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PHYSICAL REVIEW A

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Electric Dipole Moment of the Thallium Atom*

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Results of a search for the electric dipole moment of the thallium atom using previously described techniques and apparatus are reported.

Previously described^{1,2} atomic-beam techniques and apparatus were used to search for an electric dipole moment (EDM) of the Tl atom. The motivation for the Tl experiments was twofold: (1) Such experiments are sensitive to $\vec{1} \cdot \vec{E}$ type parity and time-reversal violating interactions³ because Tl has a *P* ground state whereas alkali atoms used in the previous work have *S* ground states, and (2) the relativistic enhancement of an assumed electron EDM in Tl may be comparable to that for Cs because of thallium's high atomic number (*Z* = 81)⁴.

Experiments were performed in the same way as those for alkali atoms^{1,2}. The flop-in transition $(F=1, m_F=0) \leftrightarrow (F=1, -1)$ in the $6^2P_{1/2}$ ground state of Tl was monitored. The transition was induced in a Ramsey double-hairpin structure in a uniform magnetic field (\dot{H}) of a few gauss. Intense beams of Tl, which lasted for more than 24 h of running time, were produced by a high-temperature stainless-steel oven.⁵ Tl was detected by surface ionization on a hot platinum strip. The ions were collected by a negatively biased copper cylinder which surrounded the Pt wire. A quadrupole mass spectrometer⁶ was used to identify T1. The spectrometer was not used, however, during the electric field experiments. In spite of thallium's low ionization efficiency on Pt (~1 to $2\%^7$), we were able to detect Tl with a signal-to-noise ratio comparable to that achieved in the previously reported^{1,2} alkali-atom experiments. This is attributed to our ability to form TI beams of sufficient intensity to compensate for the low ionization efficiency. At resonance, the recovered beam was $\sim 25 \times 10^{-10}$ A. The width at half-intensity of the central Ramsey peak was ~ 1.5 kc. The details of the apparatus have been published elsewhere in the literature.⁸

To detect a possible EDM in Tl, a large ac voltage was applied to parallel metal electric field plates situated between the rf loops of the Ramsey double-hairpin structure. The shift δf in the Bohr resonance frequency due to the applied electric field (\vec{E}) is written

$$\delta f = k_1 E^2 + k_2 E , \qquad (1)$$

where k_1E^2 represents the quadratic Stark interaction and k_2E represents a possible EDM effect plus instrumental effects simulating an EDM signal. Applying a large 25-cps voltage alone to the plates, only k_2E produces a 25-cps component in the detector signal (assuming no harmonics in the ac voltage). This signal is separated from the 50cps component due to the quadratic Stark interaction by means of phase-sensitive detection techniques. The system is calibrated by observing the signal at the detector corresponding to frequency modulation of the resonance rf by a known amount.

A 25-cps component of the detector signal was observed when ac alone was applied. Figure 1 shows typical behavior of this signal (expressed as a shift δf in the Bohr resonance frequency) as a function of the amplitude of the ac field. The curvature was caused by harmonics of the fundamental frequency in the output of the high-voltage transformer. This harmonic effect was observed in the alkali-atom EDM experiments,² and the method used there to subtract it out was applied to Tl. We determined $k_2 = (4.4 \pm 0.2) \times 10^{-5} \text{ cps}/(\text{V/cm})$ where the uncertainty is the standard deviation of the mean, based on nine experiments. The "filling factor" correction to account for the fact that the electric field plates do not extend the entire distance between the rf loops has already been included.

Also, the shift in the resonance frequency due to

the quadratic Stark interaction was observed. This was accomplished in two ways: (i) Measuring the 50-cps component in the signal at the hot wire detector as a function of \vec{E} ; and (ii) applying ac and dc to the plates and measuring the 25-cps component in the detector signal as a function of the dc. Results by both methods agree within experimental error. A preliminary result is k_1 $=(-4.1\pm0.3)\times10^{-8}$ cps/(V/cm)², where the large uncertainty was adopted to cover the likelihood that a magnetic instrumental effect was interfering with the measurements. Instrumental effects possibly affecting k_1 have been discussed in Ref. 8. A theoretical estimate of the magnitude of k_1 is not as straightforward as that provided in Ref. 8 for the alkalis, because we cannot assume with much accuracy that the polarizability is due to a single state admixture by \vec{E} . Significant contributions from the near-lying 6D and 7S states are expected.

1

While our result for k_2 is consistent with an EDM of the TI atom of $|D_{\text{TI}}|/e = 5 \times 10^{-19}$ cm, ¹⁰ it is likely that k_2 was due to an instrumental effect. The principal instrumental effect appeared to be the interaction between the magnetic dipole moment of the atom and the motional magnetic field $(\bar{\nabla}/c) \times \vec{E}$ experienced by the atom as it moves with velocity $\bar{\nabla}$ through the electric field \vec{E} .^{1, 2} A resonance shift, linear in \vec{E} , results if \vec{E} is not parallel to \vec{H} . As discussed in Ref. 2, this effect was studied in several alkalis having different magnetic moments and velocities. We did not pursue a similar study for TI. Instead, the present type of experiment was abandoned in favor of



digital signal-processing techniques and a longer resonance region using rectangular Helmholtz coils to produce \vec{H} .^{5, 11} The $(\vec{\nabla}/c) \times \vec{E}$ effect is then controlled electronically. Also, other instrumental effects capable of simulating an EDMproduced signal, such as the harmonic effect and a displacement-current effect, are eliminated or substantially reduced. Experiments on Tl using

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In spite of our uncertainty in the magnitude of instrumental effects contributing to k_2 , it is unlikely that the EDM of the Tl atom is much larger than $5 \times 10^{-19} e$ cm which establishes for the first time a limit to the EDM of a non-S-state atom.

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PHYSICAL REVIEW A

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Microwave Spectrum of Molecular Oxygen[†]

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This paper discusses an error in an earlier paper by West and Mizushima.

The microwave spectrum of oxygen results from transitions between fine-structure levels of the ${}^{3}\Sigma \overline{g}$ molecular ground state. The spectrum comprises a number of lines near 60 GHz and a single line at 118.750 GHz. Transitions between the levels J = K + 1 are termed K^{+} lines, and between J = K and J = K - 1, K^{-} lines. J is the total angular momentum, and K, the rotational angular momentum, is constrained to odd values by the Bose sym-

metry of the two identical O¹⁶ nuclei. The Hamiltonian for the fine structure of oxygen is¹

$$H = B\left(\vec{K}\right)^2 + \frac{2}{3}\lambda\left(3S_{z} - (\vec{S})^2\right) + \mu \vec{K} \cdot \vec{S}$$

S is the spin angular momentum operator, and S_z is its component along the internuclear axis. The term in *B*, the rotational constant, is the en-