Theoretical study of some experimentally relevant states of dysprosium

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Configuration interaction method is used to calculate transition amplitudes and other properties of the low states of dysprosium which are used in cooling and in the study of the time variation of the fine structure constant and violation of fundamental symmetries. The branching ratio for the cooling state to decay to states other than ground states is found to be smaller than 10^{-4}. The matrix element of the weak interaction between degenerate states at $E = 19797.96$ cm^{-1} is about 4 Hz which is consistent with the experimental limit $|H_W| = |2.3 \pm 2.9(\text{stat.}) \pm 0.7(\text{syst.})|$ Hz [A. T. Nguyen, D. Budker, D. DeMille, and M. Zolotorev, Phys. Rev. A **56**, 3453 (1997)] and points to feasibility of its experimental measurement. Applications include the search for physics beyond the standard model using the parity nonconservation (PNC) isotopic chain approach.

I. INTRODUCTION

Atomic dysprosium is proved to be a valuable object to study fundamental problems of modern physics. It has been recently used in search for the time variation of the fine structure constant [1], study of possible variation of the fine structure constant due to variation of the gravitation field [2], measurement of the parity nonconservation (PNC) [3], etc. Recent progress in trapping and cooling of dysprosium atoms [4–6] opens new exciting possibilities.

The most interesting feature of dysprosium which has inspired its use in the study of PNC and variation of the fine structure constant is the existence of two almost degenerate states of the same total momentum and opposite parity. Energy interval is so small that its actual value is determined by hyperfine structure and isotope shift. This leads to strong enhancement of both effects. The states are at the energy $E = 19797.96$ cm^{-1} and both have total momentum $J = 10$. Following Ref. [7] we use notation A for the even state and notation B for the odd state.

Another state of great interest is the odd state at $E = 23736.60$ cm^{-1} which is used in cooling. We use notation C for this state. Our present work mostly focuses on these three states while some other states are also considered.

The use of the rare-earth atoms, including dysprosium, in atomic PNC study was first suggested in Ref. [8], and for the search of the time variation of the fundamental constants in Refs. [9,10]. Dysprosium was studied theoretically in our previous works [11–13]. The work of Refs. [12,13] links the change of frequency of the transition between states A and B to the time variation of the fine structure constant. It was used in Refs. [1,2] for the interpretation of the measurements. In Ref. [11] we calculated the matrix element of the spin-independent parity-violating weak interaction between states A and B.

Dysprosium has many stable isotopes and is a good candidate to study PNC ratio for isotope chains. Such study may reveal important information on the physics beyond the standard model [14]. Accurate atomic calculations of the PNC effect are not needed for this study. However, reliable estimation of the effect is important to determine the feasibility of the measurements. The result of our previous calculations is $\langle A|W|B \rangle = 70(40)$ Hz [11]. Later measurements [3] lead to the

limit $\langle A|W|B \rangle = 2.3 \pm 2.9(\text{stat.}) \pm 0.7(\text{syst.})$. Although the measured value is not in strong disagreement with theoretical prediction, given the large uncertainty of the latter, the experiment did not confirm the large PNC effect that was hoped for in this system.

In present paper we revisited the PNC calculations and found that inclusion of more configurations pull the value of the weak matrix element down to about 4 Hz which is consistent with the measurements. The small value of the matrix element is the result of strong cancellation of different contributions. Dominant contributions are larger than the final result by more than one order of magnitude. This means that further cancellation to even smaller number is highly unlikely and the measurements might be possible on about the same level of sensitivity which has been already achieved in Ref. [3].

Another motivation for this work is due to dysprosium cooling at Berkeley [4,6] and Urbana [5]. We study the cooling state C to find the branching ratio of the transitions from this state to the states other than the ground state and to the ground state. High value ($\gg 10^{-4}$) for this ratio would be a problem for cooling. Our present calculations show that the branching ratio is in fact smaller than 10^{-4}.

II. METHOD

In present work we use the version of the configuration interaction (CI) method which was first developed for iron atom [15] and then used for other many-electron atoms including dysprosium [13]. See these works for the detailed discussion.

The effective Hamiltonian for N_v valence electrons ($N_v = 12$ for dysprosium) has the form

$$\hat{H}^{\text{eff}} = \sum_{i=1}^{N_v} \hat{h}_{1i} + \sum_{i<j}^{N_v} e^2/r_{ij}, \tag{1}$$

$\hat{h}_1(r_i)$ is the one-electron part of the Hamiltonian

$$\hat{h}_1 = c\boldsymbol{\alpha} \cdot \mathbf{p} + (\beta - 1)mc^2 - \frac{Ze^2}{r} + V_{\text{core}} + \delta V. \tag{2}$$

Here α and β are Dirac matrices, V_{core} is Hartree-Fock potential due to core electrons and δV is the term which simulates the

TABLE I. Configurations and effective core polarizabilities α_p (a.u.) used in the calculations.

N	Parity	Configuration	α_p
1	Even	$4f^{10}6s^2$	0.4
2	Even	$4f^{10}6s5d$	0.4006
3	Even	$4f^96s^26p$	0.4039
4	Even	$4f^95d6s6p$	0.389
5	Even	$4f^{10}6p^2$	0.4
6	Even	$4f^95d^26p$	0.4
7	Odd	$4f^95d^26s$	0.3947
8	Odd	$4f^95d6s^2$	0.3994
9	Odd	$4f^{10}6s6p$	0.397
10	Odd	$4f^{10}5d6p$	0.4
11	Odd	$4f^95d6p^2$	0.4
12	Odd	$4f^96s6p^2$	0.4

effect of the correlations between core and valence electrons. It is often called *polarization potential* and has the form

$$\delta V = -\frac{\alpha_p}{2(r^4 + a^4)}. \quad (3)$$

Here α_p is polarization of the core and a is a cut-off parameter (we use $a = a_B$).

Table I lists configurations considered in present work. The self-consistent Hartree-Fock procedure is done for every configuration separately. Then valence states found in the Hartree-Fock calculations are used as basis states for the CI calculations. It is important for the CI method that the atomic core remains the same for all configurations. We use the core which corresponds to the ground state configuration. Change in the core due to change of the valence state is small and can be neglected. This is because core states are not sensitive to the potential from the electrons which are on large distances (like $6s$, $6p$, and $5d$ electrons). The $4f$ electrons are on smaller distances and have larger effect on atomic core. However, in all the cases (see Table I) only one among about ten $4f$

electrons changes its state. Therefore their effect on atomic core is also small. More detailed discussion on the effect of valence electrons on atomic core can be found in Refs. [16,17].

The form of the δV in (3) is chosen to coincide with the standard polarization potential on large distances ($-\alpha_p/2r^4$). We treat α_p as fitting parameters. The values of α_p for each configuration of interest are presented in Table I. They are chosen to fit the experimental position of the configurations relative to each other. For all configurations the values of α_p are very close. This is not a surprise since the core is the same for every configuration. Small difference in α_p for different configurations simulates the effect of incompleteness of the basis and other imperfections in the calculations.

To calculate electric dipole transition amplitudes, lifetimes, magnetic dipole, and electric quadrupole hyperfine structure constants we use the time-dependent Hartree-Fock method (equivalent of the random phase approximation) combined with the CI technique, see Ref. [18] for a detailed discussion.

III. RESULTS AND DISCUSSION

Table II presents the results of calculations for the energies, g-factors, magnetic dipole, and electric quadrupole hyperfine structure constants and lifetimes of some low states of dysprosium. This includes the degenerate states A and B, cooling state C, and other odd states D to G which may also present an interest for cooling and quantum information processing [19]. Calculated values are compared with available experimental data. Good agreement for the energies is mostly due to the fitting. Theoretical uncertainty for the hyperfine structure constants is on the level of 20 to 30%. The uncertainty for lifetimes is determined by uncertainties for electric dipole transition amplitudes. The uncertainties for the amplitudes is also on the level of 20 to 30% with the exception of the extremely small amplitudes ($\ll 1$ a.u.) where uncertainty might be higher. The amplitudes are presented in Table III. Maximum

TABLE II. Experimental and theoretical energies, g-factors, hyperfine structure constants, and lifetimes for some low-lying states of ^{163}Dy. For theoretical uncertainties see discussion in the text.

N	leading config.	Term	J	Energies (cm^{-1})		g-factors		A (MHz)		B (MHz)		Lifetime	
				Expt.[a]	Calc.	Expt.[a]	Calc.	Expt.	Calc.	Expt.	Calc.	Expt.	Calc.
						States of special interest							
g.s.[b]	$4f^{10}6s^2$	^5I	8	0.00	0	1.24	1.24	163[c]	160	1153[c]	1193		
A	$4f^{10}5d6s$	$^3[10]$	10	19798	19786	1.21	1.21	159[d]	140	1865[d]	1629	7.9 μs[d]	16 μs
B	$4f^95d^26s$	$^7H^o$	10	19798	19770	1.367	1.368	218[d]	202	2060[d]	2413	>200 μs[d]	0.14 s
C	$4f^{10}6s6p$	$(8,1)^o$	9	23737	25200	1.22	1.22	122[e]	136	1842[e]	2096	4.8 ns[d]	4.7 ns
						Some other states							
D	$4f^95d6s^2$	$^7H^o$	8	7566	7563	1.35	1.35		131		1901		6.9 ms
E	$4f^95d6s^2$	$^7I^o$	9	9991	9944	1.32	1.32		125		2901		3 ms
F	$4f^95d6s^2$	$^5K^o$	9	13496	14634	1.23	1.23		144		4000		21 μs
G	$4f^{10}6s6p$	$(8,2)^o$	9	17727	18092	1.25	1.26		194		1582	2 μs[d]	2.9 μs

[a]Reference [20].
[b]Ground state.
[c]References [21–23].
[d]Reference [7].
[e]Reference [24].

TABLE III. Decay channels for states A–G from Table II.

Upper state	Lower state Config.	Lower state Term	J	Energy (cm^{-1})	ω (cm^{-1})	$\lvert\langle f\lVert\mathbf{D}\rVert i\rangle\rvert$ (a.u.)	Rate (s^{-1})
A	$4f^9 5d6s^2$	$^7K^o$	10	12893	6905	0.056	0.982×10^2
	$4f^{10}6s6p$	$(8,2)^o$	10	17513	2285	0.276	0.874×10^2
	$4f^9 5d6s^2$	$^7I^o$	9	9991	9807	0.049	0.218×10^3
	$4f^9 5d6s^2$	$^5K^o$	9	13496	6302	0.192	0.887×10^3
	$4f^{10}6s6p$	$(8,1)^o$	9	15972	3826	2.34	0.296×10^5
	$4f^9 5d6s^2$	$^7K^o$	9	16717	3080	0.441	0.547×10^3
	$4f^{10}6s6p$	$(8,2)^o$	9	17727	2071	5.86	0.294×10^5
B	$4f^{10}5d6s$	$^3[9]$	10	18463	1335	0.069	0.109×10^1
	$4f^{10}5d6s$	$^3[8]$	9	17515	2283	0.073	0.608×10^1
	$4f^{10}5d6s$	$^3[9]$	9	19241	557	0.0045	0.339×10^{-3}
C	$4f^{10}6s^2$	^5I	8	0	23737	12.28	0.215×10^9
	$4f^{10}5d6s$	$^3[7]$	8	17613	6123	0.221	0.120×10^4
	$4f^{10}5d6s$	$^3[8]$	8	18903	4833	0.265	0.845×10^3
	$4f^{10}6s^2$	$^3K2$	8	19019	4717	0.057	0.362×10^2
	$4f^{10}5d6s$	$^3[9]$	8	20194	3543	0.036	0.601×10^1
	$4f^9 6s6p$	$(^{15}/_2,^1/_2)$	8	20790	2947	0.099	0.265×10^2
	$4f^{10}5d6s$	$^3[7]$	8	21603	2134	0.226	0.530×10^2
	$4f^{10}5d6s$	$^3[8]$	9	17515	6222	0.411	0.434×10^4
	$4f^{10}5d6s$	$^3[9]$	9	19241	4496	0.122	0.144×10^3
	$4f^{10}5d6s$	$^3[10]$	9	20209	3528	0.222	0.231×10^3
	$4f^{10}5d6s$	$^1[9]$	9	22046	1691	0.659	0.224×10^3
	$4f^{10}5d6s$		9	23218	518	0.0076	0.846×10^3
	$4f^{10}5d6s$	$^3[9]$	10	18462	5274	0.502	0.394×10^4
	$4f^{10}5d6s$	$^3[10]$	10	19798	3939	0.467	0.142×10^4
	$4f^{10}5d6s$	$^1[10]$	10	22487	1249	1.039	0.225×10^3
D	$4f^{10}6s^2$	^5I	8	0	7566	0.053	0.146×10^3
	$4f^{10}6s^2$	^5I	7	4134	3431	0.0017	0.136×10^{-1}
E	$4f^{10}6s^2$	^5I	8	0	9991	0.059	0.369×10^3
F	$4f^{10}6s^2$	^5I	8	0	13496	0.424	0.471×10^5
G	$4f^{10}6s^2$	^5I	8	0	17727	0.897	0.478×10^6

disagreement between theory and experiment in Table II is for the lifetime of the state A where calculated value is two times larger than the experimental one. The lifetimes of the most of the states are sensitive to the mixing of the $4f^{10}6s6p$ with other odd configurations. For example, the experimental lifetime of the state C is reproduced in the calculations when state C is a pure state of the $4f^{10}6s6p$ configuration with very little admixture of other configurations. This has been achieved by properly choosing the fitting parameters α_p (see Table I).

Table III shows decay channels for the states listed in Table II. The data are based on the calculated electric dipole transition amplitudes and experimental energies. No electric quadrupole or magnetic dipole amplitudes were taken into account. Test calculations show that their contribution is negligible. The analysis of the data for state A shows that it decays mostly to two states of the $4f^{10}6s6p$ configuration. This is because the leading configuration of state A is the $4f^{10}6s5d$ configuration and an electric dipole transition between the two states can be reduced to the allowed single-electron $6p \rightarrow 5d$ transition. In contrast, transitions to the states of the $4f^9 5d6s^2$ configuration can only go via configuration mixing. The calculated lifetime of the state A is larger than the experiment. This means that the transition amplitudes are smaller. It is

unlikely that the amplitudes of the transitions between state A and states of the $4f^{10}6s6p$ configuration are underestimated. This is because the latter states are almost pure, with only about 3% admixture of other configurations. It is more likely that some of the other amplitudes are underestimated because of too small mixture of the $4f^5 5d6s^2$ and $4f^{10}6s6p$ configurations.

The analysis of the decay channels of the cooling level C (see Table III) also indicates the sensitivity of the data to the mixing of the $4f^5 5d6s^2$ and $4f^{10}6s6p$ configurations. Good agreement with experiment for the lifetime of the state C can only be achieved if this state is a pure state of the $4f^{10}6s6p$ configuration. On the other hand, this state is very close to the states of the $4f^5 5d6s^2$ configuration. Manipulating with the fitting parameters α_p (Table I) can easily lead to a situation when the states of the two configurations are strongly mixed and the lifetime of the state C is larger than the experiment. Therefore, the lifetime of this state should be monitored in the fitting process.

The data in Table III allows to estimate the ratio of the following probabilities: (a) decay of state C to all lower state other than the ground state, and (b) decay of the state C into the ground state. This ratio is $(6 \pm 3) \times 10^{-5}$. The fraction of atoms lost into metastable states must be even smaller because the number above includes all channels of decay

TABLE IV. Contributions to the electric dipole transition amplitude between states A and B (a.u.).

Configurations		Single-electron	Partial	Sum
State A	State B	matrix element	contribution	(a.u.)
$4f^{10}5d6s$	$4f^95d^26s$	$\langle 4f_{5/2}|D|5d_{3/2}\rangle$	-0.2634	-0.2634
$4f^{10}5d6s$	$4f^95d^26s$	$\langle 4f_{5/2}|D|5d_{5/2}\rangle$	-0.0106	-0.2740
$4f^{10}5d6s$	$4f^95d^26s$	$\langle 4f_{7/2}|D|5d_{5/2}\rangle$	0.2723	-0.0017

without considering which of them end up in a metastable state. However, some lower states quickly decay to the ground state and do not produce any losses. A more detailed analysis would require a lot of extra work, however it is not needed since the branching ratio $<10^{-4}$ is sufficiently good for the cooling [5].

Table III also shows the data for states D–G which might be useful for cooling and information processing [19].

Our calculated value for the electric dipole reduced matric element between states A and B is 0.024 ± 0.010 a.u. This agrees well with the experimental value of $0.015(1)$ a.u. from Ref. [7]. Note that this amplitude is zero in the nonrelativistic limit. This is because states A and B have different spin (see Table II) and electric dipole operator cannot change it. In relativistic calculations the amplitude is not zero. However, it is small and this small value is the result of strong cancellations between different contributions. Table IV shows the largest in absolute values contributions to the electric dipole transition amplitude between states A and B. All these contributions are due to the $4f - 5d$ electric dipole transitions within the main configurations of states A and B. The sum is only 0.0017 a.u. which is more than an order of magnitude smaller than the final answer. The final amplitude is the sum of many smaller contributions which contain all possible single-electron transitions.

The situation is even more complicated for the matrix element of the weak interaction between states A and B. Table V shows dominant contributions to this matrix element. There are strong cancellations between different contributions, mostly between terms containing the $\langle 4f_{5/2}|H_W|5d_{5/2}\rangle$ and $\langle 6s_{1/2}|H_W|6p_{1/2}\rangle$ single-electron matrix elements. The former of these matrix elements is small. It is not zero due to the effect of core polarization by the weak interaction: weak interaction acts on s and p core electrons changing the self-consistent Hartree-Fock potential which in turn leads to the $4f$-$5d$ transition between valence states. The $\langle 6s_{1/2}|H_W|6p_{1/2}\rangle$ inte-

TABLE V. Contributions to the matrix element of the weak interaction between states A and B (Hz).

Configurations		Single-electron	Partial	Sum
Left	Right	matrix element	contribution	(Hz)
$4f^{10}5d6s$	$4f^95d^26s$	$\langle 4f_{5/2}|H_W|5d_{5/2}\rangle$	38.79	38.79
$4f^{10}5d6s$	$4f^{10}5d6p$	$\langle 6s_{1/2}|H_W|6p_{1/2}\rangle$	-49.04	-10.25
$4f^{10}5d6s$	$4f^{10}6s6p$	$\langle 5d_{3/2}|H_W|6p_{3/2}\rangle$	1.60	-8.65
$4f^95d6s6p$	$4f^95d6s^2$	$\langle 6p_{1/2}|H_W|6s_{1/2}\rangle$	-1.35	-10.00
$4f^95d6s6p$	$4f^95d^26s$	$\langle 6p_{3/2}|H_W|5d_{3/2}\rangle$	-4.28	-14.27
$4f^95d^26p$	$4f^95d^26s$	$\langle 6p_{1/2}|H_W|6s_{1/2}\rangle$	11.84	-2.44

gral is not small but its contribution is suppressed by small admixture of the configurations containing the $6s$ and $6p$ states to the main configurations. This makes the result very sensitive to configuration mixing. We found that inclusion of all configurations listed in Table I is important for the weak matrix element. The effect of some configurations is indirect, via changing the coefficients of configuration mixing. For example, the sum of all largest contributions listed in Table V, -2.44 Hz is very close to the final answer -2.13 Hz. This may make an impression that the configurations not listed in Table V do not contribute. However, if, e.g., the $4f^96s^26p$ configuration is not included, the weak matrix element becomes larger by more than four times. This configuration was missed in our previous calculations [11] which is probably one of the reasons of disagreement between theory and experiment. The result of Ref. [11] corresponds to the situation when the weak matrix element between states A and B is dominated by the contribution of the $\langle 4f_{5/2}|H_W|5d_{5/2}\rangle$ single-electron matrix element (first line of Table V). This is in spite of the fact that most of the other configurations were also included. Probably incomplete inclusion of other configurations leads to underestimation of the configuration mixing which suppresses the contributions containing other single-electron matrix elements. This is supported by the results of another test. A small change in the fitting procedure, which has little effect on all values calculated in present paper, have significant effect on the weak matrix element between states A and B increasing it from 2 Hz to 6 Hz. Similar change happens when Breit or QED corrections are included (see discussion below). Therefore, we put the final value as $|H_W| = 4 \pm 4$ Hz.

This result for the weak matrix element is consistent with the experimental value of $|\langle A|H_W|B\rangle| = 2.3 \pm 2.9(\text{stat.}) \pm 0.7(\text{syst.})|$ [3]. This is about ten times smaller than the values of individual contributions (see Table V). Since there is no law of physics to make this matrix element exactly zero we believe that any further cancellation which would make the result even smaller than 4 Hz is highly unlikely. This probably means that the measurements of the PNC effect can be done on about the same level of sensitivity which has been already achieved in Ref. [3].

In the end we would like to discuss the effect of Breit and quantum electrodynamic (QED) corrections on the values calculated in the present paper. These effects were studied in detail for such atoms as Cs, Tl, Fr, etc., in a number of recent publications [25–32]. Direct rescaling from the studied atoms to dysprosium suggests that the effect should be small: about 1% for the weak matrix elements and the hyperfine structure constants and even smaller for the transition amplitudes and the energies. However, direct rescaling is not reliable for an atom like dysprosium which have more complicated electron structure than atoms studied in cited works.

Therefore, we perform full-scale tests of the effects of the Breit and QED corrections in dysprosium on the results listed above. For Breit interaction we use the technique developed in Ref. [31]. We replace Coulomb interaction on every stage of the calculations by the sum of the Coulomb and Breit interaction. This also includes the CI calculations which were absent in our previous work. For the QED corrections we use the QED potential approach developed in Ref. [32].

We found that the effect of Breit interaction is indeed often larger than those given by rescaling from other atoms. However it is always within the uncertainty of the calculations. For example, the effect of Breit interaction on the E1 transition amplitudes varies from 0.5 to 15% being large for small amplitudes. Note that uncertainty due to correlations are also large for small amplitudes and usually it is significantly larger than 15%.

The largest effect of the Breit interaction is on the weak matrix element between states A and B. For the choice of the fitting parameters which give the value of 6 Hz for this matrix element the inclusion of Breit interaction reduces this value to 1.5 Hz. However, as it was discussed above, small variation of the fitting parameters changes the value of this matrix element from 2 to 6 Hz. Therefore, here again we can say that the effect of Breit is within the uncertainty of the calculations.

The effect of QED corrections is always small. Even for the extremely unstable weak matrix element between states A and B the QED correction is only 15%. It is much smaller for all other vales calculated in present paper.

In the end we conclude that the main source of the uncertainty of present calculations is incomplete treatment of the correlations. Breit and QED corrections give noticeable contributions only in cases were the uncertainty is very high due to instability of the results.

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