Fine Structure Anomalies and Search for Variation of the Fine Structure Constant in Laboratory Experiments

V. A. Dzuba* and V. V. Flambaum†

1 School of Physics, University of New South Wales, Sydney 2052, Australia
2 Physics Division, Argonne National Laboratory, Argonne, Illinois 60439-4843, USA
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Configuration interaction in many-electron atoms may cause anomalies in the fine structure which make the intervals small and very sensitive to variation of the fine structure constant. Repeated precision measurements of these intervals over long period of time can put strong constrain on possible time variation of the fine structure constant. We consider the $5p^4 \ 3P_{2,1,0}$ fine structure multiplet in the ground state of neutral tellurium as an example. Here the effect of change of the fine structure constant is enhanced about one hundred times in the relative change of the small energy interval between the $^3P_1$ and $^3P_0$ states.

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I. INTRODUCTION

The possibility of fundamental constants to vary is predicted by theories unifying gravity with other interactions (see, e.g. review by Uzan [1]). There is an evidence found in quasar absorption spectra that the fine structure constant $\alpha$ ($\alpha = e^2/hc$) might be smaller in early cosmological epoch [2, 3, 4, 5]. However, similar analysis applied by other authors to different sets of data does not support this claim [7]. Recent progress in improving atomic clocks and developing optical frequency standards (see, e.g. [8]) made it possible to put strong constrains on possible time variation of fundamental constants in laboratory measurements. In particular, constrains obtained for the fine structure constant $\alpha$ need only about one order of magnitude improvement to see whether they are consistent with the quasar absorption spectra data if the same rate of change is assumed over cosmological time. These constrains were obtained with the full use of the technique developed for highly accurate optical standards. However, the choice of atomic transitions was not optimal for searching of variation of the fine structure constant. Frequencies used so far change at about the same rate as $\alpha$ if $\alpha$ changes, i.e. there is no enhancement.

An alternative approach was suggested in Refs. [2, 10]. It was proposed to measure small frequency between two close states with different dependence on $\alpha$. Here small change of $\alpha$ may lead to orders of magnitude larger relative change in frequency. The enhancement factor $k$ can be expressed as $k = 2q_{12}/\omega$, where $q_{12}$ is the difference in relativistic energy shifts of two levels and $\omega$ is transition frequency. A good candidate for such experiments is dysprosium atom [9, 10, 11]. It has two almost degenerate states of opposite parity and corresponding enhancement factor is of order of $10^3$ [10]. The measurements for dysprosium are currently underway at Berkeley [11].

While it is hard to find anything better than dysprosium in terms of enhancement factor, it has its disadvantages too. The levels involved are not so narrow. While one of the states lives long enough to cause no problem the other level does not. One has to work inside the line width to have the desirable accuracy. This puts certain limitations on achievable constrains on $\alpha$-variation. Therefore, it would be important to find something which combines the advantages of having metastable states and strong enhancement. A number of such transitions were considered in recent work [12]. It has been suggested to look at close metastable states of different configurations. Energies of different configurations change at different rates when $\alpha$ is changing. This, together with small energy intervals between the states ensures strong enhancement [12].

In present work we consider slightly different possibility. We suggest to study anomalously small fine structure intervals in ground configurations of many-electron atoms. The use of the ground state fine structure multiplet ensures that states are metastable. They can only decay via $M1$-transitions which are very much suppressed due to small value of transition frequency. On the other hand, configuration interaction in many-electron atoms can reduce fine structure intervals and lead to strong sensitivity to the change in the fine structure constant. In next sections we consider in detail fine structure of the ground state of tellurium and discuss some other possibilities.

II. THEORY AND RESULTS FOR TELLURIUM

To study sensitivity of atomic frequencies to variation of the fine structure constant $\alpha$ it is convenient to present them in the vicinity of the physical value of $\alpha$ ($\alpha = \alpha_0 = 1/137.036$) in the form

$$\epsilon = \epsilon_0 + q\alpha,$$

(1)
where \( x = (\alpha/\alpha_0)^2 - 1 \) and coefficient \( q \) defines the sensitivity of the frequency to variation of \( \alpha \). In general, its value can be found from atomic calculations. If \( \alpha \) changes, the relative frequency changes at the rate

\[
\frac{\Delta \omega}{\omega} = \frac{2q_{12} \Delta \alpha}{\omega_0 \alpha_0} = k \frac{\Delta \alpha}{\alpha_0},
\]

where \( k = 2q_{12}/\omega_0 \) is enhancement factor. To search for variation of the fine structure constant one needs to compare atomic frequencies with different enhancement factors over long period of time. The larger this difference the more sensitive the experiment to variation of \( \alpha \). Note that \( k \) can have negative value which means that changes of \( \alpha \) and frequency go in opposite directions: frequency decreases when \( \alpha \) increases and vise versa.

For “normal” fine structure intervals which are proportional to \((Z\alpha)^2\) formula (1) is valid for all values of \( \alpha \), \( 0 < \alpha < \alpha_0 \). Therefore, \( q_{12} = \omega_0 \) and \( k = 2 \). In other words, the factor \( k \) is the same for all “normal” fine structure intervals and comparison between them cannot reveal any drift of \( \alpha \). These fine structure intervals can still be used in search for variation of the fundamental constants if they are compared to hyperfine structure or to frequencies of suitable optical transitions.

Situation changes dramatically if fine structure multiplet is strongly perturbed by configuration interaction with neighboring states. We consider neutral tellurium atom in its ground state as an example.

Experimental and theoretical energies of the ground state \( 5p^4 \) configuration of tellurium are presented in Table I. There are strong anomalies in the fine structure multiplet \( ^3P_{2,1,0} \). The \(^3P_2 - ^3P_1 \) and \(^3P_1 - ^3P_0 \) intervals have opposite signs and differ in value more than hundred times. This is because of configuration interaction between the \(^3P_3 \) and \(^1D_2 \) states and between the \(^3P_0 \) and \(^1S_0 \) states while the \(^3P_1 \) state has no close neighbors to mix with. Configuration interaction leads to accidental almost exact cancellation between spin-orbit and Coulomb terms in the energy interval between the \(^3P_1 \) and \(^3P_0 \) states. Since spin-orbit interaction is much more sensitive to the change of \( \alpha \) than the Coulomb term, it is natural to expect that the \(^3P_1 - ^3P_0 \) energy interval is very sensitive to the variation of \( \alpha \).

To check how strong is sensitivity of the fine structure intervals of Te I we perform model configuration interaction (CI) calculations for the \( 5p^4 \) configuration of the atom. First, we perform Hartree-Fock calculations for open shells to find the \( 5p_{1/2} \) and \( 5p_{3/2} \) states of neutral tellurium. Then we use the CI technique to construct four-electron states of the \( 5p^4 \) configuration and to calculate their energies (in fact, in this approximation CI technique is reduced to diagonalization of the interaction Hamiltonian describing direct mixing of different \( 5p^4 \) states). It turns out that some extra fitting is needed to have good agreement with experiment. Namely, we reduce the value of the \( F_2(5p_{3/2}, 5p_{3/2}) \) Coulomb integral by 25%. This reduction simulates the effect of screening of Coulomb interaction between valence electrons by core electrons. The results for energies are presented in Table II. One can see that in spite of simple approximation used in calculations the agreement with experiment is very good.

Coefficients \( q \) (see Eq.(1)) are found by varying \( \alpha \) in computer codes:

\[
q = \frac{\epsilon(x = +0.1) - \epsilon(x = -0.1)}{0.2}.
\]

The results for \( q \) are also presented in Table II. Since we have good agreement with experiment for the energies it is natural to assume that the accuracy for the \( q \)-coefficients is also good.

Table II presents frequencies and enhancement factors for transitions between all states of the \(^3P_{0,1,2} \) fine structure multiplet. The enhancement for the \(^3P_1 - ^3P_0 \) is more than one hundred due to anomalously small frequency of the transition. Ratio of this small frequency to almost any other atomic frequency is extremely sensitive to variation of \( \alpha \). However, other transitions from Table II can also be used.

Because of very exotic behavior of the fine structure intervals of Te I it is very interesting to see what happens to them when \( \alpha \) varies from zero (non-relativistic limit) to its physical value \( \alpha_0 \). We have performed such calculations and results for five low states of the \( 5p^4 \) ground-state configuration of Te I are presented in Fig. III. Valence energies (energy to remove all four valence electrons from atom) are shown as functions of \((\alpha/\alpha_0)^2\). All three energies of the \(^3P_{0,1,2} \) multiplet have the same value at \( \alpha = 0 \) and fine structure intervals are proportional to \( \alpha^2 \) at small \( \alpha \). At larger values of \( \alpha \) interaction with the \(^1D_2 \) and \(^1S_0 \) leads to significant perturbation of the \(^3P \) multiplet. In particular, repulsion between the \(^1S_0 \) and \(^3P_0 \) levels causes the latter to cross with the \(^3P_1 \) level in the vicinity of the physical value of \( \alpha \). This explains anomalous behavior of the fine structure intervals of Te I.
Let us mention in the end of this section that both upper states of the $^3P$ multiplet are metastable. The $^3P_1$ state decays mostly by M1 transition to the ground state. Its lifetime is about 1 sec. The $^3P_0$ state decays via E2 transition to the ground state and corresponding lifetime (if no other factors are involved) is about $4 \times 10^3$ sec.

III. POLONIUM AND CERIUM

There are many other examples of the anomalous fine structure in the ground and low excited states of many-electron atoms which involve metastable states and large enhancement and therefore suitable for search of variation of the fine structure constant. The actual choice between them would involve consideration of many other factors which are important for experimentalists but not discussed in present paper. Below we discuss two more examples just to have broader picture.

Brief look at the spectra of elements presented in Moore’s tables reveals that practically all elements with the $np^4$ configuration in the ground state have anomalies in the fine structure (though none of them has so small energy interval as Te I). The most interesting case after Te I is probably polonium (Po I). It has the largest nuclear charge $Z$ ($Z = 84$) among the $np^4$ elements which mean strong relativistic effects and large $q$ and $k$ coefficients (see formulas (1) and (2)). Experimental and theoretical data similar to those considered for Te I are presented in Tables III and IV. We see that enhancement factors $k$ are large and different for different transitions. This is exactly what is needed for the search of variation of the fine structure constant.

Another interesting example is the positive ion of cerium (Ce II). An extract from the tables presenting experimental energies and $g$-factors Lande of lower states of Ce II are presented in Table V together with non-relativistic (NR) values of the $g$-factors. Non-relativistic $g$-factors were calculated according to the formula

$$g = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}.$$  

The data presented in Table V reveals an interesting picture. Fine structure multiplets of Ce II intersect. There are many states with the same total momentum $J$ within an energy interval spanned by singe fine structure multiplet. For example, there are four (!) states of $J = 4.5$ within energy interval of the lowest fine structure multiplet $^4H_6$. It is clear that they must be strongly mixed. Fine structure intervals do not obey Lande’s rule and experimental $g$-factors deviate significantly from the non-relativistic values. All these suggest strong configuration mixing and sensitivity of the intervals to variation of $\alpha$. An interval of particular interest is the $^4H_{11/2} - ^4H_{9/2}$ one. It is small, only 299 cm$^{-1}$ and states involved are strongly mixed with other close states. This ensures strong enhancement of the change of $\alpha$ in the relative change of frequency.

Calculations for Ce II are more difficult than for Te I and Po I because of valence states of high angular momentum ($5d$ and $4f$). Therefore we believe that it is premature to attempt them now. The presence of enhancement is obvious but its actual value would become important only on the stage of planning the measurements. We are ready to perform the calculations if there is any interest from experimentalists.
TABLE V: Energies (cm$^{-1}$) and $g$-factors of lower states of Ce II.

| Config. | Term | $J$  | $E$(exp) | $g$(exp) | $g$(NR) |
|---------|------|------|----------|----------|---------|
| $4f5d^2$ | $^{4}H$ | 3.5  | 0.000     | 0.794    | 0.667   |
|         |      | 4.5  | 2581.257  | 1.023    | 0.970   |
|         |      | 5.5  | 2879.695  | 1.123    | 1.133   |
|         |      | 6.5  | 4203.934  | 1.189    | 1.231   |
| $4f5d^2$ | $^{4}I$ | 4.5  | 1410.304  | 0.856    | 0.727   |
|         |      | 5.5  | 2563.233  | 0.968    | 0.965   |
|         |      | 6.5  | 3793.634  | 1.128    | 1.107   |
|         |      | 7.5  | 5455.845  | 1.196    | 1.200   |
| $4f5d^2$ |      | 3.5  | 1873.934  | 0.806    | 1.000   |
| $4f5d^2$ |      | 0.5  | 2140.492  | 0.985    | 1.000   |

Cases of fine structure anomalies similar to what is presented here for Ce II can be easily found in spectra of many other rare-earth elements. Which of them are suitable for the search of $\alpha$ variation is the question which needs further consideration.

IV. CONCLUSION

We present an alternative way to search for variation of the fine structure constant in laboratory measurements. We propose to use fine structure intervals in the ground or low excited states of many-electron atoms which are strongly perturbed by configuration interaction with neighboring states. This method has double advantages. On one hand, the use of low states ensures that they are metastable. This is important for very accurate frequency measurements. On the other hand, strong perturbation may lead to anomalously small fine structure interval and strong enhancement of the relative sensitivity of the frequencies to the change of the fine structure constant. Because of the high relative sensitivity one does need extremely accurate absolute measurements of the frequencies (this is the difference with conventional atomic clock measurements). Large value of the effect/frequency ratio may also help to reduce importance of some systematic effects (e.g. the Doppler shift and broadening). Note, however, that we do not consider in this paper any practical measurement scheme.

Enhanced and highly non-linear dependence of the small fine structure intervals on the magnitude of the relativistic corrections also presents certain theoretical interest.

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