

Energy levels and lifetimes of Gd IV and enhancement of the electron electric dipole moment

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(Received 17 June 2002; published 17 September 2002)

We have calculated energy levels and lifetimes of $4f^7$ and $4f^65d$ configurations of Gd IV using Hartree-Fock and configuration-interaction methods. This allows us to reduce significantly the uncertainty of the theoretical determination of the electron electric dipole moment (EDM) enhancement factor in this ion and, correspondingly, in gadolinium-containing garnets for which such measurements were recently proposed. Our new value for the EDM enhancement factor of Gd^{3+} is -2.2 ± 0.5 . Calculations of energy levels and lifetimes for Eu III are used to control the accuracy.

DOI: 10.1103/PhysRevA.66.032105

PACS number(s): 11.30.Er, 31.15.Ne, 32.10.Dk

I. INTRODUCTION

There have been recent suggestions by Lamoreaux [1] and Hunter [2] for searches of the electron electric dipole moment (EDM) in solid-state experiments with the compounds gadolinium gallium garnet $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ and Gadolinium Iron Garnet $\text{Gd}_3\text{Fe}_5\text{O}_{12}$. It is known that an EDM of a system in a stationary quantum state violates both time-reversal (T) and space-reflection (P) symmetries. This is why searches for EDMs of elementary particles, atoms, and molecules are very important for studies of violations of fundamental symmetries [3]. The best limit on the electron EDM comes from the Berkeley experiment of Regan *et al* [4] with an atomic thallium beam, $d_e < 1.6 \times 10^{-27} e$ cm. There are ideas on how to improve the sensitivity substantially working with PbO [5] and YbF [6] molecules. An alternative that can provide a real breakthrough is to use solids containing uncompensated electron spins. This idea was already suggested in 1968 by Shapiro [7]. Application of a strong electric field to electrons bound within a solid would align the EDMs of the unpaired electrons. This should lead to a simultaneous alignment of the electron spins; the magnetic field arising from this alignment could be detected experimentally. Another possibility is to polarize electrons by the external magnetic field. This causes alignment of electron EDMs, and hence induces a voltage across the sample that could be detected. An experiment of this kind has been performed with nickel-zinc ferrite [8], however, due to experimental limitations, the result was not very impressive. Interest in this approach has been renewed recently owing to the suggestions by Lamoreaux [1] and Hunter [2] to perform similar experiments with gadolinium gallium garnet and gadolinium iron garnet, employing new experimental techniques. The estimates of sensitivity

presented in Ref. [1] look highly promising; an improvement by several orders of magnitude is feasible.

The first calculations of the expected effects have been performed in recent papers by Buhmann *et al.* [9] and Kuenzi *et al.* [10] using a semiempirical approach. The compounds under consideration contain Gd^{3+} ions, see Ref. [11], that give the most important contributions to the effect owing to their large nuclear charge. Therefore, from the theoretical point of view, the problem can be split into two closely connected, but still distinct, parts. The first part is the evaluation of the EDM of a Gd^{3+} ion induced by an assumed electron EDM, and the second part is an account of the combined electron-lattice dynamics of the solid. It has been shown by Buhmann *et al.* [9] that the main contribution to the Gd^{3+} EDM comes from mixing between $4f$ and $5d$ electrons. This mixing depends on the energy separation between $4f^7$ and $4f^65d$ configurations. There are experimental data on the relevant energy intervals [12]. However, Ref. [12] does not contain identification of all possible levels; therefore, one cannot rely completely on the data. It has been pointed out by Refs. [9,10] that accurate calculations of the energy levels of the $4f^65d$ configuration of Gd^{3+} ion are needed to improve the accuracy of the EDM calculation.

There were recent measurements of lifetimes of some states of the $4f^65d$ configuration of Eu III [13], which has an electronic structure similar to Gd IV. Therefore, calculations of lifetimes in Eu III provide a good test of the accuracy of $E1$ -transition amplitudes which determine both the lifetimes and the EDM enhancement factor. In the present work we perform calculations of the energy levels and the lifetimes of both Eu III and Gd IV.

II. CALCULATION OF ENERGIES

We use two different sets of computer codes to do our calculations. One is our own configuration interaction program and the second is a code written by Cowan [14] and freely available via the Internet [22]. We use both codes to compute energy levels of Eu III and Gd IV. Calculations for the Eu III are mostly done to control the accuracy. These two

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ions have similar electronic structure, however, much of the reliable experimental data is available for Eu III while limited data are available for Gd IV. We restrict our study to the lowest odd configuration $4f^7$ and even configuration $4f^65d$; these are the configurations most relevant to the atomic EDM.

We will describe our approach in detail while restricting our comments on the Cowan code to a few general remarks. Since our calculations are relativistic, we will use the abbreviation RCI (relativistic configuration interaction) to refer to them. We start our calculations using the relativistic Hartree-Fock (HF) method. Calculations of the self-consistent field are done for an ion in its ground state. This is an open-shell system with seven out of 14 electrons in its outermost $4f$ subshell. Therefore, we apply 50% weighting to the contribution of the $4f$ subshell to the HF potential. This weighting is further reduced to $\sim 46\%$ when the interaction of a $4f$ electron with other electrons of the same subshell is calculated ($6/13 \approx 0.46$). Note, that our calculations are relativistic and we apply the same weighting to both $4f_{5/2}$ and $4f_{7/2}$ subshells. RCI results for the $4f^65d$ configuration are sensitive to how the $5d$ state is calculated. It is natural to calculate it in the field of the $4f^6$ subshell, which means that HF potential is modified by removing a contribution of one $4f$ electron. However, the $5d$ state obtained in this way is still not good enough to achieve accurate energy levels. This is probably because the self-consistent field is calculated for the configuration $4f^7$ and not for $4f^65d$. Therefore, we further modify the $5d$ state by introducing a correction to the HF potential in which this state is calculated as

$$\delta V = - \frac{\alpha}{2(a^4 + r^4)}. \quad (1)$$

Here α is polarizability of an ion in the $4f^6$ configuration, a is a cutoff parameter introduced to remove the singularity in the origin. Potential (1) describes the effect of core polarization by the field of external electron. We treat α as a fitting parameter. Its value ($\alpha = 0.5a_B^3$) has been chosen to obtain accurate energy levels for Eu III. The value of a is not very important because the $5d$ wave function is small at short distances. We use $a = a_B$. We use the same values of a and α for both Eu III and Gd IV.

We now have four single-electron basis states, $4f_{5/2}$, $4f_{7/2}$, $5d_{3/2}$, and $5d_{5/2}$. Many-electron basis states for the RCI calculations are constructed by distributing seven electrons over these states in all possible ways. Then, many-electron states of definite parity and total angular momentum J are constructed. The actual matrix size depends on the configuration considered and the value of the total angular momentum J ; it varies between 1 ($4f^7, J=25/2$) and 377 ($4f^65d, J=9/2$).

Energy intervals in the RCI calculations are sensitive to the value of Slater integrals ($F_2(4f,4f)$, $F_2(4f,5d)$, etc.). In the HF approximation, the value of these integrals, and consequently the energy intervals, are overestimated. This is because of the screening of the Coulomb interaction between valence electrons by core electrons (see, e.g., Ref. [15]). In

TABLE I. Lifetimes of $4f^65d$ $^8P_{5/2,7/2,9/2}$ states of Eu III (ns).

	Expt. ^a	Cowan ^b	Cowan ^c	RPA ^d
$^8P_{5/2}$	65(7)	16	24	30
$^8P_{7/2}$	46(5)	15	15	28
$^8P_{9/2}$	36(4)	10	11	19

^aZhiguo *et al.* [13].

^bThis work, $R_{4f,5d} = 0.77a_B$.

^cMashonkina *et al.* [16].

^dThis work, $R_{4f,5d} = 0.56a_B$.

the present work we include this screening semiempirically by introducing screening factor $f_2 = 0.8$. The value of this factor was chosen to fit energy intervals in Eu III. Thus, in the end, we have two fitting parameters, a core polarizability α and a screening factor f_2 . The values of both of these factors are chosen for Eu III and then the same values are used for Gd IV.

Calculations with the Cowan code are very similar to the RCI calculations. This is also a configuration interaction method, although in its nonrelativistic realization. There are also two fitting procedures in the Cowan code. One is scaling of the Coulomb integrals by a factor of 0.85. This is very similar to our screening of Coulomb interaction. Another fitting that we use in the Cowan code is changing ‘‘by hand’’ the average energy of the ground state configuration. This is equivalent to shifting all energy intervals between the ground and the excited configuration by the same value. A few more details about calculations with the Cowan code will be given in Sec. IV.

III. CALCULATION OF LIFETIMES

Lifetimes of the three $4f^65d$ $^8P_{5/2,7/2,9/2}$ states of Eu III have been recently measured by Zhiguo *et al.* [13]. Calculations using the Cowan code give values which are about three times smaller (see Table I). It is important to investigate the source of this discrepancy.

In a single-configuration approximation which we use in the present work, lifetimes of all states of the $4f^65d$ configuration are determined by a single radial integral

$$R_{4f,5d} = \int_0^\infty R_{4f}(r)R_{5d}(r)r^3 dr, \quad (2)$$

and can be presented in the form

$$\tau_i = A_i / R_{4f,5d}^2, \quad (3)$$

where i denotes a particular energy level. The parameter A_i is sensitive to the mixing of states (correlations between

TABLE II. Energies (cm^{-1}) and lifetimes τ (sec) for Eu III calculated by RCI (E^M) and Cowan codes (E^C) in $[4f^7 + 4f^6 6p]$, $[4f^6 5d + 4f^6 6s]$ model spaces. Energies are given relative to the ground states $4f^7 \ ^8S_{7/2}$. Comparison with recommended NIST data (E^N) [20].

LSJ	Energies (cm^{-1})			LSJ	Energies (cm^{-1})			τ^C (sec)	LSJ	Energies (cm^{-1})			τ^C (sec)
	E^M	E^C	E^N		E^M	E^C	E^N			E^M	E^C	E^N	
	4f ⁷ states				4f ⁶ 5d states					4f ⁶ 5d states			
⁶ P _{3/2}	34182	30406		⁸ H _{3/2}	34103	33642	33856	1.503[1]	⁸ P _{5/2}	48111	41756	39769	1.638[−8]
⁶ P _{5/2}	33691	30001	28629	⁸ H _{5/2}	34800	34160	34394	1.221[0]	⁸ P _{7/2}	49410	42139	40871	1.485[−8]
⁶ P _{7/2}	33319	29581	28200	⁸ H _{7/2}	35722	34848	35109	4.196[−1]	⁸ P _{9/2}	50613	43423	42084	1.024[−8]
				⁸ H _{9/2}	36833	35680	35972	1.418[−1]					
⁶ I _{7/2}	34702	32295	31746	⁸ H _{11/2}	38101	36634	36962	4.524[−2]	⁶ P _{3/2}	43323	39747		6.317[−6]
⁶ I _{9/2}	34972	32560	31954	⁸ H _{13/2}	39512	37696	38067	1.472[−2]	⁶ P _{5/2}	45407	40763	40898	3.977[−8]
⁶ I _{11/2}	35217	32783	32180	⁸ H _{15/2}	41065	38865	38290	5.781[−3]	⁶ P _{7/2}	48088	43250	42530	4.040[−8]
⁶ I _{13/2}	35396	32921	32314	⁸ H _{17/2}	42784	40158	40659	4.292[−3]					
⁶ I _{15/2}	35465	32926	32308						⁶ H _{5/2}	48728	44055	43396	1.956[−6]
⁶ I _{17/2}	35352	32716	32073	⁸ D _{3/2}	37444	35902	35627	9.567[−5]	⁶ H _{7/2}	49463	44593	43885	1.837[−6]
				⁸ D _{5/2}	38726	37059		1.455[−5]	⁶ H _{9/2}	50336	45240	44554	1.671[−6]
⁶ D _{1/2}	39714	37159		⁸ D _{7/2}	40016	38129	38229	4.369[−6]	⁶ H _{11/2}	51311	45975	45313	1.483[−6]
⁶ D _{3/2}	40048	37342		⁸ D _{9/2}	41275	39101	39226	3.804[−6]	⁶ H _{13/2}	52362	46786	46150	1.271[−6]
⁶ D _{5/2}	40295	37457		⁸ D _{11/2}	42482	40017	40133	3.052[−1]	⁶ H _{15/2}	53495	47678	47069	1.051[−6]
⁶ D _{7/2}	40145	37275											
⁶ D _{9/2}	39231	36562		⁸ F _{1/2}	40362	38832		2.978[−3]	⁶ F _{1/2}	52135	48094		4.873[−6]
				⁸ F _{3/2}	40999	39334	39014	5.622[−4]	⁶ F _{3/2}	54591	48700		3.291[−6]
⁶ G _{3/2}	49215	47912		⁸ F _{5/2}	41813	39968	39636	7.318[−6]	⁶ F _{5/2}	55764	47434	46108	1.865[−6]
⁶ G _{5/2}	48780	47186		⁸ F _{7/2}	42741	40692	40372	1.259[−5]	⁶ F _{7/2}	56851	48188	46793	1.495[−6]
⁶ G _{7/2}	48539	46501		⁸ F _{9/2}	43751	41474	41150	3.811[−5]	⁶ F _{9/2}	57693	49130	47714	9.094[−7]
⁶ G _{9/2}	48578	47555		⁸ F _{11/2}	44824	42294	41988	9.703[−4]	⁶ F _{11/2}	59967	50735	49086	7.764[−6]
⁶ G _{11/2}	48743	47645		⁸ F _{13/2}	45957	43138	42850	4.093[−4]					
⁶ G _{13/2}	49859	49110							⁶ D _{1/2}	64852	46902		1.796[−6]
				⁸ G _{1/2}	39692	38440	38050	1.586[−3]	⁶ D _{3/2}	65882	46948		2.090[−6]
⁶ F _{1/2}	52339	49089		⁸ G _{3/2}	39989	38657	38337	9.284[−4]	⁶ D _{5/2}	65929	49445	48496	2.595[−6]
⁶ F _{3/2}	52865	51589		⁸ G _{5/2}	40642	39160	38829	1.417[−5]	⁶ D _{7/2}	66715	50206	49293	1.911[−6]
⁶ F _{5/2}	53356	51293		⁸ G _{7/2}	41583	39898	39580	1.581[−5]	⁶ D _{9/2}	67322	50846	49957	1.094[−6]
⁶ F _{7/2}	53699	51605		⁸ G _{9/2}	42747	40814	40518	4.568[−5]					
⁶ F _{9/2}	53744	51812		⁸ G _{11/2}	44073	41848	41572	3.865[−4]	⁶ G _{3/2}	52052	50504		8.288[−5]
⁶ F _{11/2}	53341	51165		⁸ G _{13/2}	45474	42926	42658	1.222[−4]	⁶ G _{5/2}	52902	51193	49906	2.662[−5]
				⁸ G _{15/2}	46832	43952	43658	3.452[−5]	⁶ G _{7/2}	54046	51896		1.733[−5]
									⁶ G _{9/2}	55329	52533		1.639[−5]
									⁶ G _{11/2}	57039	53055	51651	4.744[−5]
									⁶ G _{13/2}	60645	53464	52100	1.108[−4]

valence electrons) while there are also many-body corrections to $R_{4f,5d}$ due to the correlations between valence and core electrons. Since the ratio of experimental and calculated lifetimes is almost the same for all three 8P states (see Table I), it is natural to assume that the most of discrepancy comes from many-body corrections to $R_{4f,5d}$. Note, that the ratio of the experimental and calculated lifetimes is even more stable in the work of Mashonkina *et al.* [16]. The ratio is 3.0 ± 0.3 and the corresponding lifetimes are presented in Table I. Calculations in this work were also done with the Cowan code, however, the mixing of states was more carefully considered.

In the HF approximation, $R_{4f,5d} = 0.77a_B$. Now we calculate a correction to this value due to core polarization by the dipole electric field of the emitted photon. We do this in the

random-phase approximation (RPA) using the time-dependent Hartree-Fock method (TDHF) [17]. The TDHF equations can be written in the form

$$(\hat{H}_0 - \epsilon_i) \delta\psi_i = -(\hat{f} + \hat{\delta}V) \psi_i, \quad (4)$$

where \hat{H}_0 is HF Hamiltonian. The single-electron orbital ψ_i satisfies HF equation

$$(\hat{H}_0 - \epsilon_i) \psi_i = 0,$$

TABLE III. Energies (cm^{-1}) and lifetimes τ (sec) for Gd IV calculated by RCI (E^M) and Cowan codes (E^C). Energies are given relative to the ground states $4f^7 {}^8S_{7/2}$. Comparison with experimental data from Ref. [12] (E^{expt}).

LSJ	Energies (cm^{-1})			LSJ	Energies (cm^{-1})			τ^C (sec)	LSJ	Energies (cm^{-1})			τ^C (sec)
	E^M	E^C	E^{expt}		E^M	E^C	E^{expt}			E^M	E^C	E^{expt}	
	4f ⁷ states				4f ⁶ 5d states					4f ⁶ 5d states			
⁶ P _{3/2}	38308	34114	33262	⁸ H _{3/2}	92479	98073	3.364[−5]	⁶ P _{3/2}	103091	105613	104264	2.131[−8]	
⁶ P _{5/2}	37638	33577	32680	⁸ H _{5/2}	93338	98750	1.624[−5]	⁸ P _{5/2}	105108	106266	106493	1.696[−9]	
⁶ P _{7/2}	37103	33018	32084	⁸ H _{7/2}	94467	99641	9.024[−6]	⁶ P _{7/2}	107493	109547	109005	4.311[−9]	
				⁸ H _{9/2}	95821	100711	5.447[−6]						
⁶ I _{7/2}	38833	36109	35808	⁸ H _{11/2}	97368	101933	3.541[−6]	⁶ H _{5/2}	108974	110344		1.093[−8]	
⁶ I _{9/2}	39191	36468	36151	⁸ H _{13/2}	99097	103295	2.527[−6]	⁶ H _{7/2}	109790	110989		1.086[−8]	
⁶ I _{11/2}	39504	36766	36430	⁸ H _{15/2}	101027	104809	2.160[−6]	⁶ H _{9/2}	110760	111763		1.075[−8]	
⁶ I _{13/2}	39722	36950	36508	⁸ H _{17/2}	103239	106541	6.287[−6]	⁶ H _{11/2}	111838	112641		1.067[−8]	
⁶ I _{15/2}	39782	36957	36547					⁶ H _{13/2}	113003	113620		1.069[−8]	
⁶ I _{17/2}	39586	36677	36206	⁸ D _{3/2}	96222	100611	6.089[−8]	⁶ H _{15/2}	114270	114729		1.089[−8]	
				⁸ D _{5/2}	98072	102237	1.195[−7]						
⁶ D _{1/2}	44618	41738	40444	⁸ D _{7/2}	99762	103840	4.803[−7]	⁶ F _{1/2}	112718	115165		1.405[−8]	
⁶ D _{3/2}	45060	41983	40694	⁸ D _{9/2}	101204	105041	4.429[−7]	⁶ F _{3/2}	112551	115883		1.317[−8]	
⁶ D _{5/2}	45363	42131	40857	⁸ D _{11/2}	102467	106068	2.438[−6]	⁶ F _{5/2}	113476	114242	111745	1.068[−8]	
⁶ D _{7/2}	45120	41877	40599					⁶ F _{7/2}	114715	115140	113129	9.812[−9]	
⁶ D _{9/2}	43876	40934	39508	⁸ F _{1/2}	99582	103800	7.829[−7]	⁶ F _{9/2}	116120	116239	114214	9.076[−9]	
				⁸ F _{3/2}	100315	104908	8.558[−7]	⁶ F _{11/2}	118102	118166		1.166[−8]	
⁶ G _{3/2}	54999	53498	50633	⁸ F _{5/2}	101260	104774	1.958[−7]						
⁶ G _{5/2}	54398	52556	49825	⁸ F _{7/2}	102350	106595	1.884[−7]	⁶ D _{1/2}	114452	113648		1.207[−8]	
⁶ G _{7/2}	54061	51626	49526	⁸ F _{9/2}	103560	107573	7.487[−7]	⁶ D _{3/2}	115542	113646		1.196[−8]	
⁶ G _{9/2}	54132	53018	49652	⁸ F _{11/2}	104888	108599	2.167[−6]	⁶ D _{5/2}	116881	116788		1.229[−8]	
⁶ G _{11/2}	54277	53113	49652	⁸ F _{13/2}	106352	109718	4.888[−7]	⁶ D _{7/2}	118099	117691	116230	1.156[−8]	
⁶ G _{13/2}	55684	54908	51360					⁶ D _{9/2}	119010	118415	117229	1.059[−8]	
				⁸ G _{1/2}	98870	104285	1.263[−7]						
⁶ F _{1/2}	58661	55022		⁸ G _{3/2}	99263	104094	6.047[−7]	⁶ G _{3/2}	116779	117952		1.700[−8]	
⁶ F _{3/2}	59353	58025		⁸ G _{5/2}	100104	105690	8.230[−8]	⁶ G _{5/2}	118903	118862	118109	1.553[−8]	
⁶ F _{5/2}	59953	57566		⁸ G _{7/2}	101315	105781	2.170[−7]	⁶ G _{7/2}	119962	119714	119292	1.528[−8]	
⁶ F _{7/2}	60303	57909		⁸ G _{9/2}	102816	107028	5.390[−7]	⁶ G _{9/2}	120838	120449	120220	1.513[−8]	
⁶ F _{9/2}	60310	58200		⁸ G _{11/2}	104455	108380	5.352[−7]	⁶ G _{11/2}	121510	121003	121063	1.496[−8]	
⁶ F _{11/2}	59774	57330		⁸ G _{13/2}	106038	109641	2.946[−7]	⁶ G _{13/2}	122130	121368	121725	1.393[−8]	
				⁸ G _{15/2}	107581	110766	8.983[−8]						
⁴ N _{17/2}	60545	55382						⁶ G _{3/2}	118067	119462		7.208[−8]	
⁴ N _{19/2}	61512	56379		⁸ P _{5/2}	107010	107583	2.264[−9]	⁶ G _{5/2}	119183	121201		1.101[−7]	
⁴ N _{21/2}	62009	56524		⁸ P _{7/2}	108921	107524	1.180[−9]	⁶ G _{7/2}	122136	123606		1.262[−7]	
⁴ N _{23/2}	61817	56827		⁸ P _{9/2}	109318	108884	9.829[−10]	⁶ G _{9/2}	125622	126445		1.554[−7]	
								⁶ G _{11/2}	128024	129607		1.376[−7]	
								⁶ G _{13/2}	129323				

\hat{f} is the operator of the external electric field, $\delta\psi_i$ is a correction to the orbital ψ_i due to external field \hat{f} , and δV is the modification of the HF potential induced by corrections to the core states. Equations (4) are solved self-consistently for all core states. Note that since Eu III and Gd IV are open-shell systems, the same weighting procedure described in the preceding section must be applied to the left-handside and right-handside of Eq. (4). The transition amplitude between the states $4f$ and $5d$ in the RPA is

$$\langle 4f | \hat{f} + \delta V | 5d \rangle \quad (5)$$

(the HF approximation corresponds to $\delta V = 0$). Core polarization reduces the value of the $R_{4f,5d}$ radial integral bringing lifetimes into better agreement with experiment (see column “RPA” in Table I).

The remaining discrepancy should be attributed to correlations. A detailed investigation of correlations leads beyond the scope of the present work, but one should note that correlations increase the density of the external electron at short distances. Therefore, owing to normalization, it must decrease the density at large distances, thereby decreasing the value of the radial integral. Calculated lifetimes are also sensitive to mixing of states. Analysis of the RCI and Cowan

code calculations shows that the smallest mixing is for the $^8P_{9/2}$ state. Therefore, we can use this state to extract the value of $R_{4f,5d}$ that ensures the best fit of the experimental data. This value is $R_{4f,5d}=0.41a_B$ for Eu⁺². To calculate lifetimes which correspond to the “best fit” value of $R_{4f,5d}$, one need only multiply the results from the Cowan code by a factor of 3.6. In summary, the values of $R_{4f,5d}$ for Eu III and Gd IV are

	Eu III	Gd IV
HF	$0.77a_B$	$0.63a_B$
RPA	$0.56a_B$	$0.42a_B$
Best fit	$0.41a_B$	$0.34a_B$

IV. RESULTS

In Table II, we list and compare energies of $4f^7$ and $4f^65d$ states in Eu III calculated using the RCI code and the Cowan code. Energies are given relative to the ground state $4f^7\ ^8S_{7/2}$. As mentioned above, both codes permit us to obtain results that are generally in good agreement with experimental energies by scaling the electrostatic Slater parameters to simulate correlation effects (Refs. [18,19]). We use the scaling factor of 0.8 in RCI code and 0.85 in the Cowan code. Also the energies of the $4f^65dLSJ$ levels are shifted by $13\,500\text{ cm}^{-1}$ in the Cowan code relative to the ground state $4f^7\ ^8S_{7/2}$. In the RCI code we do not shift the energies but modify the $5d$ state as was described in Sec. II, to improve the energy interval between the $4f^7$ and $4f^65d$ configurations. In Table II, energies of the $4f^7\ LSJ$, $4f^65dLSJ$, and $4f^66sLSJ$ levels in Eu III are compared with recommended data from the National Institute for Standards and Technology (NIST) by Martin *et al.* [20]. The 105 levels obtained from spectral analysis by Sugar and Spector [21] given in the NIST publication, classify about 300 of the observed lines. It should be noted that the spectral analysis in Ref. [21] was based on the Cowan code, probably, a simpler version than we use here.

It should be noted that we use different coupling schemes in the RCI (jj coupling) and Cowan codes (LS coupling) to build energy matrices. We use, for convenience, LS coupling labeling of states; however, neither jj nor LS coupling can describe the physical states properly. To combine together our results obtained with different coupling schemes, we calculated Landé g factors for each level. For low-lying states, the g factors are very close to their nonrelativistic values and identification of levels is easy. However, higher in the spectrum, strong mixing between states makes level identification difficult. We restricted our calculations to levels which are reliably identified in both calculations.

As can be seen from Table II that results of both calculations for Eu III are in good agreement with one another and with experiment. This gives us confidence in similar calculations for Gd IV.

The RCI calculations for Gd IV are done in exactly the same way as for Eu III. All fitting parameters were chosen for Eu III and no *a priori* data on Gd IV was used in the calculations. In a sense, we can say that the Gd IV calculations are predictive. They produce an energy spectrum of Gd IV re-

gardless of what is known about it. In contrast, the calculations with the Cowan code are not exactly the same for both ions. While we use the same scaling factor for the Coulomb integrals (0.85), the energy shift for the $4f^65d$ configuration is larger for Gd IV ($18\,000\text{ cm}^{-1}$) than for Eu ($13\,500\text{ cm}^{-1}$). A larger energy shift is needed to obtain good agreement with available experimental data.

In Table III, we compare energies of the $4f^7\ LSJ$ and $4f^65dLSJ$ levels with available experimental data and predicted data given by Kielkopf and Crosswhite [12]. It can be seen from Table III that for the $4f^7$ configuration the energies obtained by Cowan code E^C are in better agreement with energies from Ref. [12] than are energies obtained by RCI code E^M . However, for the $4f^65d$ configuration, results of both calculations are in very good agreement with each other and with [12].

In Tables (II and III), we present lifetimes of the $4f^65dLSJ$ levels calculated using the Cowan code with the HF value of the $R_{4f,5d}$ radial integral. To get more accurate predictions for the lifetimes one should multiply the values presented in tables by the factor of 3.6 (see Sec. I).

V. CONCLUSION

In a recent work on calculation of the EDM enhancement factor (K) in Gd IV [9], the result was presented in a form of two different numbers: $K_A \approx -6.4$ and $K_B \approx -3.3$. These two numbers were based on different assumptions about the energy splitting between $4f$ and $5d$ states of Gd IV. The first number (K_A) corresponds to $E_{5d} - E_{4f} \approx 40\,000\text{ cm}^{-1}$ which is a result of extrapolation from Eu III. The second number (K_B) corresponds to $E_{5d} - E_{4f} \approx 100\,000\text{ cm}^{-1}$ which is based on available experimental data for Gd IV (too incomplete at that time to be fully trusted).

The present work clearly indicates that the correct energy splitting is closer to $100\,000\text{ cm}^{-1}$ and consequently, the enhancement factor is rather -3.3 .

Furthermore, an analysis of lifetimes of Eu III suggests that core polarization by the electric field of an external photon is an important effect for both ions, Eu III and Gd IV. It reduces the value of the $R_{4f,5d}$ radial integral by a factor of about 1.5. This effect was not included in the calculation of the EDM enhancement factor (K_{EDM}) [9]. Only contributions proportional to the $R_{4f,5d}$ radial integral were considered in that work. To include core polarization by the electric field, one should divide the final answer of Ref. [9] by the factor of 1.5. This leaves us with $K_{\text{EDM}} = -2.2(-3.3/1.5)$.

ACKNOWLEDGMENTS

One of the authors (V.A.D.) is grateful to the Physics Department of the University of Notre Dame for the hospitality and support during his visit. The work of W.R.J. was supported in part by National Science Foundation Grant No. PHY-01-39928. U.I.S. acknowledges partial support by Grant No. B516165 from Lawrence Livermore National Laboratory.

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