Relativistic configuration interaction plus linearized-coupled-cluster calculations of U²⁺ energies, *g* factors, transition rates, and lifetimes

I. Savukov,[1] U. I. Safronova,[2] and M. S. Safronova[3,4]

[1]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

[2]*Physics Department, University of Nevada, Reno, Nevada 89557, USA*

[3]*Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA*

[4]*Joint Quantum Institute, NIST and the University of Maryland, College Park, Maryland 20742, USA*

Excitation energies, term designations, *g* factors, transition rates, and lifetimes of U²⁺ are determined using a relativistic configuration interaction (CI) + linearized-coupled-cluster (LCC) approach. The CI-LCC energies are compared with CI + many-body-perturbation-theory (MBPT) and available experimental energies. Close agreement has been found with experiment, within hundreds of cm⁻¹. In addition, lifetimes of higher levels have been calculated for comparison with three experimentally measured lifetimes, and close agreement has been found within the experimental error. CI-LCC calculations constitute a benchmark test of the CI + all-order method in complex relativistic systems such as actinides and their ions with many valence electrons. The theory yields many energy levels, *g* factors, transition rates, and lifetimes of U²⁺ that are not available from experiment. The theory can be applied to other multivalence atoms and ions, which would be of interest to many applications.

PACS number(s): 31.15.ac, 31.15.ag, 31.15.aj

I. INTRODUCTION

Atomic properties of actinides, such as energy levels, are needed in many applications, from nuclear forensic to industrial uses to quantum chemistry calculations. In particular, energies of actinide ions are needed for calibration of model potentials in chemical calculations of molecules containing actinide atoms used to reduce large full Hilbert space [1,2]. In addition to being of practical interest, actinides are an intriguing research subject, since they are considered among the most complex atoms that pose several challenges. First, relativistic effects are important and have to be treated consistently. Second, actinides have many valence electrons, including those in the *f* shell, that generate a very large number of possible states, and valence-valence interactions between these states have to be treated in all orders, for example with the configuration-interaction (CI) method. Finally, valence-core interactions are also strong, as will be evident below from differences between experimental and 2nd-order many-body perturbation theory (MBPT) single-electron energies in U⁵⁺ ion, and the CI + 2nd-order MBPT approach can be inaccurate. Thus more elaborate approaches, such as CI + all-order, may be required. The study of the U²⁺ ion is important for developing theory for more complex actinide ions since the valence CI space contains only configurations with four elections in our method and can still be saturated. From the point of view of testing the theory, many experimental U²⁺ energy levels are available to gauge the precision of the theory; in addition, substantial gaps in experimental data exist that can be filled with the theoretical calculations, provided theory is proved to be satisfactory.

Because of the aforementioned challenges, there were no reliable *ab initio* or semiempirical calculations of U²⁺ energy levels reported in the literature. An early attempt to estimate a few energy levels was made by Brewer [3] using trends in energies of different actinide atoms. It was estimated that the ground state was the odd $5f^3 6d\ ^5L_6$ and the first even state $5f^4\ ^5I_4$ had energy 1000 ± 1000 cm⁻¹ from the ground

state. Considering the uncertainty, either of these states could have been the ground state. Palmer and Engleman [4] used the predictions by Brewer [3] to assign labels to two of the lowest states: $5f^3 6d\ (^4I)^5L_6$ to the ground state and $5f^3 6d\ (^4I)^5K_5$ to the next odd state. Experimentally many actinide ions are difficult to deal with and the available data are generally limited to only energy levels. Few data for transition rates or lifetimes are available for actinides. Spectroscopic measurements of lines in discharges, where different stages of ionicity coexist and many levels are simultaneously excited, were converted to energy levels using a fitting procedure. The assignment of labels was done using a parametric method following the Slater-Condon method [5,6]. Apart from the problem of level identification, some effort was focused on calculations of ionization potentials with the approach of model potentials and pseudopotentials [7,8], which are widely used in quantum chemistry. As it is evident from the literature search, data for multiple-charge actinide ions is scarce, and there is a great need for developing an *ab initio* approach, such as described in this paper.

Recently, atomic properties of the neutral thorium and its ions were evaluated by Safronova *et al.* [9]. Excitation energies, term designations, and *g* factors of Th, Th⁺, and Th²⁺ were determined using a relativistic hybrid CI + linearized-coupled-cluster methods (LCC) [10]. The results were compared with other theoretical and experimental values where available. The neutral Th and Th-like uranium have similar electronic structure: a radon core ([Xe]$4f^{14}5d^{10}6s^2 6p^6$ where [Xe] = $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^6$) and four valence electrons. The ground state of neutral thorium is $6s^2 7s^2\ ^3F_2$; its experimental energies are compiled in the recently updated website [11]. In the case of Th-like uranium, U²⁺, the experimental data are quite old and less complete [4,12]. The $5f^4\ ^5F_4$ level was determined to be the ground state of U²⁺ [12], but level identification was not presented and only odd-parity states were referenced. The low-lying valence configurations of Th and Th-like U are very different, with

dominant even configurations being $6s^2 7s^2$ and $6d^3 7s$ in Th and $5f^4$ and $5f^2 6d^2$ in Th-like uranium. Large correlation effects for systems with nf electrons were discussed by Safronova *et al.* [13]. For example, the correction due to high partial waves is largely determined by a number of nf electrons in a configuration [13].

In the present paper, we evaluate energies of U^{2+} using the CI + LCC approach. The energies of odd- and even-parity complex states with $J = 0$–7 were evaluated. Each complex includes 12 states, which gives together 192 states. Electric-multipole matrix elements ($E1$, $E2$, and $E3$) and magnetic-multipole matrix elements ($M1$, $M2$, and $M3$) were calculated. We use these matrix elements to evaluate transition rates, oscillator strengths, and lifetimes.

II. CI + MPBT APPROACH

It is known that it is important to consider valence-valence interactions using the nonperturbative CI method, while weaker valence-core interactions can be included using pseudopotentials or many-body perturbation theory. Recently, we have studied the Si atom [14], which has corrections beyond the 2nd order quite small, much smaller than missing corrections from incomplete valence-valence CI space. However, in the current case of U^{2+}, it appears that the 2nd order is not sufficient, with errors from the omission of higher-order corrections on the order 1000 cm^{-1}. In the present study, we find that by scaling correlation corrections, especially the single-electron part with $l = 0$, much better agreement can be achieved. Thus we include CI-scaled MBPT energies for comparison with experiment and CI-LCC calculations.

A CI-MBPT method developed for open shell atoms with multiple valence electrons is used in the current calculations (see, for example, Ref. [15]). The effective CI-MBPT Hamiltonian for U^{2+} is split into two parts:

$$H^{\text{eff}} = \sum_{i=1}^{M} h_{1i} + \sum_{i \neq j}^{M} h_{2ij}. \quad (1)$$

The one-electron contribution

$$h_1 = c\alpha \cdot \mathbf{p} + (\beta - 1)mc^2 - Ze^2/r + V^{N-4} + \Sigma_1 \quad (2)$$

in addition to the V^{N-4} DHF potential contains the valence electron self-energy correction, Σ_1 [16]. In the current CI-MBPT program, the self-energy correction is calculated with the 2nd-order MBPT. The two electron Hamiltonian is

$$h_2 = e^2/|\mathbf{r}_1 - \mathbf{r}_2| + \Sigma_2, \quad (3)$$

where Σ_2 is the term accounting for Coulomb interaction screening arising from the presence of the core [17]. In the CI-MBPT calculations, the screening is calculated to the 2nd order.

To understand the valence-core effects, we compared monovalent U^{5+} energies calculated with the 2nd-order MBPT and the LCC method with experiment in Table I. As expected, the agreement with experiment is better for the LCC method. More specifically, the accuracy for the $7s$ and $7p$ states is worse than 1000 cm^{-1} in the case of MBPT, while the LCC method gives deviations less than 1000 cm^{-1}, except for the $7p_{3/2}$ state. Since low-lying U^{2+} levels do not contain substantial

TABLE I. Comparison of U^{5+} 2nd-order MBPT and LCC energy levels with theoretical [18] and experimental [12] results.

Level	Expt.	2nd-MBPT	Diff.	LCC	Diff.	Ref. [18]	Diff.
$5f_{5/2}$	0	0	0	0	0	0	
$5f_{7/2}$	7609	7914	−306	7481	128	7611	−2
$6d_{3/2}$	91 000	90 165	835	90 593	407	91 502	−502
$6d_{5/2}$	100 511	100 347	163	99 841	670	101 056	−545
$7s_{1/2}$	141 447	138 422	3025	141 103	344	141 118	329
$7p_{1/2}$	193 340	191 308	2032	192 508	832	196 146	2806
$7p_{3/2}$	215 886	214 328	1558	213 197	2689	217 482	−1596

contributions from the $7p$ states, it is expected that the accuracy of the CI-LCC approach for these levels would be on the order of 500 cm^{-1}. In the case of the CI-MBPT method, because the contribution from the $7s$ state is significant and the error of MBPT for this state is as large as 3000 cm^{-1}, the expected accuracy of CI-MBPT will be on the order of 1000 cm^{-1}. To amend this, we introduced scaling factors in front of Σ_1 in our calculations to correct single-valence MBPT energies and Σ_2 to correct Coulomb screening to higher orders. We find, indeed, that results improve substantially, especially after scaling of Σ_1 for the s wave to account for the $7s$ state energy shift, and the agreement approaches that for the CI-LCC method.

III. CI + LCC METHOD

In the CI + LCC approach introduced in Ref. [19], corrections to the effective Hamiltonian Σ_1 and Σ_2 are calculated using a modified version of the LCC (all-order) method with single and double excitations (LCCSD) described in Refs. [20,21]. As a result, the effective Hamiltonian contains dominant core and core-valence correlation corrections to all orders. The main issue is to efficiently calculate the LCC correction to $\Sigma_2(ijkl)$.

The implementation of this approach proceeds as follows.

(1) The 2nd-order corrections Σ_1 and Σ_2 to the effective Hamiltonian are calculated in the same way as in the CI + MBPT method.

(2) The single-double (SD) LCC calculations are carried out for Rn-like U^{6+} core with 24 subshells. Single and double excitations are allowed from *all* 24 core subshells.

(3) Using the core LCC results, the single-double (SD) core-valence calculations are carried out for 21 valence states: $7s$-$9s$, $7p_{1/2}$-$9p_{1/2}$, $7p_{3/2}$-$9p_{3/2}$, $6d_{3/2}$-$8d_{3/2}$, $6d_{5/2}$-$8d_{5/2}$, $5f_{5/2}$-$7f_{5/2}$, and $5f_{7/2}$-$7f_{7/2}$. Core excitations are also allowed from *all* 24 core subshells. The LCC method is modified to exclude the valence diagram that will be later accounted for by the CI. This part of the calculation produces the Σ_1 and $\Sigma_2(ijva)$ quantities, where i and j can be any excited state, a are core states, and v are the 21 states on the above list.

(4) The $\Sigma_2(ijvw)$ corrections to the CI Hamiltonian are calculated, with w also taken from the above valence list. We have tested that restricting the LCC calculation to 21 valence electrons results in sufficient numerical accuracy. We note that the remaining $\Sigma_2(ijkl)$ elements are still corrected in 2nd order. More details of the CI + LCC approaches are described

TABLE II. CI + LCC excitation energies (cm^{-1}) and g factors of the lowest states of Th-like U^{2+}. Nonrelativistic values of g factors (g_{nr}) are given by Eq. (4).

Conf.	Term	g factors nr	Present	Energy	Conf.	Term	g factors nr	Present	Energy
		Odd-parity states					Even-parity states		
$5f^3 6d$	$^3K_6$	0.857	0.745	0	$5f^4$	$^5I_4$	0.600	0.639	1846
$5f^3 6d$	$^5K_6^a$	0.905	0.921	4524	$5f^4$	$^1G_4$	1.000	1.001	10 298
$5f^3 6d$	$^5K_6^b$	0.905	0.985	7787	$5f^4$	$^3F_4$	1.250	1.225	14 103
$5f^3 7s$	$^5I_6$	1.071	1.055	8364	$5f^4$	$^3G_4^a$	1.050	1.049	15 890
$5f^3 6d$	$^1I_6$	1.000	1.008	11 880	$5f^4$	$^5G_4^a$	1.150	1.123	19 102
$5f^3 6d$	$^5I_6^a$	1.071	1.053	12 735	$5f^2 6d^2$	$^3H_4^a$	0.800	0.811	19 276
$5f^3 7s$	$^3I_6$	1.024	1.020	12 955	$5f^4$	$^3G_4^b$	1.050	1.048	20 792
$5f^3 6d$	$^5I_6^b$	1.071	1.071	14 601	$5f^4$	$^5G_4^b$	1.150	1.175	22 870
$5f^3 6d$	$^3H_6$	1.167	1.152	14 998	$5f^4$	$^5G_4^c$	1.150	1.115	24 491
$5f^3 6d$	$^3I_6$	1.024	1.033	16 404	$5f^2 6d^2$	$^3H_4^b$	0.800	0.877	25 481
$5f^3 6d$	$^5H_6$	1.214	1.223	17 772	$5f^4$	$^5G_4^d$	1.150	1.089	26 277
$5f^3 7s$	$^5H_6$	1.214	1.204	20 496	$5f^4$	$^3G_4^c$	1.050	1.068	26 792
$5f^3 7s$	$^5I_4$	0.600	0.625	3430	$5f^4$	$^5I_5^a$	0.900	0.907	4791
$5f^3 6d$	$^3H_4^a$	0.800	0.777	6406	$5f^4$	$^3G_5^a$	1.200	1.209	13 939
$5f^3 6d$	$^3H_4^b$	0.800	0.760	8021	$5f^4$	$^5G_5$	1.267	1.260	16 779
$5f^3 6d$	$^5H_4$	0.900	0.904	8692	$5f^4$	$^3G_5^b$	1.200	1.152	18 681
$5f^3 6d$	$^1G_4$	1.000	1.020	12 066	$5f^4$	$^5I_5^b$	0.900	0.922	20 037
$5f^3 7s$	$^5H_4$	0.900	0.938	14 563	$5f^2 6d^2$	$^5K_5^a$	0.667	0.761	22 250
$5f^3 7s$	$^3F_4$	1.250	1.235	16 247	$5f^4$	$^5H_5$	1.100	1.092	23 803
$5f^3 7s$	$^5G_4^a$	1.150	1.135	18 308	$5f^2 6d^2$	$^5I_5$	0.900	0.942	24 088
$5f^3 7s$	$^5G_4^b$	1.150	1.123	20 031	$5f^2 6d^2$	$^3I_5$	0.833	0.886	26 484
$5f^3 7s$	$^3G_4^a$	1.050	1.043	20 649	$5f^4$	$^3H_5^a$	1.033	1.059	26 652
$5f^3 7s$	$^3G_4^b$	1.050	1.094	22 110	$5f^4$	$^3H_5^a$	1.033	1.044	27 856
$5f^3 7s$	$^3G_4^c$	1.050	1.078	23 126	$5f^2 6d^2$	$^1H_5$	1.000	1.017	28 574
$5f^3 6d$	$^5K_5$	0.667	0.726	565	$5f^4$	$^5I_6$	1.071	1.056	7441
$5f^3 7s$	$^3I_5^a$	0.833	0.887	4415	$5f^4$	$^3K_6^a$	0.857	0.875	14 059
$5f^3 6d$	$^3I_5$	0.833	0.890	6782	$5f^2 6d^2$	$^3K_6$	0.857	0.781	17 963
$5f^3 7s$	$^3I_5^b$	0.833	0.870	8431	$5f^4$	$^5H_6$	1.214	1.240	19 152
$5f^3 6d$	$^1H_5$	1.000	0.929	9943	$5f^4$	$^3H_6$	1.167	1.176	21 212
$5f^3 6d$	$^1H_5^a$	1.000	1.004	11 598	$5f^2 6d^2$	$^5I_6$	1.071	1.056	22 974
$5f^3 6d$	$^3H_5$	1.033	1.077	12 590	$5f^4$	$^3K_6^b$	0.857	0.875	24 397
$5f^3 6d$	$^1H_5^b$	1.000	1.009	13 218	$5f^4$	$^3I_6$	1.024	1.024	26 200
$5f^3 7s$	$^5H_5$	1.100	1.120	14 498	$5f^2 6d^2$	$^1I_6$	1.000	1.002	27 060
$5f^3 6d$	$^3G_5$	1.200	1.165	15 875	$5f^2 6d^2$	$^5K_6$	0.905	0.934	27 476
$5f^3 6d$	$^5H_5$	1.100	1.097	17 867	$5f^4$	$^1I_6$	1.000	0.979	29 103
$5f^3 7s$	$^5G_5$	1.267	1.264	18 932	$5f^2 6d^2$	$^3I_6$	1.024	1.037	29 407
$5f^3 6d$	$^3L_7$	0.875	0.918	4136	$5f^4$	$^3I_7$	1.143	1.142	9769
$5f^3 6d$	$^3K_7$	1.018	1.039	8061	$5f^4$	$^1K_7$	1.000	0.973	15 546
$5f^3 6d$	$^5I_7^a$	1.179	1.151	11 898	$5f^4$	$^3L_7$	0.875	0.950	20 868
$5f^3 6d$	$^1K_7$	1.000	0.975	12 584	$5f^2 6d^2$	$^3L_7$	0.875	0.932	23 126
$5f^3 6d$	$^5K_7$	1.054	1.039	13 569	$5f^4$	$^5K_7^a$	1.054	1.097	23 764
$5f^3 6d$	$^3I_7$	1.143	1.144	15 495	$5f^2 6d^2$	$^3K_7$	1.018	1.019	28 042
$5f^3 7s$	$^3I_7$	1.143	1.109	16 414	$5f^4$	$^3K_7$	1.018	1.024	29 918
$5f^3 7s$	$^3I_7$	1.143	1.127	17 054	$5f^2 6d^2$	$^3I_7$	1.143	1.114	31 000
$5f^3 6d$	$^5I_7^b$	1.179	1.178	18 514	$5f^4$	$^5K_7^b$	1.054	1.042	31 419
$5f^3 6d$	$^3I_7$	1.143	1.110	20 398	$5f^4$	$^5K_7^c$	1.054	1.053	31 898
$5f^3 7s$	$^3K_7$	1.018	1.021	21 482	$5f^2 6d^2$	$^5K_7$	1.054	1.081	34 670
$5f^3 7s$	$^1K_7$	1.000	0.970	23 987	$5f^2 6d^2$	$3K_7$	1.018	1.012	35 556

TABLE III. Excitation energies (cm^{-1}) of U^{2+} evaluated using the CI + LCC approach are compared with compilation from Ref. [12]. The LCC energies for the odd-parity states in Table II and in a similar way CI-MBPT energies are shifted by 210 cm^{-1} to facilitate comparison with the original experimental data. CI-2nd-order MBPT calculations are done for odd states to evaluate theoretical accuracy of the CI-LCC approach, which is on the order of a few 100 cm^{-1}, except for $J = 3$ odd states. Even states have very limited experimental data, so the comparison there not only serves to test theoretical accuracy but also to fill in the gaps in the experimental data.

Conf.	Term	Energy				Conf. + Term	Conf.	Term	Energy		Conf. + Term
Present		CI + LCC	CI + 2nd	Expt. [12]	Dev.	[12]	Present		CI + LCC	[12]	
		Odd-parity states							Even-parity states		
$5f^36d$	$^3G_3^a$	4303	6162	4611	−98	$5f^36d$ ^5H	$5f^4$	$^5I_4$	0	0	$5f^4$ $^5I_4$
$5f^36d$	$^3G_3^b$	9108	9882	8569	749	$5f^36d$	$5f^4$	$^1G_4$	8438		
$5f^36d$	$^3F_3$	10677	12525	9186	1701	$5f^36d$	$5f^4$	$^3F_4$	12243		
$5f^36d$	$^3G_3^c$	12956	12906	11948	1218	$5f^36d$	$5f^4$	$^3G_4^a$	14030		
$5f^37s$	$^5G_3^b$	17105		17058	257	$5f^36d$	$5f^4$	$^5G_4^b$	21010	24249	4
$5f^37s$	$^3D_3$	25548					$5f^4$	$^3G_4^c$	24932	24935	4
$5f^37s$	$^5I_4$	3640	2771	3745	105	$5f^37s$ ^5I	$5f^4$	$^5I_5^a$	2932	3037	$5f^4$ $^5I_5$
$5f^36d$	$^3H_4^a$	6616	6721	6286	540	$5f^36d$ ^5I	$5f^4$	$^3G_5^a$	12080		
$5f^36d$	$^3H_4^b$	8231	8478	7894	547	$5f^36d$	$5f^4$	$^5G_5$	14919		
$5f^36d$	$^5H_4$	8902	10284	9113	−1	$5f^36d$	$5f^4$	$^3G_5^b$	16821		
$5f^37s$	$^5H_4$	14773	13022	14669	314	$5f^36d$ (4)5	$5f^26d^2$	$^5K_5^a$	20390		
$5f^37s$	$^5G_4^a$	18518					$5f^26d^2$	$^5I_5$	22228	23531	$5f^26d^2$ 5
$5f^37s$	$^3G_4^b$	22320					$5f^4b$	$^3H_5$	25996	25611	5
$5f^36d$	$^5K_5$	775	1133	885	100	$5f^36d$ ^5K	$5f^4$	$^5I_6$	5582	5719	$5f^4$ $^5I_6$
$5f^37s$	$^3I_5^a$	4625	4040	4718	117	$5f^37s$ ^5I	$5f^4a$	$^3K_6$	12199		
$5f^36d$	$^3I_5$	6782	7959	7288	−86	$5f^36d$	$5f^26d^2$	$^3K_6$	16104		
$5f^37s$	$^3I_5^b$	8641	8130	8816	35	$5f^37s$ ^3I	$5f^4$	$^5H_6$	17293		
$5f^36d$	$^1H_5$	10153	9840	9864	499	$5f^36d$ ^5I	$5f^4$	$^3H_6$	19353	19417	$5f^26d^2$ $^5L_6$
$5f^36d$	$^3H_5$	12800	12535	13024	−14	$5f^36d$ (5)4	$5f^4b$	$^3K_6$	22537		
$5f^36d$	$^1H_5^b$	13428	13964	13192	446	$5f^36d$ (5)4	$5f^4$	$^3I_6$	24340	24539	$5f^26d^2$ 6(7)
$5f^37s$	$^5H_5$	14708	14769	14669	249	$5f^36d$ (4)5	$5f^26d^2$	$^1I_6$	25201		
$5f^36d$	$^3G_5$	16085	15310	15008	1287	$5f^36d$ (4)5	$5f^26d^2$	$^5K_6$	25616		
$5f^36d$	$^5H_5$	18077	16750	17250	1037	$5f^36d$ (4)5	$5f^4$	$^1I_6$	27243		
$5f^37s$	$^5G_5$	19142	18154	18510	842	$5f^36d$ (5)6	$5f^26d^2$	$^3I_6$	27548		
$5f^36d$	$^3K_6$	210	210	210	210	$5f^36d$ $^5L_6$	$5f^4$	$^3I_7$	7910		
$5f^36d$	$^5K_6^a$	4734	4670	4940	4	$5f^36d$ $^5K_6$	$5f^4$	$^1K_7$	13687		
$5f^36d$	$^5K_6^b$	7997	7716	7894	313	$5f^36d$ 6	$5f^4$	$^3L_7$	19008		
$5f^37s$	$^5I_6$	8574	9150	8778	6	$5f^37s$ $^5I_6$	$5f^26d^2$	$^3L_7$	21266		
$5f^36d$	$^1I_6$	12090	12359	12210	90		$5f^4a$	$^5K_7$	21905		
$5f^36d$	$^5I_6^a$	12945	12628	12636	519	$5f^36d$ ^5I	$5f^26d^2$	$^3K_7$	26182	25507	$5f^26d$ $^5L_7$
$5f^36d$	$^5H_6$	17982	18321	18510	−318	(5)6	$5f^26d^2$	$^5K_7$	32810		
$5f^37s$	$^5H_6$	20706	20197	20689	227	(5)6	$5f^26d^2$	$^3K_7$	33696	33993	7(6)
$5f^36d$	$^3L_7$	4346	4556	4504	52	$5f^36d$ $^5L_7$					
$5f^36d$	$^3K_7$	8271	8481	8437	44	$5f^36d$ $^5K_7$					
$5f^36d$	$^5I_7^a$	12108	12318	12025	293	$5f^37s$ $^5I_7$					

in Ref. [19]. All of the 2nd-order and the LCC calculations include partial waves with $l = 0$–6.

(5) The CI method [22] is then used to treat valence-valence correlations, with the CI code modified to include the effective Hamiltonian constructed as described above. The CI space includes configurations with four valence electrons in our approach and is constructed as described, for example, in Ref. [23]. Briefly, we start with $5f^4$, $5f^36d$, $5f^37s$, $5f^37p$, $6d^25f^2$, and $6d5f^27s$ configurations and allow up to two replacements of any of the configuration electrons to the set of $13s12pdfg$ orbitals to construct the configurations for the CI calculation.

The CI + LCC method was used to evaluate properties of atomic systems with two to four valence electrons [24–30]. This method was also used to calculate atomic properties of the superheavy elements No, Lr, and Rf by Dzuba *et al.* [31]. The $7s^2$ and $7snl$ states were considered for nobelium atoms, the $7s^26d$, and $7s7p6d$ states were considered for lawrencium atoms, and the $7s^26d^2$, $7s^27p6d$, and $7s7p6d^2$ states were considered for rutherfordium atoms [31].

The CI + LCC method was used to calculate energies in Ce, Ce$^+$, La, Ce^{2+}, and La$^+$, respectively [13], and to study various correlation corrections in these systems. The differences between neutral and low-ionized systems were considered. The ground states in Ce^{2+} and La$^+$ are $4f^2\,^3H_4$ and $5d^2\,^3D_2$ instead of the usual the $ns^2\,^1S_0$ ground state in Pb^{2+} [24], Tl$^+$ [25], and Si^{2+} [26].

IV. RESULTS

A. Excitation energies in U^{2+}

Excitation energies for the lowest states of U^{2+} are presented in Tables II and III. To save space, we list results in Table II for only 96 states instead of the 192 states that we included in our calculations. We presented results for $J = 4$, 5, 6, and 7 even- and odd-parity states in Table II. The results are ordered by energy within each J for both even and odd states. Since the ground state is the odd $J = 6$ state in our calculations, we list the $J = 6$ set of odd states first, and list the $J = 4$, 5, and 7 odd results below.

The g factors were also evaluated and compared to nonrelativistic (nr) values of g factors given by Eq. (4) for identification of the LS terms:

$$g_{nr} = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}, \quad (4)$$

where J is total angular momentum of the atom, L is its angular momentum, and S is the spin ($\mathbf{J} = \mathbf{L} + \mathbf{S}$).

Our results give the odd ground state $5f^36d\,^3K_6$ instead of the even $5f^4\,^5I_4$ state listed as a ground state in Ref. [12]. The energy level of the $5f^4\,^5I_4$ level relative our ground $5f^36d\,^3K_6$

state is 1846 cm^{-1}. Table II data are all counted from the ground $5f^36d\,^3K_6$ state. In order to provide comparison with Ref. [12] in Table III, we count the energies of odd and even states from the corresponding lowest levels, $5f^4\,^5I_4$ for even-parity states and $5f^36d\,^3K_6$ for odd-parity states. We added 210 cm^{-1} to the odd states to align theoretical and experimental levels with respect to the $5f^36d\,^3K_6$ level.

Our and Blaise and Wyart's [12] assignments of configurations and LSJ parameters are shown in separate columns. Some of the energy levels listed in Ref. [12] are only identified by the total angular momentum J and not by a complete LSJ term designation. Such designations are always approximate and sometimes ambiguous, as in cases of strong configuration mixing.

Note that $5f^4, 5f^26d^2$ and $5f^36d, 5f^37s$ are dominant configurations for even- and odd-parity states, respectively, among the considered levels.

We find that CI + LCC calculations are in very good agreement with experiment (see Table III) considering the complexity of this ion for theory. More specifically, in most cases for odd states, the deviation was a few 100 cm^{-1} out of 10 000 cm^{-1}, but four levels had differences exceeding 1000 cm^{-1}. The experimental data for even states are fairly incomplete; nevertheless, because of large spacing between theoretical levels and the established accuracy for the odd states, the comparison can be also done for even levels, confirming the experimental levels. In addition, the CI-LCC calculations provide many missing energy levels. This information can be used for the experimental search of these levels and for the analysis of lifetimes requiring branching ratios data. The deviation from experiment in even states is more or

TABLE IV. Wavelengths (λ in Å); multipole matrix elements Z_{M1}^{CI+LCC}, Z_{E2}^{CI+LCC}, and Z_{M3}^{CI+LCC} ($E2$ in a.u.; $M1$ and $M3$ in units of μ_B, half a.u.); and transition rates A_r^{M1}, A_r^{E2}, and A_r^{M3} (in s^{-1}) evaluated using the CI + LCC approach. The numbers in brackets represent powers of 10.

Conf.	Term	Conf.	Term	λ	Z_{M1}^{CI+LCC}	Z_{E2}^{CI+LCC}	Z_{M3}^{CI+LCC}	A_r^{M1}	A_r^{E2}/A_r^{M1}	A_r^{M3}/A_r^{M1}
$5f^4$	$^5G_4^c$	$5f^4$	$^5G_4^d$	55 960	0.150	1.347	3.093	3.84[−04]	1.1[−03]	6.7[−18]
$5f^4$	$^5G_4^c$	$5f^4$	$^3G_4^c$	43 459	0.029	0.704	0.096	3.03[−05]	1.3[−02]	4.8[−19]
$5f^26d^2$	$^3H_4^a$	$5f^4$	$^3G_4^c$	13 305	0.029	0.923	3.570	1.10[−03]	2.3[−01]	7.3[−14]
$5f^4$	$^3F_4$	$5f^4$	$^5G_4^b$	11 406	0.011	0.706	10.316	2.37[−04]	1.4[+00]	8.3[−12]
$5f^4$	$^1G_4$	$5f^26d^2$	$^3H_4^a$	11 138	0.389	1.023	2.290	3.29[−01]	2.3[−03]	3.5[−16]
$5f^4$	$^3F_4$	$5f^4$	$^5G_4^c$	9626	0.064	0.737	0.921	1.40[−02]	5.8[−02]	3.7[−15]
$5f^4$	$^1G_4$	$5f^4$	$^3G_4^b$	9528	0.088	0.421	0.985	2.68[−02]	1.0[−02]	2.4[−15]
$5f^4$	$^3G_4^a$	$5f^4$	$^3G_4^c$	9173	0.282	0.419	5.538	3.10[−01]	1.1[−03]	8.4[−15]
$5f^4$	$^3F_4$	$5f^26d^2$	$^3H_4^b$	8788	0.055	0.585	1.206	1.36[−02]	6.0[−02]	1.2[−14]
$5f^4$	$^1G_4$	$5f^4$	$^5G_4^b$	7954	0.105	1.266	2.976	6.53[−02]	9.6[−02]	3.1[−14]
$5f^4$	$^3F_4$	$5f^4$	$^3G_4^c$	7881	0.073	0.101	2.499	3.24[−02]	1.3[−03]	4.7[−14]
$5f^4$	$^1G_4$	$5f^4$	$^5G_4^c$	7046	0.061	0.406	0.647	3.22[−02]	3.7[−02]	7.0[−15]
$5f^4$	$^1G_4$	$5f^26d^2$	$^3H_4^b$	6586	0.032	1.212	0.130	1.05[−02]	1.4[+00]	1.4[−15]
$5f^4$	$^1G_4$	$5f^4$	$^5G_4^d$	6258	0.050	0.102	1.702	3.06[−02]	4.4[−03]	1.2[−13]
$5f^4$	$^1G_4$	$5f^4$	$^3G_4^c$	6063	0.068	0.833	3.607	6.28[−02]	1.7[−01]	3.2[−13]
$5f^4$	$^5I_4$	$5f^4$	$^5G_4^a$	5805	0.033	0.729	1.808	1.73[−02]	5.9[−01]	4.0[−13]
$5f^4$	$^5I_4$	$5f^26d^2$	$^3H_4^a$	5746	0.006	1.438	2.485	5.12[−04]	8.1[+01]	2.7[−11]
$5f^4$	$^5I_4$	$5f^4$	$^5G_4^b$	4763	0.047	0.162	1.031	6.04[−02]	2.2[−02]	1.5[−13]
$5f^4$	$^5I_4$	$5f^26d^2$	$^3H_4^b$	4236	0.006	0.268	0.449	1.64[−03]	4.0[+00]	2.4[−12]
$5f^4$	$^5I_4$	$5f^4$	$^3G_4^c$	4013	0.045	0.233	1.257	9.56[−02]	6.8[−02]	4.6[−13]

TABLE V. Excitation energies (cm^{-1}), wavelengths (λ in Å), dipole matrix elements $Z^{\text{CI+LCC}}$ in a.u., oscillator strengths f in arbitrary units, and transition rates (A_r in s^{-1}) evaluated using the CI + LCC approach. The numbers in brackets represent powers of 10.

Conf.	Term	Conf.	Term	Energies in cm^{-1}			λ	$Z^{\text{CI+LCC}}$	gf	gA_r
Low level		Upper level		Low	Upper	ΔE	(Å)	(a.u.)	(arb. units)	(s^{-1})
$5f^4$	$^5I_4$	$5f^36d$	$^5H_3$	1846	13 056	11 210	8921	1.90650	0.1238	1.037[7]
$5f^4$	$^5I_6$	$5f^36d$	$^5H_5$	7441	17 855	10 414	9602	2.17260	0.1493	1.080[7]
$5f^36d$	$^3K_6$	$5f^26d^2$	$^3K_6$	0	17 963	17 963	5567	1.47800	0.1192	2.565[7]
$5f^36d$	$^3K_6$	$5f^26d^2$	$^5K_5^a$	0	22 250	22 250	4494	1.47350	0.1467	4.846[7]
$5f^36d$	$^5K_5$	$5f^26d^2$	$^5K_5^a$	553	22 250	21 697	4609	1.43010	0.1348	4.233[7]
$5f^36d$	$^3L_7$	$5f^26d^2$	$^3L_7$	4124	23 126	19 002	5263	1.60390	0.1485	3.576[7]
$5f^36d$	$^5K_6^a$	$5f^26d^2$	$^5I_5$	4511	24 088	19 577	5108	0.85588	0.0436	1.114[7]
$5f^36d$	$^3L_7$	$5f^4$	$^3K_6$	4124	24 397	20 273	4933	0.76973	0.0365	1.000[7]
$5f^36d$	$^3L_7$	$5f^26d^2$	$^1I_6$	4124	27 060	22 936	4360	0.89745	0.0561	1.969[7]
$5f^36d$	$^5K_6^a$	$5f^26d^2$	$^1I_6$	4511	27 060	22 549	4435	0.71342	0.0349	1.182[7]
$5f^36d$	^3L	$5f^26d^2$	$^5K_6$	4124	27 476	23 352	4282	1.37940	0.1350	4.909[7]
$5f^36d$	$^5K_6^a$	$5f^26d^2$	$^5K_6$	4511	27 476	22 965	4354	1.01970	0.0725	2.552[7]
$5f^36d$	$^3K_7$	$5f^26d^2$	$^3K_7$	8049	28 042	19 993	5002	1.00590	0.0614	1.638[7]
$5f^36d$	$^5K_6^b$	$5f^26d^2$	$^1H_5$	7775	28 574	20 799	4808	1.23680	0.0966	2.789[7]
$5f^36d$	$^3K_7$	$5f^26d^2$	$^3I_7$	8049	31 000	22 951	4357	0.68950	0.0331	1.165[7]
$5f^36d$	$^3K_7$	$5f^4$	$^5K_7$	8049	31 419	23 370	4279	0.72833	0.0377	1.372[7]
$5f^36d$	$^3K_7$	$5f^4$	$^5K_7$	8049	31 898	23 849	4193	0.81787	0.0485	1.838[7]
$5f^36d$	$^5D_1$	$5f^26d^2$	$^5D_1$	9607	33 609	24 002	4166	0.62427	0.0284	1.092[7]
$5f^36d$	$^5G_2$	$5f^26d^2$	$^3D_1$	9070	37 657	28 587	3498	0.81754	0.0580	3.164[7]
$5f^36d$	$^3F_2^b$	$5f^26d^2$	$^3D_1$	12 855	37 657	24 802	4032	0.60089	0.0272	1.116[7]

less similar. Levels with unusually large deviations might need additional theoretical and experimental verification.

The CI-MBPT method requires adjustments of scaling factors in front of Σ_1 and to a lesser extent in front of Σ_2 to approach the accuracy of the LCC approach, with the most important being the adjustment of the s-wave correction of Σ_1, as we have already discussed. The comparison with CI-MBPT calculations serves two purposes. One is to understand the strength of valence-core interactions needed to estimate theoretical accuracy. Second, it is important to answer the question of whether the CI-MBPT method, as much simpler and now available as open source software [23], can be used for calculations of actinide properties. Although such calculations require adjustments of correlation corrections, such adjustments improve agreement and simplify identification. The situation is similar to that with the Cowan code; however, in contrast, the number of adjustable parameters is much smaller.

B. Multipole matrix elements, transition rates, and lifetimes in Th-like U^{2+}

We evaluated 3024 $E1$, $M2$, and $E3$ matrix elements that included transitions between even-parity states with $J = 0$ to $J = 7$ and odd-parity states with $J = 0$ to $J = 7$. As we noted above, we calculated 12 even- and odd-parity states for each of the $J = 0$–7. Therefore, each set of matrix element calculations between J and J' sets includes 144 transitions, with $21 J - J'$ cases. We also evaluate multipole $M1$, $E2$, and $M3$ matrix elements for 64 transitions inside of even-parity ($5f^4 + 5f^26d^2$) sets of states. Such large numbers of transitions are needed for the evaluation of lifetimes.

Our CI + LCC results for the multipole matrix elements $Z^{\text{CI+LCC}}$, oscillator strengths f, transition rates A_r, and lifetimes $\tau^{\text{CI+LCC}}$ in Th-like U^{2+} are listed in Tables IV, V, VI, and VII. Results for the effective multipole operator include random phase approximation (RPA) corrections. The code packages for the calculation of matrix elements and RPA correction to the matrix elements are the same for CI + MBPT and CI + LCC approaches and are described in detail in Ref. [23]. The expected accuracy for strong transitions, evaluated from the accuracy of transition energies, is on the order of 10%. The package has only length form output for the electric-dipole transitions, so the difference between length and velocity forms cannot be used for testing the accuracy of the matrix elements.

The multipole A_r^{Ek} ($E1$, $E2$, and $E3$) and multipole A_r^{Mk} ($M1$, $M2$, and $M3$) transition probabilities (s^{-1}) are obtained in terms of matrix elements Z_{Ek} and Z_{Mk} (a.u.) and transition energies ΔE (a.u.) as

$$A_r^{Ek} = \frac{C^{(k)}[\Delta E]^{2k+1}}{(2J+1)}(Z_{Ek})^2, \quad C^{(1)} = 2.14200 \times 10^{10},$$

$$C^{(2)} = 5.70322 \times 10^4, \quad C^{(3)} = 7.71311 \times 10^{-2},$$

$$A_r^{Mk} = \frac{D^{(k)}[\Delta E]^{2k+1}}{(2J+1)}(Z_{Mk})^2, \quad D^{(1)} = 2.85161 \times 10^5,$$

$$D^{(2)} = 7.59260 \times 10^{-1}, \quad D^{(3)} = 1.02683 \times 10^{-6}. \tag{5}$$

TABLE VI. Lifetimes τ^{CI+LCC} (in ms), branching ratio, transition rates A_r (in s^{-1}), and reduced matrix elements Z^{CI+LCC} (in a.u.) for electric-dipole ($E1$) transitions in U^{2+} ion evaluated in the CI + LCC approximation. The numbers in brackets represent powers of 10.

Conf.	Term	Conf.	Term	Conf.	Term	Energies (cm^{-1})		λ	Z^{CI+LCC}	A_r^{CI+LCC}	Branch.	τ^{CI+LCC}
Level		Lower level		Upper level		Lower	Upper	(Å)	(a.u.)	(s^{-1})	ratio	(msec)
$5f^4$	$^5I_6$	$5f^3 6d$	$^5Ka_6$	$5f^4$	$^5I_6$	4511	7441	34 130	0.237	2.19[2]	0.66	3.009
		$5f^3 6d$	$^5K_5$	$5f^4$	$^5I_6$	553	7441	14 518	0.040	8.28[1]	0.25	
$5f^3 6d$	$^3H_4^b$	$5f^4$	$^5I_4$	$5f^3 6d$	$^3H_4^b$	1846	8009	16 226	1.330	9.32[4]	0.95	0.010
		$5f^4$	$^5I_5^a$	$5f^3 6d$	$^3H_4^b$	4791	8009	31 075	0.770	4.44[3]	0.05	
$5f^3 7s$	$^3I_5^b$	$5f^4$	$^5I_5^a$	$5f^3 7s$	$^3I_5^b$	4791	8419	27 563	0.130	1.48[2]	0.54	3.650
		$5f^4$	$^5I_4$	$5f^3 7s$	$^3I_5^b$	1846	8419	15 214	0.049	1.26[2]	0.46	
$5f^4$	$^5F_1$	$5f^3 6d$	0	$5f^4$	$^5F_1$	5165	9682	22 139	0.079	3.92[2]	0.76	1.928
		$5f^3 6d$	$^5G_2$	$5f^4$	$^5F_1$	9070	9682	163 399	0.905	1.27[2]	0.24	
$5f^4$	$^3I_7$	$5f^3 6d$	$^5K_6^b$	$5f^4$	$^3I_7$	7775	9769	50 150	0.423	1.91[2]	0.64	3.364
		$5f^3 6d$	$^3K_7$	$5f^4$	$^3I_7$	8049	9769	58 140	0.304	6.35[1]	0.21	
$5f^3 6d$	$^1H_5$	$5f^4$	$^5I_5^a$	$5f^3 6d$	$^1H_5$	4791	9931	19 455	2.092	1.09[5]	0.86	0.008
		$5f^4$	$^5I_4$	$5f^3 6d$	$^1H_5$	1846	9931	12 369	0.425	1.76[4]	0.14	
$5f^4$	$^1G_4$	$5f^3 6d$	$^3I_5$	$5f^4$	$^1G_4$	6769	10 298	28 337	0.736	5.36[3]	0.64	0.120
		$5f^3 6d$	$^3H_4^a$	$5f^4$	$^1G_4$	6393	10 298	25 608	0.308	1.27[3]	0.15	
$5f^3 7s$	$^5F_1$	$5f^4$	$^1D_2^a$	$5f^3 7s$	$^5F_1$	8591	11 074	40 274	0.086	7.67[1]	0.88	11.509
		$5f^4$	$^5F_1$	$5f^3 7s$	$^5F_1$	9682	11 074	71 839	0.075	1.02[1]	0.12	
$5f^4$	$^3F_3^a$	$5f^3 6d$	$^3H_4^a$	$5f^4$	$^3F_3^a$	6393	11 312	20 329	0.408	5.75[3]	0.52	0.090
		$5f^3 6d$	$^3G_3^a$	$5f^4$	$^3F_3^a$	4081	11 312	13 829	0.144	2.27[3]	0.21	0.090
		$5f^3 6d$	$^5H_4$	$5f^4$	$^3F_3^a$	8679	11 312	37 979	0.617	2.01[3]	0.18	
$5f^3 6d$	$^1H_5^a$	$5f^4$	$^5I_5^a$	$5f^3 6d$	$^1H_5^a$	4791	11 586	14 717	0.520	1.56[4]	0.67	0.043
		$5f^4$	$^5I_4$	$5f^3 6d$	$^1H_5^a$	1846	11 586	10 267	0.159	4.32[3]	0.19	
		$5f^4$	$^5I_6$	$5f^3 6d$	$^1H_5^a$	7441	11 586	24 125	0.507	3.37[3]	0.14	
$5f^4$	$^3F_2^a$	$5f^3 6d$	$^3G_3^a$	$5f^4$	$^3F_2^a$	4081	11 601	13 298	0.334	1.92[4]	0.81	0.042
		$5f^3 6d$	$^3G_3^b$	$5f^4$	$^3F_2^a$	8886	11 601	36 832	0.576	2.70[3]	0.11	
		$5f^3 6d$	$^5G_2$	$5f^4$	$^3F_2^a$	9070	11 601	39 510	0.478	1.50[3]	0.06	
$5f^3 7s$	$^1D_2$	$5f^4$	$^1D_2^a$	$5f^3 7s$	$^1D_2$	8591	11 698	32 185	0.081	8.02[1]	0.82	10.165
		$5f^4$	$^5F_1$	$5f^3 7s$	$^1D_2$	9682	11 698	49 603	0.074	1.82[1]	0.18	
$5f^3 6d$	$^1I_6$	$5f^4$	$^5I_6$	$5f^3 6d$	$^1I_6$	7441	11 868	22 589	0.345	1.61[3]	0.62	0.385
		$5f^4$	$^5I_5^a$	$5f^3 6d$	$^1I_6$	4791	11 868	14 130	0.114	7.16[2]	0.28	

C. Multipole matrix elements and transition rates

In Table IV, we list wavelengths; multipole matrix elements Z^{CI+LCC}_{M1}, Z^{CI+LCC}_{E2}, and Z^{CI+LCC}_{M3}; and transition rates A_r^{M1}, A_r^{E2}, and A_r^{M3} evaluated using the CI + LCC approach. We evaluate 64 transitions between the even-parity states ($5f^4 + 5f^2 6d^2$), but list only 20 transitions in Table IV to save space. The $E2/M1$ and $M3/E1$ ratios of transition rates are shown in the two last columns of Table IV. The ratios of A_r^{E2}/A_r^{M1} are generally small, 10^{-3}–10^{-2}. However, there are transitions with A_r^{E2} being larger than the A_r^{M1}. The A_r^{M3}/A_r^{M1} ratio is much smaller than A_r^{E2}/A_r^{M1}, as expected, 10^{-19}–10^{-11}.

In Table V, we list excitation energies, wavelengths, dipole matrix elements Z^{CI+LCC}, oscillator strengths f, and transition rates evaluated using the CI + LCC approach. In Table V, we present results for 20 transitions among the 3024 transitions that we considered. We choose transitions with the largest values of A_r. It should be noted that we evaluated also values of A_r for magnetic-quadrupole $M2$ and electric-octupole $E3$

transitions. We find that the A_r^{M2}/A_r^{E1} ratio is small, about 10^{-6}. The A_r^{E3}/A_r^{E1} ratio is extremely small, about 10^{-16}, as expected, and we did not include those transitions in Table V and the other two tables with results for lifetimes and branching ratios.

D. Branching ratios and lifetimes in U^{2+}

In Table VI, we list lifetimes τ^{CI+LCC}, branching ratios, transition rates A_r, and reduced matrix elements Z^{CI+LCC} for electric-dipole transitions. We evaluate the results for 188 levels in Th-like U^{2+}, which excludes several metastable levels with no contributing $E1$ transitions; however, we show data for only 12 levels in Table VI for illustration.

In order to determine the lifetimes listed in the last column of Table VI, we sum over all possible radiative transitions. The number of contributing transitions increases significantly for higher levels. For example, 8 transitions contribute to the lifetime of the relatively low-lying $5f^4$ $^3F_3$ state, $E(5f^4\ ^3F_3) = 11\,601$ cm^{-1}. However, only one transition,

TABLE VII. Lifetimes $\tau^{\mathrm{CI+LCC}}$ (in ms) and sum of transition rates $\sum A_r$ (in s^{-1}) for electric-dipole ($E1$) transitions in U^{2+} ion evaluated in the CI + LCC approximation. The numbers in brackets represent powers of 10.

Conf.	Term	Energies	$\sum A_r^{\mathrm{CI+LCC}}$	$\tau^{\mathrm{CI+LCC}}$	Conf.	Term	Energies	$\sum A_r^{\mathrm{CI+LCC}}$	$\tau^{\mathrm{CI+LCC}}$
		Odd-parity states					Even-parity states		
$5f^36d$	$^3K_6$	0.0			$5f^4$	$^5I_4$	1846	2.89[+2]	3.46[+0]
$5f^36d$	$^5K_6^a$	4511			$5f^4$	$^1G_4$	10 298	8.37[+3]	1.20[−1]
$5f^36d$	$^5K_6^b$	7775	2.63[+1]	3.81[+1]	$5f^4$	$^3F_4$	14 103	4.03[+3]	2.48[−1]
$5f^37s$	$^5I_6$	8352	1.39[+0]	7.21[+2]	$5f^4$	$^3G_4^a$	15 890	2.32[+4]	4.30[−2]
$5f^36d$	$^1I_6$	11 868	2.60[+3]	3.85[−1]	$5f^4$	$^5G_4^a$	19 102	2.82[+5]	3.55[−3]
$5f^36d$	$^5I_6^a$	12 723	1.17[+5]	8.54[−3]	$5f^26d^2$	$^3H_4^a$	19 276	1.61[+6]	6.20[−4]
$5f^37s$	$^3I_6$	12 943	1.95[+4]	5.12[−2]	$5f^4$	$^3G_4^b$	20 792	1.18[+5]	8.49[−3]
$5f^36d$	$^5I_6^b$	14 589	5.06[+4]	1.98[−2]	$5f^4$	$^5G_4^b$	22 870	1.63[+4]	6.15[−2]
$5f^36d$	$^3H_6$	14 986	7.90[+3]	1.27[−1]	$5f^4$	$^5G_4^c$	24 491	9.12[+4]	1.10[−2]
$5f^36d$	$^3I_6$	16 392	5.97[+4]	1.68[−2]	$5f^26d^2$	$^3H_4^b$	25 481	2.35[+6]	4.26[−4]
$5f^37s$	$^5I_4$	3418	6.06[+0]	1.65[+2]	$5f^4$	$^5Ia_5$	4791	2.74[+2]	3.65[+0]
$5f^36d$	$^3H_4^a$	6393	3.96[+4]	2.53[−2]	$5f^4$	$^3G_5^a$	13 939	1.23[+4]	8.15[−2]
$5f^36d$	$^3H_4^b$	8009	9.77[+4]	1.02[−2]	$5f^4$	$^5G_5$	16 779	1.09[+4]	9.17[−2]
$5f^36d$	$^5H_4$	8679	3.60[+4]	2.78[−2]	$5f^4$	$^3G_5^b$	18 681	3.54[+4]	2.83[−2]
$5f^36d$	$^1G_4$	12 054	2.37[+3]	4.23[−1]	$5f^4$	$^5Ib_5$	20 037	8.11[+4]	1.23[−2]
$5f^37s$	$^5H_4$	14 551	9.13[+3]	1.10[−1]	$5f^26d^2$	$^5K_5^a$	22 250	9.04[+6]	1.11[−4]
$5f^37s$	$^3F_4$	16 235	1.26[+3]	7.92[−1]	$5f^4$	$^5H_5$	23 803	5.00[+5]	2.00[−3]
$5f^37s$	$^5G_4^a$	18 296	6.58[+3]	1.52[−1]	$5f^26d^2$	$^5I_5$	24 088	2.61[+6]	3.84[−4]
$5f^36d$	$^5K_5$	553			$5f^4$	$^5I_6$	7441	3.32[+2]	3.01[+0]
$5f^37s$	$^3I_5^a$	4403	3.74[+0]	2.68[+2]	$5f^4$	$^3K_6$	14 059	1.75[+4]	5.72[−2]
$5f^36d$	$^3I_5$	6769	2.76[+2]	3.62[+0]	$5f^26d^2$	$^3K_6$	17 963	2.43[+6]	4.11[−4]
$5f^37s$	$^3I_5^b$	8419	2.74[+2]	3.65[+0]	$5f^4$	$^5H_6$	19 152	4.30[+4]	2.33[−2]
$5f^36d$	$^1H_5$	9931	1.28[+5]	7.84[−3]	$5f^4$	$^3H_6$	21 212	4.56[+4]	2.19[−2]
$5f^36d$	$^1H_5^a$	11 586	2.33[+4]	4.29[−2]	$5f^26d^2$	$^5I_6$	22 974	7.41[+4]	1.35[−2]
$5f^36d$	$^3H_5$	12 578	1.87[+4]	5.35[−2]	$5f^4b$	$^3K_6$	24 397	2.73[+6]	3.66[−4]
$5f^36d$	$^3L_7$	4124			$5f^4$	$^3I_7$	9769	2.97[+2]	3.36[+0]
$5f^36d$	$^3K_7$	8049	2.49[+1]	4.02[+1]	$5f^4$	$^1K_7$	15 546	1.88[+4]	5.31[−2]
$5f^36d$	$^5I_7^a$	11 886	7.94[+1]	1.26[+1]	$5f^4$	$^3L_7$	20 868	3.36[+4]	2.97[−2]
$5f^36d$	$^1K_7$	12 571	4.53[+2]	2.21[+0]	$5f^26d^2$	$^3L_7$	23 126	3.04[+6]	3.30[−4]
$5f^36d$	$^5K_7$	13 557	9.07[+3]	1.10[−1]	$5f^4a$	$^5K_7$	23 764	8.84[+4]	1.13[−2]

$5f^36d\ ^3G_3$-$5f^4\ ^3F_3$, contributes significantly, and the total contribution of other 7 transitions to the $5f^4\ ^3F_3$ lifetime is equal to 19%. The final values of $\tau^{\mathrm{CI+all}}$ for 12 lowest-lying levels are listed in the last column of Table VI. The term designation for those levels are in the first column of Table VI.

In Table VII, we present results for the other 92 $E1$ transitions for low-lying levels. In this table, we list lifetimes $\tau^{\mathrm{CI+LCC}}$ (in ms) and sum of transition rates for 12 odd-parity and even-parity states with $J = 4$–7. The largest value of the lifetime is about 721 ms for the $5f^37s\ ^5I_6$ level with excitation energy equal to 8352 cm^{-1}. Unfortunately, we did not find any theoretical or experimental results to compare with our A_r and τ values for the low-lying states listed in Table VII.

We found only one work that reported lifetime measurements of U^{2+} [32], with data given for five levels. The corresponding excitation energies for these levels are in the higher range of 29 000–37 000 cm^{-1}. In order to compare with the lifetimes listed in Ref. [32], we made additional calculations of energies and transition rates using the CI+LCC

method with larger configuration sets. We evaluated energies for the 30 even-parity states with $J = 4$ and 5 to reach the required higher-energy levels. Results of our calculations are presented in Table VIII where we list energies and lifetimes in the intervals of energies 28 000–31 000 cm^{-1} and 30 000–34 000 cm^{-1} for even-parity states with $J = 4$ and 5, respectively. As a result, we were able to compare our CI + LCC results with three lifetime values given in Ref. [32].

Energies of the levels quoted in Ref. [32] were taken from compilation of Ref. [12]. In order to be sure that our identification of levels in Table VIII is correct, we compare also our CI + LCC results with energies from Ref. [12]. Unfortunately, we found only few results, with missing full terms designation and only J being listed.

The theoretical lifetime of 97.5 ns for the the $5f^26d^2$ level with $J = 4$ agrees with experiment 104 ± 10 ns within the experimental precision. The difference in corresponding energies is about 1.5%. The theoretical lifetime for the $5f^26d7s\ ^5H_5$ level, 162 ns, is also in agreement with the

TABLE VIII. Energy levels (cm^{-1}), g factors, and lifetimes (ns) in U^{2+}. Nonrelativistic values of g factors (g_{nr}) are given by Eq. (4). The first column gives the dominant contribution for the configuration. Experimental lifetimes are taken from Ref. [32]. Energy levels are from the experimental compilation of Ref. [12].

	Level		g factors		Energies		Conf.	Lifetimes	
%	Conf.	Level	Present	nr	Present	Expt. [12]	Expt. [12]	Present	Expt. [32]
36	$5f^26d^2$	$^5H_4$	0.898	0.900	28 466	28 922	$5f^26d^2\,^5I$	97.5	104 ± 10
45	$5f^4$	$^1G_4$	0.997	1.000	28 695			167.	
37	$5f^26d7s$	$^5H_4$	0.875	0.900	28 957	28 773	$5f^26d7s\,^5I$	65.0	
56	$5f^26d^2$	$^5H_4$	0.912	0.900	29 349			73.0	
34	$5f^26d7s$	$^3G_4$	1.056	1.050	29 617			307.	
36	$5f^26d^2$	$^5H_4$	0.904	0.900	30 100	31 469	$5f^26d^2$	58.6	41 ± 3
62	$5f^26d^2$	$^3G_4$	1.074	1.050	30 398			212.	
56	$5f^26d^2$	$^3G_4$	1.051	1.050	31 183			117.	
35	$5f^26d7s$	$^5H_4$	0.950	0.900	31 417	32 020	$5f^26d7s$	310.	
38	$5f^26d^2$	$^1G_4$	0.993	1.000	31 840			226.	
65	$5f^26d^2$	$^3G_4$	1.099	1.050	31 994			233.	
49	$5f^26d^2$	$^3H_5$	1.027	1.033	30 170			93.4	
33	$5f^26d7s$	$^1H_5$	0.972	1.000	30 373			84.7	
38	$5f^26d^2$	$^5I_5$	0.960	0.900	31 315			51.7	
62	$5f^26d^2$	$^1H_5$	1.001	1.000	31 821	32 511		79.8	
59	$5f^26d^2$	$^1H_5$	1.002	1.000	32 028	32 945	$5f^26d^2$	48.0	
32	$5f^26d7s$	$^5H_5$	1.077	1.100	32 391	33 237	$5f^26d7s$	162.	150 ± 15
45	$5f^4$	$^1H_5$	0.972	1.000	32 602			31.8	
65	$5f^26d^2$	$^1H_5$	1.008	1.000	32 912	33 546		70.2	
28	$5f^26d^2$	$^5H_5$	1.115	1.100	33 536			34.2	
35	$5f^26d^2$	$^3H_5$	1.024	1.033	33 876			13.4	
37	$5f^26d^2$	$^5H_5$	1.072	1.100	34 216	34 453		15.6	

experiment, 150 ± 15 ns, while the difference in energies is larger, 2.5%. No term identification is given in Ref. [12] for this level. We find about 30% difference in the lifetime and 4.4% in the energy for the $5f^26d^2\,^5H_4$ level.

In order to obtain these lifetimes, we sum transition rates for 36 odd-parity levels with $J = 3$–5 and 36 odd-parity levels with $J = 4$–6. The branching ratios of the odd-parity states with $J = 3$, 4, and 5 are equal to 19.5%, 12.5%, and 68%, respectively, for the $5f^26d^2$ level with $J = 4$ (97.5 ns). For the $5f^26d7s$ level with $J = 5(162$ ns$)$, the branching ratios of the odd-parity states with $J = 4$, 5, and 6 are equal to 12.3%, 42.3%, and 45.5%, respectively.

V. CONCLUSIONS

In this paper, we calculated energy levels, g factors, transition probabilities, and lifetimes for U^{2+} ions. Results for energies and lifetimes are in good agreement with experiment, where available. We compared CI-LCC and CI-2nd-order MBPT calculations. CI-LCC results are in good agreement with experiment without any adjustable parameters, while the CI-MBPT method after scaling correlation corrections

achieves similar agreement, except for even $J = 3$ levels. This paper is focused on CI-LCC calculations, and CI-MPBT results are presented to give an idea of valence-core effects and to give an estimate of the theoretical accuracy. Both theoretical methods can be applied to other systems, for example U$^+$ and neutral U. The CI-MPBT method has the advantage of simplicity, but requires adjustments of correlation corrections. The *ab initio* CI-LCC method, on the other hand, is more accurate, but this method is more complicated and, unlike the CI-MBPT method [23], its availability is limited.

ACKNOWLEDGMENTS

[1] A. Weigand, X. Cao, V. Vallet, J.-P. Flament, and M. Dolg, J. Phys. Chem. A **113**, 11509 (2009).

[2] M. Dolg and X. Cao, J. Phys. Chem. A **113**, 12573 (2009).

[3] L. Brewer, J. Opt. Soc. Am. **61**, 1666 (1971).

[4] B. A. Palmer and R. Engleman, Jr., J. Opt. Soc. Am. B **1**, 609 (1984).

[5] J. Blaise, J. F. Wyart, B. A. Palmer, J. R. Engleman, and F. Launay, in 19th Conf. Eur. Group At. Spectrosc., Dublin, Eire, Europhys. Conf. Abstr. 11E, Comm. A3 (1987).

[6] J. Blaise and J. F. Wyart, in 16th Conf. Eur. Group At. Spectrosc., London, UK, Europhys. Conf. Abstr. 8H, A4 (1984).

[7] X. Cao and M. Dolg, Mol. Phys. **101**, 961 (2003).

[8] W. Liu, W. Küchle, and M. Dolg, Phys. Rev. A **58**, 1103 (1998).

[9] M. S. Safronova, U. I. Safronova, and C. W. Clark, Phys. Rev. A **90**, 032512 (2014).

[10] M. S. Safronova and W. R. Johnson, *Advances in Atomic, Molecular, and Optical Physics* (Academic Press, San Diego, CA, 2007), Vol. 55, p. 194.

[11] http://web2.lac.u-psud.fr/lac/Database/Tab-energy/Thorium/Th-el-dir.html.

[12] J. Blaise and J.-F. Wyart, *International Tables of Selected Constants* (Centre National de la Recherche Scientifique, Paris, 1992), Vol. 20.

[13] M. S. Safronova, U. I. Safronova, and C. W. Clark, Phys. Rev. A **91**, 022504 (2015).

[14] I. M. Savukov, Phys. Rev. A **91**, 022514 (2015).

[15] V. A. Dzuba, Phys. Rev. A **71**, 062501 (2005).

[16] V. A. Dzuba, V. V. Flambaum, P. G. Silvestrov, and O. P. Sushkov, J. Phys. B **20**, 3297 (1987).

[17] V. A. Dzuba, V. V. Flambaum, and O. P. Sushkov, Phys. Lett. A **140**, 493 (1989).

[18] E. Eliav and U. Kaldor, Chem. Phys. **392**, 78 (2012).

[19] M. S. Safronova, M. G. Kozlov, W. R. Johnson, and D. Jiang, Phys. Rev. A **80**, 012516 (2009).

[20] M. S. Safronova, W. R. Johnson, and A. Derevianko, Phys. Rev. A **60**, 4476 (1999).

[21] M. S. Safronova and U. I. Safronova, Phys. Rev. A **83**, 012503 (2011).

[22] S. A. Kotochigova and I. I. Tupitsyn, J. Phys. B **20**, 4759 (1987).

[23] M. G. Kozlov, S. G. Porsev, M. S. Safronova, and I. I. Tupitsyn, Comput. Phys. Commun. **195**, 199 (2015).

[24] M. S. Safronova, M. G. Kozlov, and U. I. Safronova, Phys. Rev. A **85**, 012507 (2012).

[25] Z. Zuhrianda, M. S. Safronova, and M. G. Kozlov, Phys. Rev. A **85**, 022513 (2012).

[26] M. S. Safronova, S. G. Porsev, M. G. Kozlov, and C. W. Clark, Phys. Rev. A **85**, 052506 (2012).

[27] S. G. Porsev, M. S. Safronova, and M. G. Kozlov, Phys. Rev. Lett. **108**, 173001 (2012).

[28] M. S. Safronova, U. I. Safronova, and S. G. Porsev, Phys. Rev. A **87**, 032513 (2013).

[29] M. S. Safronova and P. K. Majumder, Phys. Rev. A **87**, 042502 (2013).

[30] M. S. Safronova, V. A. Dzuba, V. V. Flambaum, U. I. Safronova, S. G. Porsev, and M. G. Kozlov, Phys. Rev. Lett. **113**, 030801 (2014).

[31] V. A. Dzuba, M. S. Safronova, and U. I. Safronova, Phys. Rev. A **90**, 012504 (2014).

[32] E. Biémont, P. Palmer, P. Quinet, S. Svanberg, and Z. G. Zhang, J. Phys. B **35**, 1701 (2002).