Electric-dipole-moment enhancement factor for the thallium atom, and a new upper limit on the electric dipole moment of the electron*

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Some time ago, an accurate upper limit on a possible permanent electric dipole moment of the thallium atom in the 6 $^2P_{1/2}$ ground state was obtained by Gould. The result was $D_{\rm Tl} = [(1.3 \pm 2.4) \times 10^{-21}$ cm$]e$. In connection with this value, we have carried out a calculation of the electric dipole enhancement factor $R_{\rm Tl}$, which is defined as the ratio $D_{\rm Tl}/D_e$, where D_e is the corresponding upper limit on a possible electric dipole moment of the (valence) electron. A value $R_{\rm Tl} = 700$ was obtained, which leads to an upper limit $D_e = [(1.9 \pm 3.4) \times 10^{-24}$ cm$]e$. This result is comparable with the value $D_e < (3 \times 10^{-24}$ cm$)e$ previously obtained by Weisskopf *et al.* from measurements on the cesium atom, and with the result of Player and Sandars of $[(0.7 \pm 2.2) \times 10^{-24}$ cm$]e$ obtained from the search for an electric dipole moment in the $^3P_2$ metastable state of xenon. All three results set a stringent upper limit on the amount of a possible violation of T and P invariance in electromagnetic interactions.

I. INTRODUCTION

A few years ago, an accurate upper limit on the electric dipole moment (EDM) of the neutral thallium atom was obtained by Gould.[1] This quantity is of interest, because as has been shown by one of us[2] (P.G.H.S.), the EDM of the thallium atom $D_{\rm Tl}$ reflects a possible electric dipole moment of the electron D_e, and therefore an upper limit on D_e is implicitly determined by the experiment of Gould on $D_{\rm Tl}$. The enhancement factor which relates $D_{\rm Tl}$ to D_e has been introduced (Ref. 2), and is denoted by R. Thus $D_{\rm Tl} = RD_e$. The factor R arises from the electric dipole moment in the thallium core induced by the presumed intrinsic EDM of the valence ($6p$) electron. This enhancement effect is considerable (R is of the order of 100–1000 for heavy atoms), and it is analogous to the antishielding of nuclear quadrupole moments by the electron core, which has been extensively discussed and evaluated elsewhere (ionic antishielding factor γ_∞).[3,4]

Calculations of R for the EDM of the alkali atoms were first carried out in Ref. 2. These results were subsequently verified.[5,6] For the case of the alkali-metal atoms, the values of R of Ref. 2 increase from 0.32 for Na to 24 for Rb and 119 for Cs. For the case of francium ($Z = 87$), an approximate value of $R = 1150$ was also calculated.[2] These calculations of Ref. 2 all pertain to the alkali-metal atoms with a valence ns electron.

The case of Tl is obviously different because the valence electron is in a p state ($6p$), the external part of the configuration being $6s^2 6p$. In fact, it was precisely this circumstance which prompted the investigation of thallium since, as explained in Gould's paper,[1] a violation of invariance under T and P (which is necessary for the existence of an EDM) will be maximized by having a ground state with $L \neq 0$. Thus, according to Sachs,[7] the violation of T and P invariance in the electromagnetic interactions involves a $\vec{J} \cdot \vec{E}$ interaction, where $\vec{E}$ is the electric field and $J = L + \frac{1}{2}\sigma$.

A preliminary estimate of R for Tl, $R_{\rm Tl}$, was made by one of us (P. G. H. S.), namely $R_{\rm Tl} \approx 200$, and this value was used in Gould's paper. However, it was clear to Gould and co-workers that an accurate calculation of $R_{\rm Tl}$ was called for, especially in view of the accuracy of the experimental upper limit for $D_{\rm Tl}$, namely

$$D_{\rm Tl} = [(1.3 \pm 2.4) \times 10^{-21} \text{ cm}]e. \quad (1)$$

It may be noted that an earlier experiment[8] for thallium, which did not include a correction for the motional magnetic field effect, had given a value more than two orders of magnitude larger, namely $(5 \times 10^{-19}$ cm$)e$.

In the present paper, we give the results of an actual calculation of $R_{\rm Tl}$. The result is appreciably larger than the preliminary estimate, namely we find that $R_{\rm Tl} = 700 \pm 100$. Upon using $R_{\rm Tl} = 700$, the upper limit on the EDM of the electron becomes, in view of (1),

$$D_e = [(1.9 \pm 3.4) \times 10^{-24} \text{ cm}]e. \quad (2)$$

This value is comparable to the upper limit on D_e obtained by Weisskopf *et al.*[9] from an accurate experiment on the cesium atom, namely

$$|D_e| \leq (3\times 10^{-24}\ \text{cm})e, \tag{3}$$

and to the upper limit on D_e obtained by Player and Sandars,[10] namely

$$|D_e| = [(0.7 \pm 2.2)\times 10^{-24}\ \text{cm}]e. \tag{3a}$$

Thus with the revised value of R_{Tl} obtained here, the experiment of Gould[1] can be regarded as a major confirmation of the slightly earlier experiment of Weisskopf *et al.*[9] on the cesium atom, and of the experiment of Player and Sandars[10] on the $^3P_2$ metastable state of the xenon atom.

In Sec. II, we describe the calculation of R_{Tl}, starting from the basic unperturbed and perturbed wave functions of the $6p$ electron of thallium. Finally, in Sec. III, we give a brief discussion and summary of the results obtained for the upper limit on the electric dipole moment of the electron D_e.

II. CALCULATION OF THE ELECTRIC DIPOLE ENHANCEMENT FACTOR *R*

The dipole perturbations involving the outermost $6s$ and $6p$ electrons of the Tl atom are $6s\to p$, $6p\to s$, and $6p\to d$. Since the atoms in the experiment are in the $6\,^2P_{1/2}$ ground state, with $J=\frac{1}{2}$, an excitation of the $6p$ electrons to nd states (for which $J=\frac{3}{2}$ or $J=\frac{5}{2}$, but not $J=\frac{1}{2}$) is impossible, by virtue of the conservation of the angular momentum J. In a similar manner, it can also be shown that the $6s\to p$ term makes no net contribution to the dipole enhancement factor R. This leaves $6p\to s$ as the only contribution to R for the thallium atom.

For the unperturbed and the perturbed wave functions, we use the same notation as in Ref. 5. By analogy with the $ns\to p$ excitation of the ns electrons in the alkali-metal atoms, it can be shown that for $6p\to s$ in the thallium atom, we obtain

$$R_{\text{Tl}} = \tfrac{2}{3}\alpha^2 F_r \int_0^\infty u_0'(6p)u_1'(6p\to s)|V_0|\frac{dV_0}{dr}\,dr, \tag{4}$$

where $\alpha = e^2/\hbar c$, F_r is a relativistic correction factor defined below, and the integral over r is completely similar to the integral of Eq. (2) of Ref. 2. The function $u_0'(6p)$ is r times the unperturbed $6p$ radial function, normalized to 1:

$$\int_0^\infty [u_0'(6p)]^2\,dr = 1; \tag{5}$$

$u_0'(6p)$ was taken from the tabulation of Herman and Skillman,[11] who employed the Hartree-Fock-Slater method[12] to obtain the wave functions.

The potential V_0 in Eq. (4) was also obtained from the tables of Herman and Skillman,[11] and the derivative dV_0/dr was obtained by numerical differentiation of V_0 over most of the range. Near the nucleus, where V_0 is rapidly varying ($V_0 \sim -2Z/r$), dV_0/dr was obtained analytically as described in Ref. 5 [see Eqs. (14)–(17)]. In this connection, we note that the quantity R calculated here is the equivalent of S_b of Ref. 5, where the subscript b pertains to the "shielded case" of Ref. 2. Thus the perturbation $u_1'(6p\to s)$ is obtained as the solution of the following differential equation:

$$[-(d^2/dr^2) + V_0 - E_0]\,u_1'(6p\to s) = u_0'(6p)rf(r), \tag{6}$$

where V_0 and E_0 are the unperturbed potential and eigenvalue, respectively, of the Hamiltonian for the $6p$ electron.

The effective values of $V_0(r) - E_0$ are actually obtained from the equation previously introduced by one of us (R.M.S.)[13]:

$$V_0 - E_0 = \frac{1}{u_0'}\frac{d^2u_0'}{dr^2} - \frac{2}{r^2}, \tag{7}$$

where $u_0' = u_0'(6p)$ in the present case. The shielding function $f(r)$ has been previously introduced in Ref. 2:

$$f(r) = \frac{r^3 + [\alpha_c/(Z-1)]}{r^3 + [Z/(Z-1)]\alpha_c}, \tag{8}$$

to take into account the fact that the effect of an external electric field is decreased by a factor $1/Z$ at the nucleus of the atom ($r=0$), due to the shielding effect of the core.[14] For α_c, the polarizability of the Tl^+ core, we used the value $5.2A^3 = 35.1a_H^3$ obtained by Tessman, Kahn, and Shockley.[15] We note that the factor r on the right-hand side of Eq. (6) corresponds to the radial part of the potential $\mathcal{E}_0 r\cos\theta$ due to an external field $\mathcal{E}_0$.

Returning to Eq. (4), the relativistic factor F_r was introduced in Ref. 2 to describe the effect of the relativistic corrections to the otherwise nonrelativistic approximation of Eq. (4). The factor F_r, which was first tabulated by Kopfermann,[16] is defined as

$$F_r = 3/[\rho(4\rho^2 - 1)], \tag{9}$$

where $\rho = (1-\alpha^2Z^2)^{1/2} = 0.8066$ for Tl ($Z=81$), giving $F_r = 2.321$.

In the solution of Eq. (6) for $u_1'(6p\to s)$, we employed the computer program described in Ref. 17. Near the nucleus ($r\sim 0$), the inhomogeneous term becomes negligible, and $u_1'(6p\to s)$ becomes approximately proportional to the unperturbed $6s$ wave function, $u_0'(6s)$. In fact, we find that in this region, and actually up to $r\sim 1a_H$, we have $u_1'(6p\to s) \approx 11.5u_0'(6s)$, where $u_0'(6s)$ is normalized to 1, in the same manner as $u_0'(6p)$ [see Eq. (5)]. At large r, namely $r = 3.7a_H$, $u_1'(6p\to s)$ has an additional node, so that it has the same number of nodes as a $7s$ wave function.

The functions $u_1'(6p\to s)$ and $u_0'(6p)$ for the Tl atom are shown in Fig. 1. Concerning $f(r)$, we note that it has the same general shape as the functions $f(r)$ shown in Fig. 1 of Ref. 5, except that it lies somewhat below the function $f(r)$ for Cs, and attains the value $\frac{1}{2}$ at $r_{1/2}\cong \alpha_c^{1/3}=35.1^{1/3}=3.27a_H$.

The major contribution to the integral for $R_{\rm Tl}$ [Eq. (4)] arises from the region very near the nucleus. As a matter of fact, the integral from 0 to r in Eq. (4), to be denoted by $I(r)$, already attains its maximum value 9.314×10^6 at $r=0.025a_H$, at the location of the first node of $u_1'(6p\to s)$, and decreases for larger r to its asymptotic value $I(\infty)=8.686\times10^6$ [which is the integral in Eq. (4)]. The reason for the importance of the region near $r=0$ is that the integrand of Eq. (4), to be denoted by $P(r)$, approaches a constant value at $r=0$. This can be easily seen from the radial dependences of the four factors of $P(r)$, namely

$$u_0'(6p)\propto r^2,\ u_1'(6p\to s)\propto r,\ V_0\to -2Z/r,$$
$$dV_0/dr\to +2Z/r^2,\quad \text{for } r\to 0.$$

Thus

$$P(r)\propto r^2\, r\, r^{-1} r^{-2}=\text{const.}$$

The actual value of P at $r=0$, $P(0)$, has to be obtained by extrapolation from the results for $r=0.0025$, 0.005, and $0.0075a_H$, which are the points in the radial integration of Eq. (4). The values of $P(r)=u_0u_1|V_0|(dV_0/dr)$, in units 10^9, are as follows: $P(r=0)=1.304$; $P(0.0025)=1.0065$; $P(0.0050)=0.7393$; $P(0.0075)=0.5153$; $P(0.0100)=0.3427$; $P(0.0125)=0.2245$; and $P(0.0150)=0.1416$. The extrapolation to $r=0$ is quite smooth, as can be seen by comparing first and second differences.

The result for R is given by

$$R=\tfrac{2}{3}\alpha^2F_rI(\infty)=\tfrac{2}{3}\alpha^2(2.321)(8.686\times10^6)$$
$$=308.4F_r=716.0. \qquad (10)$$

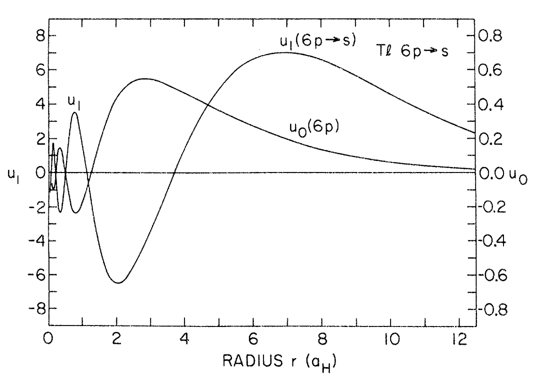

FIG. 1. Perturbed wave function $u_1(6p\to s)$ and the unperturbed $6p$ function $u_0(6p)$ for the thallium atom.

We note that even the smallest r values which contribute appreciably to R, namely $r\cong 0.001a_H$ correspond to a radius of $\sim5.3\times10^{-12}$ cm, which lies well outside the radius of the Tl nucleus, r_N, which is approximately given by $r_N=1.2A^{1/3}$ F $=1.2\times204^{1/3}\times10^{-13}=7.07\times10^{-13}$ cm. Hence the effects of nuclear structure on R are expected to be small, i.e., we can use the full nuclear potential $-2Z/r$ Ryd at the radii r for which the calculations are carried out.

We also note that an independent calculation carried out using also the Herman-Skillman potential, but a slightly different treatment near $r=0$, gave $I(\infty)=9.28\times10^6$, from which one obtains

$$R=\tfrac{2}{3}\alpha^2F_rI(\infty)=329F_r=764.0, \qquad (11)$$

which differs by only ~7% from the result of Eq. (10).

Finally, we wish to point out that an additional independent relativistic calculation, using equations derived by one of us (P.G.H.S.),[18] gives $R=640$.

Thus we may conclude from these three results that the actual value of R for thallium is ≈700, with an estimated maximum uncertainty of ±100. Such a result is also reasonable in terms of the result for francium ($Z=87$) previously obtained,[2] namely $R=1150$ for the unshielded case, which corresponds to $R\cong 800$ for the shielded case considered in the present paper.

In the following, we will use the result $R_{\rm Tl}=700$, and apply this enhancement factor to the results of Gould in Ref. 1. As mentioned in the Introduction, this procedure gives $D_e=[(1.9\pm3.4)\times10^{-24}\ \text{cm}]e$ for the upper limit on the electric dipole moment of the electron [see Eq. (2)]. This result represents a considerable improvement over the upper limit deduced by Gould on the basis of the preliminary value, $R=200$.

III. SUMMARY

In the Introduction, we have compared the present result for D_e, namely $[(1.9\pm3.4)\times10^{-24}\ \text{cm}]e$ with the upper limit given by Weisskopf *et al.* in Ref. 9, namely $[3\times10^{-24}\ \text{cm}]e$, as obtained from measurements on cesium, and with the value obtained by Player and Sandars in Ref. 10, namely $[(0.7\pm2.2)\times10^{-24}\ \text{cm}]e$. We have thus concluded that the present limit provides a major confirmation of the independent measurements for cesium and xenon. Actually, if one considers the detailed result for cesium,[9] that is $|D_{\rm Cs}|=[(0.8\pm1.8)\times10^{-22}\ \text{cm}]e$, and divides by the electric dipole enhancement factor[2] $R_{\rm Cs}=120$, one obtains

$$D_e = [(0.7 \pm 1.5)\times 10^{-24}\ \text{cm}]e, \tag{12}$$

which is a factor of ~2 smaller than the present result obtained from Gould's measurements for thallium. However, the three results for D_e (from Tl, Xe, and Cs) lie clearly in the same range of values, and they set a stringent upper limit on the amount of a possible violation of T and P invariance in the electromagnetic interactions.

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