ELECTRIC DIPOLE MOMENT OF THE CESIUM ATOM. A NEW UPPER LIMIT TO THE ELECTRIC DIPOLE MOMENT OF THE FREE ELECTRON*

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The static electric dipole moment (edm) of a nondegenerate physical system in a well-defined angular-momentum state is zero if the Hamiltonian is invariant under time reversal. This was first pointed out by Landau,[1] and has been discussed by Lee and Yang.[2] The question of time-reversal invariance has assumed new importance because of a recent experiment of Christenson et al.[3] on the decay of the K_2^0 meson, where there are indications that it may be violated in weak interactions. Many workers have searched for an edm in elementary particles, in nuclei, and in atoms; their results are summarized in Table I. In this Letter we report an experiment which sets a new and smaller upper limit to the edm of a free atom and thus tests more stringently the time-reversal invariance of the electromagnetic interactions, a topic which has been discussed by Sachs and Schwebel.[4] Our result can also be interpreted in terms of the edm of the free electron to yield an upper limit several orders of magnitude smaller than that previously available.

We have used the atomic-beam method described in the preceding Letter, which enables us to measure shifts in the position of the resonance as small as 10^{-4} of a linewidth. A beam of cesium atoms passes between two closely spaced metal plates which lie between the loops of a Ramsey double-hairpin structure. The observable Zeeman transition $(4, -4) \leftrightarrow (4, -3)$ is induced and the signal adjusted to the point of maximum slope on one side of the central peak of the resonance, when any displacement of the resonance will be reflected with maximum sensitivity as a change in the ion current at the hot-wire detector. The resonance is in effect used as a displacement amplifier, a method similar to that used by Browne[5] in a paramagnetic-resonance experiment involving electric fields. Large dc and ac (25 cps) electric fields can be applied simultaneously to the beam as it passes between the plates. In an electric field E the resonance frequency will be shifted by an amount $\delta\nu = k_1E^2 + k_2E$, where the quadratic term is due to the Stark effect,[6] and the linear term to the assumed edm of the atom. If $E = E_{dc} + E_{ac}\sin\omega t$, then $\delta\nu$ will contain components at both ω and 2ω, either of which can be singled out by phase-sensitive detection. We are interested here in the component modulated at frequency ω which takes the form

$$\delta\nu(\omega) = [2E_{dc}k_1 + k_2]E_{ac}\sin\omega t. \qquad (1)$$

This equation implies that if the atom has an edm the observed frequency shift modulated at ω must satisfy the following conditions: (1) It must be linear in E_{dc}; (2) the shift must be nonzero for $E_{dc} = 0$; (3) the magnitude of the shift when $E_{dc} = 0$ must be proportional to E_{ac}; and (4) the shift when $E_{dc} = 0$ must be in phase with that when $E_{dc} \neq 0$.

Our experimental results for the cesium atom are illustrated in Figs. 1 and 2. In Fig. 1 we see that within experimental error the frequency shift is linear with E_{dc} and has a small nonzero value when $E_{dc} = 0$. In Fig. 2 we show the frequency shift for $E_{dc} = 0$ as a function of E_{ac}; again, within experimental error the relation is linear. In addition, we have not been able to detect any phase difference be-

Table I. Upper limits on some electric-dipole moments.

Particle	Electric-dipole moment	Reference
Electron[a]	1.2×10^{-15} cm$\times e$	a
Muon[b]	$2\ \times 10^{-16}$ cm$\times e$	b
Neutron[c]	2.4×10^{-20} cm$\times e$	c
Proton[d]	1.3×10^{-13} cm$\times e$	d
^{23}Na nucleus[e]	$1\ \times 10^{-14}$ cm$\times e$	e
Rb atom[f]	$1\ \times 10^{-18}$ cm$\times e$	f
Cs atom	2.2×10^{-19} cm$\times e$	Present experiment
Electron	$2\ \times 10^{-21}$ cm$\times e$	Present experiment

[a]See reference 10.
[b]D. Berley and G. Gidal, Phys. Rev. 118, 1086 (1960).
[c]J. H. Smith, E. M. Purcell, and N. F. Ramsey, Phys. Rev. 108, 120 (1957).
[d]R. M. Sternheimer, Phys. Rev. 113, 828 (1959).
[e]T. Ramsey and L. W. Anderson, Nuovo Cimento 32, 1151 (1964).
[f]E. S. Ensberg, Bull. Am. Phys. Soc. 7, 534 (1962).

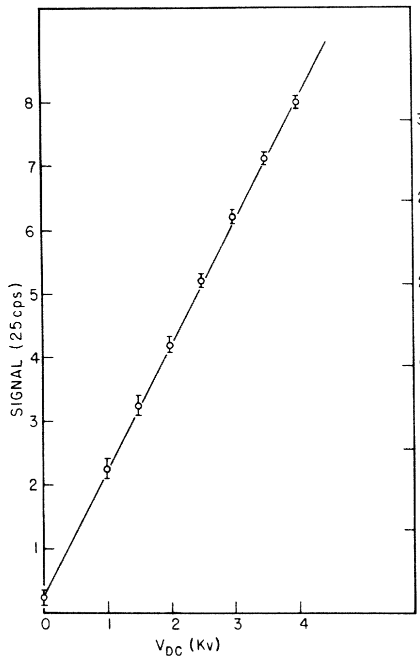

FIG. 1. Frequency shift modulated at ω vs dc voltage at fixed ac voltage.

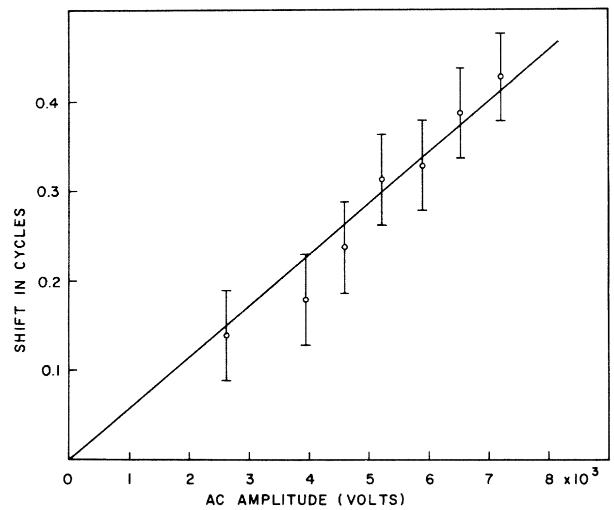

FIG. 2. Frequency shift modulated at ω vs amplitude of ac voltage with dc voltage equal to zero.

tween the shift when $E_{dc}=0$ and that when $E_{dc}\neq 0$. While these results are consistent with an edm for the cesium atom of $d_A=(2.2\pm 0.1)\times 10^{-19}$ cm$\times e$, we have not been able to rule out the possibility that they are due to an instrumental effect. The most likely cause appears to be the interaction between the magnetic moment of the atom and the motional magnetic field $H_1=v\times E$ experienced by the atom as it moves with velocity v through the electric field. With our geometry (E and H_0 parallel), this should produce a shift modulated at 2ω. However, a slight misalignment of order $\frac{1}{2}°$ would produce a shift of the observed magnitude. This should be proportional to v, and we have attempted to run beams of different velocities using ovens with heated snouts. The results have been inconclusive because of the limitations imposed by the signal-to-noise ratio when working with shifts of the order of 10^{-4} of the linewidth. Further experiments are in progress to ascertain if any part of the linear frequency shift is due to an edm. In spite of the uncertainty in the origin of the observed linear frequency shift, it is clear that the edm cannot be much larger than 2×10^{-19} cm$\times e$, the value which corresponds to the observed shift. This limit on the edm is sufficiently small that some interesting consequences follow.

We first interpret our result in terms of an edm of the free electron. This analysis must be done relativistically since one can show[7] that in the nonrelativistic limit an atom has no edm even if the constituent electrons do. Salpeter[8] has shown how an edm can be introduced into the single-electron Dirac equation in a Lorentz-covariant way. We assume that we can generalize this for a many-electron atom by adding to the unperturbed atomic Hamiltonian a perturbation

$$H_1=+d_e\sum_i\beta_i\vec{\sigma}_i\cdot\vec{\nabla}\varphi_i, \qquad (2)$$

where the sum over i spans the electrons, d_e is the edm of the free electron, and φ_i is the potential at the ith electron. The calculation on the effect of H_1 is reported elsewhere.[9] It is shown there that the expectation value of H_1 vanishes in the nonrelativistic approximation, but that if relativity is taken into account the edm of the atom is some 100 times bigger than that of the free electron. A similar relativistic enhancement of the electron edm was found by Salpeter[8] in his analysis of a related problem involving the $2s_{1/2}$ state of the hydrogen atom. Comparing the theoretical estimate above with our experimental limit on the edm of the atom, we deduce an upper limit on the electron edm of $d_e\lesssim 2\times 10^{-21}$ cm $\times e$. This limit is a factor 10^5 smaller than that previously deduced from high-energy scattering.[10]

An alternative way of looking at our results is in terms of the purity of the cesium ground state. If some p state is present so that the wave function takes the form $|s\rangle + a|p\rangle$, then the dipole moment of the atom is given by

$$d_A = -e\langle s|\vec{r}|p\rangle(a+a^*), \qquad (4)$$

where we have adopted the phase convention in which the matrix element of r is real. We assume that any admixture is due to an internal interaction within the atom. Such an interaction must violate parity in order to admix a p state into an s state. It must also violate time-reversal invariance in order to ensure that the admixture coefficient a is real; otherwise the dipole moment will vanish. From our measured limit on the edm of the atom we deduce that

$$|a| \leq 2\times10^{-12},$$

where we have obtained a rough value of the matrix element of r from the atomic polarizability.[11] To illustrate the order of magnitude of this limit, we calculate the admixture of p state into the ground s state in cesium that one would expect due to parity-nonconserving weak interactions. Carhart[12] has calculated the admixture of $2p$ state into the $2s$ state of hydrogen. His theory can readily be applied to cesium, and we find $|a| \approx 10^{-12}$. This admixture is pure imaginary so that $a+a^*=0$, and the edm is zero as it must be since Carhart's theory is time-reversal invariant. Nonetheless, the fact that weak interactions can produce admixtures of the same order as our experimental limits suggests that if time-reversal invariance is violated in the weak interactions a measurable edm of the cesium atom might result.

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[1]L. Landau, Nucl. Phys. 3, 127 (1957).
[2]T. D. Lee and C. N. Yang, Brookhaven National Laboratory Report No. BNL-443 (F91), October 1957, p. 17 (unpublished).
[3]J. H. Christenson, J. W. Cronin, V. L. Fitch, and R. Turlay, Phys. Rev. Letters 13, 138 (1964).
[4]M. Sachs and S. L. Schwebel, Ann. Phys. (N.Y.) 6, 244 (1959); 8, 475 (1959).
[5]M. E. Browne, Phys. Rev. 121, 1699 (1961).
[6]E. Lipworth, S. Legowski, and P. G. H. Sandars, Bull. Am. Phys. Soc. 9, 91 (1964).
[7]L. I. Schiff, Phys. Rev. 132, 2194 (1963).
[8]E. E. Salpeter, Phys. Rev. 112, 1642 (1958).
[9]P. G. H. Sandars, to be published.
[10]J. Goldemberg and Y. Torizuka, Phys. Rev. 129, 2580 (1963).
[11]G. E. Chamberlain and J. C. Zorn, Phys. Rev. 129, 677 (1963).
[12]R. A. Carhart, Phys. Rev. 132, 2337 (1963).

MULTIMODE EFFECTS IN STIMULATED RAMAN EMISSION

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The multimode structure of high-power Q-switched lasers has been shown to have an important effect on the second-harmonic generation of light.[1] The present note calls attention to the even more pronounced effects of the spatial and temporal distribution of the laser modes on the stimulated Raman emission.

Consider the generation of Stokes radiation by two laser modes, represented by plane waves at the same frequency ω_L but traveling in slightly different directions. The wave vectors are denoted by $\vec{k}_L$ and $\vec{k}_L'$, with corresponding complex amplitudes E_L and E_L'. The usual notion is that the gain for each mode at the Stokes frequency is proportional to the laser intensity $|E_L|^2+|E_L'|^2$. Consider, however, the pair of Stokes modes for which $\vec{k}_L-\vec{k}_S=\vec{k}_L'-\vec{k}_{S'}$. The coupled amplitude equations[2] describing the growth of these modes are

$$dE_S/dz = C\{|E_L|^2+|E_L'|^2\}E_S + CE_L^{*\prime}E_LE_{S'},$$

$$dE_{S'}/dz = CE_L'E_L^*E_S + C\{|E_L|^2+|E_L'|^2\}E_{S'}, \qquad (1)$$

where the constant C is related to the Raman susceptibility at resonance $\chi_S''(0)$ by

$$C = (2\pi\omega_S^2/c^2k_{zS})|\chi_S''(0)|. \qquad (2)$$