaction matrix elements (in a.u.) and HIT rates (s^{-1}) for $^{175}Lu^+$. RME_{T_1} and RME_{T_2} represent the magnetic dipole interaction and electric quadrupole matrix element, respectively. Numbers in square brackets stand for the power of 10 and in parentheses for error bars.

Model	($^1D_2$, $^3D_1$)			($^3D_2$, $^3D_1$)			A_{F_i}		
	ΔE	RME_{T_1}	RME_{T_2}	ΔE	RME_{T_1}	RME_{T_2}	$F_i = 5/2$	$F_i = 7/2$	$F_i = 9/2$
DHF	−4.42[−2]	3.17[−1]	3.18[−2]	−2.67[−3]	−4.65[−1]	1.25	1.43[−10]	2.16[−10]	1.65[−10]
C4V-8	−2.61[−2]	3.40[−1]	2.21[−1]	−2.89[−3]	−6.75[−1]	1.61	1.30[−9]	2.09[−9]	1.75[−9]
CC5-7	−3.39[−2]	3.62[−1]	1.92[−1]	−3.03[−3]	−6.02[−1]	1.66	1.27[−9]	1.99[−9]	1.61[−9]
MR	−2.65[−2]	3.28[−1]	2.42[−1]	−3.07[−3]	−6.49[−1]	1.65	1.23[−8]	1.96[−8]	1.62[−8]
MR + B	−2.62[−2]	3.28[−1]	2.37[−1]	−3.02[−3]	−6.43[−1]	1.65	1.11[−8]	1.76[−8]	1.45[−8]
MR + BQ	−2.62[−2]	3.29[−1]	2.38[−1]	−3.03[−3]	−6.45[−1]	1.65	1.06[−8]	1.68[−8]	1.38[−8]
Paez *et al.*[21]		3.63(30)[−1]	1.33(86)[−1]		−6.39(67)[−1]	1.33(19)			
NIST[40]	−2.52[−2]			−2.91[−3]					

6. Lifetimes of 5d6s $^3D_{2,3}$ and $^1D_2$ states

In Table 4, we present the calculated M1 and E2 transition rates between states in the 5d6s configuration to determine the lifetimes of $^3D_{2,3}$ and $^1D_2$ states. Other theoretical and experimental results are also shown for comparison. For the M1 transitions, the agreements in both the reduced matrix elements and transition rates between our results and other theoretical values[21] are within 1.8%. For the E2 transitions, our calculated reduced transition matrix elements show discrepancies of 0.9%–3.7% compared with previous theoretical results.[21] Combining the rates of $^{1,3}D_2$–$^1S_0$ E2 transition from Table 2, we obtain lifetimes of the $^3D_{2,3}$ and $^1D_2$ states in the 5d6s configuration by summing over the transition rates of the important transition channels ($\tau = 1/\sum_j A_j$). For the $^3D_2$ state, the transition rates of the $^3D_2$–$^1S_0$ E2 and $^3D_2$–$^3D_1$ M1 transitions are more than five orders of magnitude larger than that of the $^3D_2$–$^3D_1$ E2 transition. Thus, $^3D_2$–$^1S_0$ E2 and $^3D_2$–$^3D_1$ M1 transitions dominate the lifetime of the $^3D_2$ state. Our result for the $^3D_2$ state is 21.70 s, which is about 3.8% larger than the theoretical value.[21] For the $^3D_3$ state, the $^3D_3$–$^3D_2$ M1 transition is the primary transition channel. This yields a lifetime of 10.79 s, which is well consistent with the previous theoretical result.[21] For the $^1D_2$ state, the dominant transition is the $^1D_2$–$^1S_0$ E2 transition, but the $^1D_2$–$^3D_1$ M1 transitions cannot be ignored, resulting in a lifetime of 0.25 s. There are discrepancies exceeding 20% between the theoretical and experimental results[21] for the $^{1,3}D_2$ states. We attribute this discrepancy to two possible reasons: (i) Off-resonant scattering induced

by the cooling, repumping, or detection beam. (ii) Statistical uncertainties arising from longer data collection time of the metastable state. We call for more theoretical and experimental studies on determining the lifetimes of the $^{1,3}D_2$ states.

Table 4. The reduced matrix elements RME (in a.u.) and transition rates A (in s^{-1}) of M1 and E2 transitions between states in 5d6s configuration. The theoretical and experimental lifetimes τ (in s) of $^3D_{2,3}$ and $^1D_2$ states are presented in 8–10 columns. RMEs of the Babushkin gauge in the MR + BQ model are taken as the final results for the E2 transitions. Our transition rates are calculated by applying the excitation energies from the NIST database.[40] Numbers in square brackets stand for the power of 10 and in parentheses for error bars.

Upper	Lower	Transition	RME		A		τ		
			This work	Theo.[21]	This work	Theo.[21]	This work	Theo.[21]	Exp.[21]
$^3D_2$	$^3D_1$	E2	4.36	5.52	4.55[−8]	4.88[−8]	21.7	20.9	17.33(98)
	$^3D_1$	M1	2.06	2.06	5.95[−3]	5.94[−3]			
$^3D_3$	$^3D_1$	E2	1.54	1.60	3.05[−6]	3.29[−6]	10.79	10.8	
	$^3D_2$	E2	4.79	4.97	6.27[−6]	6.74[−6]			
	$^3D_2$	M1	2.09	2.09	9.26[−2]	9.27[−2]			
$^1D_2$	$^3D_1$	E2	0.99	1.02	1.13[−4]	1.21[−4]	0.25	0.23	0.19(0.4)
	$^3D_2$	E2	1.33	1.32	1.12[−4]	1.10[−4]			
	$^3D_3$	E2	1.28	1.33	1.10[−5]	1.19[−5]			
	$^3D_1$	M1	0.53	0.52	2.52[−1]	2.51[−1]			
	$^3D_2$	M1	0.22	0.22	3.03[−2]	3.01[−2]			
	$^3D_3$	M1	0.54	0.53	4.76[−2]	4.68[−2]			

7. Conclusion

In this work, we calculated the lifetimes of four low-lying excited states in Lu^+ using the MCDHF method. The electron correlation effects were systematically included using the active space approach. The effects of the Breit interaction and QED correction on the transition properties were also evaluated. Our calculated rate of the 5d6s $^3D_1$–6s^2 $^1S_0$ M1 transition was $1.69(7) \times 10^{-6}$ s^{-1}. Furthermore, we calculated the rate of the hyperfine-induced 5d6s $^3D_1$–6s^2 $^1S_0$ E2 transition in $^{175}Lu^+$. The contribution of the hyperfine interaction to the lifetime of the $^3D_1$ state was found to be negligible for determining the state lifetime. Our final lifetime of the $^3D_1$ clock state was 164.4(66) hours, falling within the uncertainty range of the latest experimental measurement. This resolves the previously reported discrepancy between theoretical and experimental results. Moreover, the lifetimes of the $^3D_{2,3}$ and $^1D_2$ states were determined to be 21.70 s, 10.79 s, and 0.25 s, respectively, which are in good agreement with previous theoretical results. However, significant discrepancies still remain between the theoretical and experimental lifetimes of the $^{1,3}D_2$ states. Our results have important implications for understanding the atomic structure of Lu^+ and provide reliable theoretical support for the design and optimization of the Lu^+ optical atomic clock.

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