the polarizing power of the mirror is not constant over the spectrum. The asymmetry was not observed for the Fe^{54}-enriched iron mirrors either, for which the real part of the coherent-scattering amplitude was close to zero, as indicated by the high polarizing efficiency ($P \approx 1$) of these mirrors due to compensation of nuclear and magnetic scattering amplitudes.

Since it was not our aim to investigate the spin-orbit effect but merely to obtain a quantitative experimental estimate for these factors in the case of reflecting materials used in practice, we did not look for the ideal conditions under which these effects might be seen. The theoretical estimates reported by Handel[2-4] do, in fact, refer to such ideal conditions, i.e., complete compensation of the real parts of the nuclear and magnetic scattering amplitudes, sufficiently large imaginary part of the amplitude, and particular purity on the reflecting surface layer, which is quite difficult to achieve in an experiment.

On the basis of our measurements, we can find no evidence for the polarization asymmetry reported by Berndorfer[1] and consider that this effect is more likely to have been due to technological factors and not the spin-orbit contribution.

It also follows from our experiments that Handel's proposals[4] regarding the use of spin-orbit effects for measuring the potential due to the electric surface dipole layer, and for producing on this basis an electric neutron polarizer, are far from experimental realization.

In conclusion, we are indebted to G. M. Drabkin and E. F. Shender for useful discussions of the experiment and the possible spin-orbit effects, and to N. V. Borovikova for preparing the samples for the experiment.

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A bound on the proton electric dipole moment derived from atomic experiments

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The upper bound $|d/e| < 5.5 \times 10^{-19}$ cm on the proton electric dipole moment (EDM) d is derived from the experimental upper bound for the EDM of the cesium atom in a state with $F = 4$. Similar measurements on an $F = 3$ state might improve this result by a factor of 1.5. An upper bound on the magnetic quadrupole moment of the nucleus (which might be induced, for example, by the EDM of the valence nucleon) is also derived from the same experiment. The search for the proton EDM in experiments with polar molecules is discussed.

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1. INTRODUCTION

It is well known[1] that elementary particles can have electric dipole moments (EDM) only if time-reversal invariance is violated. Up to now, T-odd interactions seem to have been observed only in the decay of K^0 mesons. From this it is clear why it is of interest to seek EDM of elementary particles. In particular, elementary-particle EDM would be of interest because of the light they might shed on the structure of the T-odd interaction.

Experiment gives the following upper bound for the neutron EDM, d_n: $|d_n/e| < 10^{-23}$ cm.[2] The difficulties in measuring the EDM of charged particles—electrons and protons—are obvious. Nevertheless, the idea of seeking the electron EDM via the EDM it induces in a neutral atom has proved to be very fruitful. At first glance the situation here would seem to be hopeless in view of Schiff's well-known theorem,[3] according to which the EDM of a system of nonrelativistic particles in equilibrium under the action of electrostatic forces will vanish provided the intrinsic EDM of each particle has the same spatial distribution as the charge of that same particle. As Sandars showed,[4] however, owing to relativistic effects the induced EDM of a heavy atom not only is not small, but on the contrary, is much enhanced as compared with the EDM of the electron inducing it. Calculations[4-7] show that the enhancement factor is ~130 for cesium and ~500 for thallium. Experiments with atomic cesium[8] and thallium[9] resulted in the following bounds for the electron EDM: $|d_e/e| < 3 \times 10^{-24}$ cm and $|d_e/e| < 7 \times 10^{-24}$ cm.

In the present study we derive an upper bound for the

proton EDM from an experiment with atomic cesium. This bound is of considerable interest despite the fact that it is much higher (weaker) than the bound said to have been derived from an experiment with TlF molecules.[10,11] The point is that the interpretation[10] of the TlF experiment is not entirely convincing. The bound on the proton EDM that actually follows from the TlF experiment may be, say, an order of magnitude higher. Worse than that, at present there is no practicable way in sight to extract an unambiguous bound for the proton EDM from the experiment in question. This situation will be discussed in more detail at the end of the present paper.

2. NUCLEAR MAGNETIC QUADRUPOLE MOMENT INDUCED BY THE EDM OF A VALENCE NUCLEON

We begin with a general treatment of the effects due to the presence in a nucleus of a valence nucleon having an EDM. We write the relativistic Hamiltonian for the interaction of the dipole moment d of a spin-$\frac{1}{2}$ particle with an external field $F_{\mu\nu}$ in the form

$$\hat{H}_d=\tfrac{1}{2}d\gamma_0\gamma_5\sigma_{\mu\nu}F_{\mu\nu},\quad \gamma_5=-i\gamma_0\gamma_1\gamma_2\gamma_3,\quad \sigma_{\mu\nu}=\tfrac{1}{2}(\gamma_\mu\gamma_\nu-\gamma_\nu\gamma_\mu). \tag{2.1}$$

Then we obtain the following expressions for the charge and current densities:

$$\rho_d=-d\nabla_n(\Psi^+\gamma_5\gamma_n\Psi), \tag{2.2}$$

$$\mathbf{j}_d=icd\ \mathrm{rot}\ (\Psi^+\boldsymbol{\gamma}\Psi). \tag{2.3}$$

Now we introduce a normalized two-component Schrödinger wave function ψ into Eqs. (2.2) and (2.3) in place of the Dirac bispinor Ψ, retaining only terms up to the second order in v/c. The result is

$$\rho_d=-d\nabla_n\left\{\psi^+\left[\sigma_n-\frac{1}{8m_p^2c^2}(\sigma_n(p'^2+p^2)+2(\boldsymbol{\sigma}\mathbf{p}')\sigma_n(\boldsymbol{\sigma}\mathbf{p}))\right]\psi\right\}, \tag{2.2a}$$

$$\mathbf{j}_d=\frac{d}{2m_p}\mathrm{rot}(\psi^+[\sigma\times(\mathbf{p}'+\mathbf{p})]\psi). \tag{2.3a}$$

Here $\mathbf{p}'$ and $\mathbf{p}$ are the momentum operators acting on ψ^* and ψ, respectively.

We begin with the first term in (2.2a), which corresponds to the ordinary contribution of the valence nucleon to the EDM of the nucleus. The relativistic corrections to the motion of the nucleus as a whole are negligible. But then, in view of Schiff's theorem,[3] the EDM of the nucleus, by itself, affects the EDM of the atom only via the interaction of the nucleus with the magnetic field produced by the electrons, and also via possible effects associated with the finite size of the nucleus. The EDM induced in the atom by this magnetic interaction turns out to be very small, of the order of $dmZ\alpha^2/m_p$ (m is the electron mass), as can be shown without difficulty. Nevertheless, we postpone discussion of the effects due to the finite nuclear size to Sec. 4.

The relativistic corrections to ρ_d turn out to be of two kinds. Some arise from the motion of the proton in the nucleus and reduce simply to a renormalization of the resultant EDM of the nucleus. Although these effects are not especially small, they are of no particular interest in view of Schiff's theorem. The other relativistic corrections have a structure of the form

$$\frac{\hbar^2}{m_p^2c^2}\nabla_k\nabla_m\nabla_n(\psi^+\sigma_k l_m l_n\psi)$$

(l is the orbital angular momentum of the nucleon) and correspond, in particular, to an octupole moment of the nucleus induced by the EDM of the valence nucleon. The contribution of these corrections to the EDM of the atom is also small, being of the order of $d(m/m_p)^2Z^2\alpha^2$.

Now let us consider the effects due to the space current j_d. We shall make use of the nuclear shell model to simplify expression (2.3). Judging from the Schmidt plot, the errors introduced by this approximation may amount to 30–40% for the Cs^{133} nucleus of interest to us. We begin by averaging the operator $(\mathbf{p}'+\mathbf{p})/2m_p$ over a state of the valence nucleon with a given orbital angular momentum l. Actually, we are talking about the current that produces the orbital magnetic moment of the nucleus. Assuming a point nucleus, we express the corresponding magnetic field in the form

$$\mathbf{H}=\frac{|e|\hbar}{2m_pc}\left[\nabla\times\left[\nabla\frac{1}{r}\times\mathbf{l}\right]\right]. \tag{2.4}$$

After performing some calculations involving the Maxwell equation $\mathrm{curl}\mathbf{H}=4\pi\mathbf{j}/c$, we obtain

$$\frac{1}{2m_p}\overline{(\mathbf{p}'+\mathbf{p})}=\frac{\hbar}{2m_p}[\nabla\delta(\mathbf{r})\times\mathbf{l}]\ . \tag{2.5}$$

from which follows

$$\mathbf{j}_d=-d\frac{\hbar}{2m_pc}[\nabla\times\overline{\mathbf{l}(\boldsymbol{\sigma}\nabla)}\delta(\mathbf{r})]. \tag{2.6}$$

Here the bar indicates averaging over a state of the nucleus with a given total angular momentum i. As is easy to show, the tensor $\overline{l_m\sigma_n}$ is symmetric; moreover, the term proportional to δ_{mn} in this tensor obviously does not contribute to j_d. Taking these remarks into account, we obtain

$$j_{dm}=d\frac{\hbar}{2m_p}\varepsilon_{mrn}\frac{g}{2}\left[i_ri_s+i_si_r-\frac{2}{3}\delta_{rs}i(i+1)\right]\nabla_s\nabla_n\delta(\mathbf{r}), \tag{2.7}$$

$$g=\frac{1}{4}+\frac{l(l+1)+3/4}{2i(i+1)}-\frac{3}{4}\frac{[l(l+1)-3/4]^2}{i^2(i+1)^2}.$$

For the Cs^{133} nucleus, in which the valence proton is in a $g_{7/2}$ state, we find $g=-40/189$. The vector potential produced by the current $\mathbf{j}_d$ is

$$A_m=d\frac{\hbar}{2m_pc}\varepsilon_{mrn}\frac{g}{2}\left[i_ri_s+i_si_r-\frac{2}{3}\delta_{rs}i(i+1)\right]\nabla_s\nabla_n\frac{1}{r}. \tag{2.8}$$

The physical interpretation of the effect under discussion is clear. Just as the orbital motion of a charged particle leads to a magnetic dipole moment of the system, the orbital motion of a charge having an EDM leads to a magnetic quadrupole moment of the sys-

tem. In the present case it is natural to take the tensor

$$M_{mn}=\frac{3}{2}d\frac{\hbar}{m_pc}g\left[i_mi_n+i_ni_m-\frac{2}{3}\delta_{mn}i(i+1)\right] \quad (2.9)$$

as the nuclear magnetic quadrupole moment operator. We shall characterize this operator as usual by the quantity

$$M=M_{zz}|_{i_z=i}=d\frac{\hbar}{m_pc}gi(2i-1). \quad (2.10)$$

For Cs^{133} this expression reduces to

$$M_{Cs}=-\frac{80}{9}\frac{d}{|e|}\mu_p, \quad (2.11)$$

in which $\mu_p=|e|\hbar/2m_pc$ is the nuclear magneton and e is the electron charge. The nuclear magnetic quadrupole moment can obviously differ from zero only if $i\geq 1$.

We note that Eqs. (2.5)–(2.10) are equally valid for a nucleus that contains a valence neutron having an EDM; thus, such a nucleus also acquires a magnetic quadrupole moment if $i\geq 1$.

3. THE EDM INDUCED IN AN ATOM BY THE MAGNETIC QUADRUPOLE MOMENT OF ITS NUCLEUS

As will be evident from what follows, the relativistic effects in the atom are essential to the phenomenon under discussion. Hence we write down at once the Hamiltonian for the interaction of a relativistic electron with the vector potential A of Eq. (2.8):

$$H_1=d\frac{e\hbar}{2m_pc}\alpha_k\varepsilon_{klm}\nabla_l\nabla_n\frac{1}{r}\frac{g}{2}\left[i_mi_n+i_ni_m-\frac{2}{3}\delta_{mn}i(i+1)\right], \quad (3.1)$$

in which $\boldsymbol{\alpha}=\gamma_0\boldsymbol{\gamma}$ represents the Dirac matrices for the electron.

The cesium atom has a $6s_{1/2}$ ground state, and the interaction (3.1) can cause mixing with it of states of opposite parity in which the atom has the same total angular momentum F. As a result, the atom acquires an EDM. Since the dipole matrix elements $6s-6p$ are very much larger in cesium than all the others, we shall consider only admixtures of $6p$ states to the $6s$ ground state. The electronic part of the interaction (3.1) is obviously an irreducible second-rank tensor and therefore cannot mix $s_{1/2}$ and $p_{1/2}$ states, even in the presence of relativistic effects. It therefore remains only to calculate the admixture of the $6p_{3/2}$ state.

The relativistic wave functions of the electron can be written in the form

$$\Psi_{s_{1/2}}=\begin{pmatrix} g_{1/2}(r)\Omega_{1/2\,0} \\ -if_{1/2}(r)\,(\sigma r/r)\Omega_{1/2\,0}\end{pmatrix},\quad \Psi_{p}=\begin{pmatrix} g_{1/2}(r)\Omega_{1/2\,1} \\ -if_{1/2}(r)\,(\sigma r/r)\Omega_{1/2\,1}\end{pmatrix}. \quad (3.2)$$

Here the Ω_{jl} are spherical spinors. Let us consider the radial functions g and f. Because of the singularity of interaction (3.1) (it is proportional to $1/r^3$) the main contribution to the matrix element of H_1 comes from the region of small r, where the nucleus can be regarded as unscreened and the energy of the electron can be neglected as compared with the potential. It can be shown (see, e.g., [12]) that in this region we have

$$rg(r)=-C\left(\frac{x}{2}\frac{d}{dx}-\varkappa\right)J_{2\gamma}(x),\quad rf(r)=CZ\alpha J_{2\gamma}(x); \quad (3.3)$$

$$x=(8Zr/a)^{1/2},\quad \varkappa=(-1)^{j+1/2-l}(j+1/2),\quad \gamma=[(j+1/2)^2-Z^2\alpha^2]^{1/2},$$
$$C=(-1)^{j+1/2-l}(2\mathrm{Ry}/e^2\nu^3)^{1/2}. \quad (3.4)$$

Here $\mathrm{Ry}=me^4/2\hbar^2$ is the Rydberg constant, $a=\hbar^2/me^2$ is the Bohr radius, and ν is the effective principal quantum number.

A straightforward but cumbersome calculation of the matrix element that mixes the $s_{1/2}$ and $p_{3/2}$ states in cesium leads to the following result:

$$\langle p_{1/2}|H_1|s_{1/2}\rangle=\frac{d}{ea}\frac{m}{m_p}Z^2\alpha^2\,\mathrm{Ry}\,(\nu_p\nu_s)^{-1/2}R\frac{16}{63}\begin{cases}\sqrt{35}\\ -5\sqrt{3}\end{cases}. \quad (3.5)$$

Here and below the upper number of the pair refers to a state in which the atom has the total angular momentum $F=4$, and the lower one, to a state with $F=3$. In this case the relativistic enhancement factor is

$$R=\frac{6!\Gamma(\gamma_{1/2}+\gamma_{1/2}-2)}{\Gamma(\gamma_{1/2}+\gamma_{1/2}+3)\Gamma(\gamma_{1/2}-\gamma_{1/2}+3)\Gamma(\gamma_{1/2}-\gamma_{1/2}+3)}\approx 1.3. \quad (3.6)$$

The dipole matrix element of interest to us is

$$\langle i=7/2,\ j=1/2,\ F;\ F_z=F|ez|i=7/2,\ j=3/2,\ F;\ F_z=F\rangle=ea\rho\begin{cases}1/3\sqrt{7}/5\\ 1/4\sqrt{3}\end{cases}. \quad (3.7)$$

Using (3.5) and (3.7), we obtain the following expression for the contribution D_1 from the effect under discussion to the EDM of the cesium atom:

$$D_1=d\frac{m}{m_p}Z^2\alpha^2\rho\frac{\mathrm{Ry}}{E_{6p}-E_{6s}}(\nu_{6s}\nu_{6p})^{-1/2}R\begin{cases}-32/27\\ 40/21\end{cases}. \quad (3.8)$$

Using the known values of the effective quantum numbers, $\nu_{6s}=1.87$ and $\nu_{6p}=2.35$, and the dimensionless radial matrix element,[1)] $\rho(6s,6p)\approx -5.8$, we obtain the following numerical result:

$$D_1=d\cdot 10^{-3}\begin{cases}0.78\\ -1.25\end{cases}. \quad (3.9)$$

By comparing (3.9) with the experimental upper bound $|D_{Cs}/e|<3.7\times10^{-22}$ cm for the EDM D_{Cs} of the cesium atom in the $F=4$ state,[8] we obtain the following upper bound for the proton EDM:

$$|d/e|<4.7\cdot10^{-19}\ \text{cm}. \quad (3.10)$$

We note that one could improve the bound (3.10) by a factor of about 1.5 by measuring the EDM of the cesium atom in the $F=3$ state.

Finally, we can derive an upper bound to the magnetic quadrupole moment of the Cs^{133} nucleus from the experiment with atomic cesium. Comparing (2.11) with (3.10), we obtain

$$|M_{Cs^{133}}|<6.8\cdot10^{-6}\mu_p r_{Cs}, \quad (3.11)$$

in which $r_{Cs} = 6.1 \times 10^{-13}$ cm is the radius of the Cs^{133} nucleus. The upper bound given by Sandars in[15] is 15 times higher. Since no details are given in[15], it is impossible to determine the reason for this large discrepancy.

4. THE EDM INDUCED IN AN ATOM BY THE DIPOLE MOMENT OF ITS FINITE NUCLEUS

Now let us consider another mechanism by which the EDM of a valence nucleon can induce an EDM in an atom. As Schiff pointed out,[3] a system of particles can have an EDM even in the absence of relativistic effects provided the charge and EDM distributions differ from one another in at least one of the particles. The interaction between an electron and the dipole moment of a finite nucleus, which gives rise to an EDM in the atom, can be written in the form

$$H_2 = \int d\mathbf{r}' \, [\rho_d(\mathbf{r}') - \rho_q(\mathbf{r}')] \, \mathbf{d} \nabla_{r'} \frac{e}{|\mathbf{r}-\mathbf{r}'|}. \tag{4.1}$$

Here $\mathbf{r}$ is the coordinate of the electron reckoned from the center of the nucleus, and ρ_q and ρ_d are the nuclear charge and dipole-moment densities, respectively, each normalized to unity.

Retaining only the lowest-order nonvanishing term in the expansion of (4.1) in powers of r'/r, we obtain

$$H_2 = \frac{e}{2} \int d\mathbf{r}' \, [\rho_q(\mathbf{r}') - \rho_d(\mathbf{r}')] \, d_l r_m' r_n' \nabla_l \nabla_m \nabla_n \frac{1}{r}. \tag{4.2}$$

The charge density ρ_q is obviously spherically symmetric, except possibly for corrections of the order of Z^{-1}. Even for deformed nuclei, where these corrections may be an order of magnitude larger, they can still be neglected. As for ρ_d, it is natural to assume that it coincides with the density distribution of the valence nucleon. Finally, we shall assume that the dipole moment of the nucleus is equal to the intrinsic EDM of the valence nucleon: $\mathbf{d} = d\boldsymbol{\sigma}$.

In view of all this we can rewrite Eq. (4.2) in the form

$$H_2 = \frac{ed}{2} \int d\mathbf{r}' \, r'^2 \left\{ \rho_q(r') \frac{1}{3} \delta_{mn} \langle \sigma_l \rangle - \rho_d(\mathbf{r}') \langle \sigma_l n_m n_n \rangle \right\} \nabla_l \nabla_m \nabla_n r^{-1} \tag{4.3}$$

Here the angle brackets indicate averaging over a nuclear state with the given angular momentum i.

Now let us consider the cesium atom specifically. Since we are interested primarily in the mixing of $6s$ and $6p$ states, it is sufficient to retain only the vector part $-(\frac{1}{5})(\nabla_l \delta_{mn} + \nabla_m \delta_{ln} + \nabla_n \delta_{lm}) 4\pi\delta(\mathbf{r})$ of the "electronic factor" $\nabla_l \nabla_m \nabla_n r^{-1}$. Then standard manipulations lead to the following expression for the case of Cs^{133} ($i = \frac{7}{2}$, $l = 4$):

$$H_2 = \frac{1}{189} ed(7r_q^2 - 3r_d^2) \, i\nabla \, 4\pi\delta(\mathbf{r}). \tag{4.4}$$

What can be said about the mean square radii

$$r_{q,d}^2 = \int d\mathbf{r}' \, r'^2 \rho_{q,d}(\mathbf{r}')?$$

TABLE I.

	$r_q^2 \cdot 10^{24}$, cm^2	$r_m^2 \cdot 10^{24}$, cm^2	$(r_q^2 - r_m^2)/r_0^2 = \delta r^2/r_0^2$
H_1^3 [16]	2.82±0.34	2.66±0.33	0.03±0.10
He_2^3 [16]	3.88±0.40	2.86±0.34	0.16±0.08
Sc_{21}^{45}	12.39±0.63 [17]	12.67±1.57 [18]	−0.01±0.06
V_{23}^{51}	13.10±0.65 [17]	12.89±0.72 [18]	0.01±0.05
Co_{27}^{59}	14.21±0.38 [17]	13.99±1.05 [18]	0.01±0.05

As is known (e.g., from the scattering of electrons from nuclei), the charge of the nucleus can be regarded with good accuracy as uniformly distributed throughout a sphere of radius $r_0 = 1.2 \times 10^{-13} A^{1/3}$ cm (A is the mass number of the nucleus). Then $r_q^2 = (\frac{3}{5}) r_0^2$. As regards r_d^2, it is natural to assume that it is equal to the mean square magnetic radius r_m^2 of the nucleus. The last quantity has been measured for tritium, He^3, Sc^{45}, V^{51}, and Co^{59}. Since there is no significant difference between r_q^2 and r_m^2 for any of these nuclei except He^3 (see Table I), we assume that $r_d^2 = r_q^2$ for cesium, too. As a result, we obtain the following interaction Hamiltonian for cesium:

$$H_2 = \frac{4}{315} e d r_0^2 \, i\nabla \, 4\pi\delta(\mathbf{r}). \tag{4.5}$$

It should be pointed out that for $l \geq 1$, the effect under consideration is essentially due to the orbital quadrupole moment of the nucleus. Unlike the effect discussed in the preceding sections, it is directly dependent on the nuclear radius r_0.

Hamiltonian (4.5) mixes both the $6p_{1/2}$ and the $6p_{3/2}$ states with the $6s_{1/2}$ ground state. Admixtures of higher p states can be neglected as before because the corresponding dipole matrix elements are small. A calculation differing little from the one discussed in Sec. 3 gives the following expression for the EDM induced in a cesium atom by the EDM of a valence proton on account of the finite size of the nucleus:

$$D_2 = d \frac{Z^2 r_0^2}{a^2} \rho \frac{\mathrm{Ry}}{E_{6p} - E_{6s}} (\nu_{6s} \nu_{6p})^{-1} (R_{1/2} + 2R_{3/2}) \begin{Bmatrix} 2/105 \\ 1/105 \end{Bmatrix}; \tag{4.6}$$

$$R_{1/2} = \frac{12\gamma_{1/2}}{(2\gamma_{1/2}+1)\Gamma^2(2\gamma_{1/2}+1)} \left(\frac{2Zr_0}{a} \right)^{2\gamma_{1/2}-2} \approx 2.7, \tag{4.7}$$

$$R_{3/2} = \frac{6[(\gamma_{3/2}+1)(\gamma_{3/2}+2) + Z^2\alpha^2]}{\Gamma(2\gamma_{3/2}+1)\Gamma(2\gamma_{3/2}+1)} \left(\frac{2Zr_0}{a} \right)^{2\gamma_{3/2}-3} \approx 2.2. \tag{4.8}$$

Here the R are the relativistic enhancement factors for the mixing of the $p_{1/2}$ and $p_{3/2}$ states to the ground state. To simplify the writing of formula (4.6) we have neglected the small differences in energy and dipole matrix elements between the $6p_{1/2}$ and $6p_{3/2}$ states.

On comparing Eqs. (3.8) and (4.6), we find that the EDM of the cesium atom due to the finite size of the nucleus is rather small as compared with that due to the nuclear magnetic quadrupole moment:

$$\frac{D_2}{D_1} = \frac{m_p}{m} \left(\frac{r_0}{\alpha a} \right)^2 \frac{R_{1/2} + 2R_{3/2}}{R} \begin{Bmatrix} -1/15 \\ 1/25 \end{Bmatrix} \approx \begin{Bmatrix} -0.17 \\ 0.10 \end{Bmatrix}. \tag{4.9}$$

Thus, this effect does not considerably change the bound (3.10) found in the preceding section.

Now let us discuss what limitations on the proton

EDM can be derived from an experiment with atomic thallium.[9] Both stable thallium isotopes, Tl^{203} and Tl^{205}, have spin $i=\frac{1}{2}$, so neither can have a magnetic quadrupole moment. According to the shell model, each of these isotopes has an $s_{1/2}$ valence proton. Thus, interaction (4.3) reduces for thallium to the form

$$H_2=-\tfrac{1}{3}ed\delta r^2\mathbf{i}\nabla 4\pi\delta(\mathbf{r}),\quad \delta r^2=r_q^2-r_d^2. \tag{4.10}$$

The ground state of the thallium atom belongs to the $6s^26p_{1/2}$ configuration. Here the dipole matrix elements for the transitions $6p-7s$ and $6s-6p$ are large, so it will be accurate enough to consider only admixtures of the $6s^27s$ state and states belonging to the $6s6p^2$ configuration to the $6s^26p_{1/2}$ ground state. If the total angular momentum F of the atom is zero, the EDM of the atom will obviously also be zero. We shall therefore consider states with $F=1$.

A calculation similar to the one described above gives the following expression for the contribution to the EDM of thallium from a $6s^27s$ admixture:

$$D^{(1)}=-\frac{8}{27}d\frac{Z^2\delta r^2}{a^2}\rho(6p_{1/2},7s)\frac{\mathrm{Ry}}{E_{7s}-E_{6p_{1/2}}}(\nu_{7s}\nu_{6p})^{-1/2}R_{1/2}. \tag{4.11}$$

In calculating the contribution from states belonging to the $6s6p^2$ configuration it is convenient to use the method of second quantization, the application of which to a related problem is discussed in[19]. This contribution is found to be

$$D^{(2)}=-\frac{8}{27}d\frac{Z^2\delta r^2}{a^2}\rho(6s,6p)\frac{\mathrm{Ry}}{\bar{E}-E_{6p_{1/2}}}(\nu_{6p}\nu_{6s})^{-1/2}(R_{1/2}+4R_{3/2}), \tag{4.12}$$

in which $\bar{E}$ is the average energy of the $6s6p^2$ band. The relativistic enhancement factors for thallium are $R_{1/2}\approx 7.9$ and $R_{3/2}\approx 4.9$ (see (4.7) and (4.8)). The values $\rho(6p_{1/2},7s)\approx 2.2$, $\rho(6s,6p)\approx -1.7$, $\bar{E}-E_{6p_{1/2}}=71\,300$ cm^{-1}, $\nu_{6p}=1.58$, $\nu_{7s}=2.19$, and $\nu_{6s}=0.99$ for the other parameters can be derived from an analysis (presented in[19]) of the experimental data on the thallium spectra and oscillator strengths.

In fact, it follows from a comparison of the calculated and experimental values of the thallium hyperfine splitting constants, and also from numerical calculations of the wave functions made by O. P. Sushkov and V. V. Flambaum, that the normalizing factor (3.4) for thallium, and hence also expressions (4.11) and (4.12), are actually larger—the latter by about 30%. Taking this into account, we find

$$D_{\mathrm{Tl}}\approx d\cdot 1.3\cdot 10^{-3}\delta r^2/r_0^2. \tag{4.13}$$

It is very difficult to estimate δr^2 with any reliability. In calculating the bound on the proton EDM from the results of an experiment with TlF molecules, Sandars approximated the potential for the valence nucleon in the thallium nucleus by an infinitely deep square well of radius r_0 and obtained the following result: $r_q^2-r_d^2=\delta r^2=(4/15)r_0^2=0.27r_0^2$.[10] A similar calculation for Sc^{45}, V^{51}, and Co^{59}, all of which have a valence proton in the $f_{7/2}$ state, gives $\delta r^2=0.13r_0^2$, which is clearly in contradiction with experiment (see the table). It is almost obvious that these calculations underestimate r_m^2 (or r_d^2) and correspondingly overestimate δr^2. Actually, according to a communication from V. B. Telitsyn, calculations using the more realistic Saxon–Woods potential lead to $\delta r^2=0.12r_0^2$ for thallium and to $\delta r^2=-0.03r_0^2$ for the excited $f_{7/2}$ state for the proton in Y^{89}, which is at least in qualitative agreement with the experimental data given in the table. However, even this calculation is not accurate enough for our purposes. It is enough to make an error of 15–20% in calculating r_m^2 (and such an error seems quite possible in these calculations) to reduce the value of δr^2 obtained for thallium, for example, by several more times. Neither is an experimental determination of r_m^2 for the thallium nucleus practicable at present. We recall that the contribution from the magnetic moment to the scattering of an electron by a nucleus is Z^2 times smaller than the contribution from the nuclear charge, and $Z=81$ for thallium. An experimental determination of the magnetic radius of F^{19}, in which, as in thallium, the valence proton is in an s state, would be very useful in this connection.

Thus, in view of the appreciably higher accuracy of the cesium experiments, it is evident from formulas (3.9), (4.9), and (4.13) that atomic cesium is a much more suitable material for investigating the proton EDM than atomic thallium, even if Sandars' value $\delta r^2/r_0^2=0.27$ for the latter is correct.

5. CONCLUSION

Thus, the following bound on the proton EDM follows from the experiment with atomic cesium with allowance for the correction (4.9):

$$|d/e|<5.5\cdot 10^{-19}\ \mathrm{cm}. \tag{5.1}$$

The greatest uncertainty in this result is that associated with the error involved in using the shell model in calculating the nuclear magnetic quadrupole moment; this error may be as large as 30–40%. The errors in the atomic calculations, judging from similar calculations of the cesium hyperfine splitting, can hardly exceed 15–20%.

The upper bound to the proton EDM that is said to follow from the results of an experiment with molecular TlF[11] is 35 times lower than (5.1). However, this bound is essentially based on the estimate $\delta r^2/r_0^2=0.27$ which, as was pointed out in the preceding section, is probably much too high, and at present there are no practicable means for improving it in sight. Further, an additional error (which is apparently not easily controllable) is involved in the calculation of the molecular wave functions. At any rate, a simple estimate of the effective electric field acting on the EDM of the valence proton in the thallium nucleus in a polarized TlF molecule leads to a value that is several times smaller than that given by Sandars *et al.*,[10,11] even if the value $\delta r^2/r_0^2=0.27$ is used. Thus, the upper bound to the proton EDM given in[11] seems to be much lower than is justified by the experimental results on which it is based.

Nevertheless, Sandars' idea of measuring the proton EDM via the nuclear magnetic resonance in polar molecules is very attractive. However, lower and more reliable bounds can probably be obtained by using molecules containing heavy nuclei with spins $i > \frac{1}{2}$ (i.e., cesium). Such a nucleus can have a magnetic quadrupole moment, so the effective electric field acting on the EDM of the valence proton will be larger than in the case of thallium, where it is due entirely to the fact that δr^2 differs from zero. The advantage is essentially the same as the advantage described above of atomic cesium over atomic thallium.

In concluding I want to point out the following circumstance. Despite the difference by a factor of 2×10^5 between bound (5.1) and the corresponding bound for the electron ($|d/e| < 3\times10^{-24}$ cm[8]), these two bounds may not differ so greatly in physical significance. The point is that in many models of the T-odd interaction (see the review by Wolfenstein in[20]) the EDM of an elementary particle is proportional to its mass (or to the quark mass). In this case the proton EDM will be $m_p/m \sim 2\times10^3$ times larger than the electron EDM. Of course from this point of view the best bound is doubtless that on the neutron EDM ($|d_n/e| < 10^{-23}$ cm[2]). However, the experimenters should not allow themselves to be hypnotized by these remarks, for we are talking about a poorly explored area of physics that may have big surprises in store for us.

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[1)]The modulus of ρ is determined from experimental data[13] on the cesium oscillator strengths. The sign is determined by the following considerations. In calculating the mixing of s and p states, the wave functions for both states were taken as positive in the limit $r \to 0$. Then, since the radial quantum numbers of the states differ by unity, the wave functions must have opposite signs in the limit $r \to \infty$. Hence ρ, which is determined mainly by the behavior of the wave functions at large distances, must be negative.

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