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Pressure Shifts of Hyperfine Splitting of Hydrogen and Tritium in Argon*†

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This paper reports new measurements of the pressure shifts in argon of the hyperfine splitting of hydrogen and tritium. A spin-exchange optical pumping technique was used to make these measurements. The fractional pressure shift (pressure shift in cps/mm Hg at 0°C divided by the hyperfine splitting in kMc/sec for hydrogen in argon is -4.78 ± 0.03; the ratio of the fractional pressure shift for tritium to that for hydrogen is 1.007 ± 0.012; the temperature shift at constant density of the hydrogen hyperfine splitting is 0.012 ± 0.003 cps/°K mm Hg. This new value for the hydrogen pressure shift is then used to reanalyze the Yale data for the hyperfine splitting of muonium, under the hypothesis that the fractional pressure shift for muonium is the same as that for hydrogen. This gives for the muonium hyperfine splitting an alternative value of 4463.23(2) Mc/sec. This value of the muonium hyperfine splitting gives for the fine-structure constant α the alternative value $\alpha^{-1} = 137.0367(10)$. This value of α is compared with the values obtained from the hydrogen fine structure, hydrogen hyperfine structure, and the ac Josephson-effect determination of $2e/h$.

INTRODUCTION

Until a year ago, the accepted value for the fine-structure constant was determined from measurements by Dayhoff, Triebwasser, and Lamb[1] of the fine-structure splitting ($n=2$, ${}^2P_{3/2} \to {}^2P_{1/2}$) in atomic deuterium.[2] Their value for the fine-structure interval was obtained by adding together separate measurements of the $n=2$, ${}^2S_{1/2} \to {}^2P_{1/2}$ fine-structure interval (Lamb shift) and the $n=2$, ${}^2S_{1/2} \to {}^2P_{3/2}$ fine-structure interval. These measurements gave for the fine-structure (FS) constant the value

$$\alpha^{-1}(\mathrm{FS}) = 137.038\,8(6) \quad (4\ \mathrm{ppm}). \tag{1}$$

The error given here and elsewhere in the paper is one standard deviation. In some cases where the authors have used limits of error, we have reduced the error so that it corresponds to one standard deviation.

This value for the fine-structure constant was confirmed by the Yale measurements on the hyperfine structure of the muonium atom. Cleland, Bailey, Eckhause, Hughes, Mobley, Prepost, and Rothberg[3] made a precise measurement of the muonium hyperfine splitting and then, assuming that the muon was a point particle adequately described by quantum electrodynamics, calculated from the measured splitting a new value for the fine-structure constant. They obtained

$$\alpha^{-1}(\mathrm{M}) = 137.038\,8(12) \quad (9\ \mathrm{ppm}). \tag{2}$$

This value agrees very well with that of Dayhoff *et al.*

Recently this value for the fine-structure constant has been challenged from several different directions. First, Drell and Sullivan[4] have reanalyzed the proton-structure corrections to the hyperfine splitting of hydrogen and have tried to understand the 44 ± 20 ppm discrepancy between the experimental value of the hydrogen hyperfine splitting and the theoretical value calculated using the Lamb value for α and the proton-structure factor calculated by Iddings.[5-8] Drell and Sullivan could not find with certainty additional corrections that would change the structure correction factor, but they concluded that calculations of the proton-structure correction were sufficiently uncertain so that hitherto uncalculated contributions to the hydrogen hyperfine structure due to the proton's polarizability may very well be as large as 10 ppm. Thus Drell and Sullivan were unable to remove the discrepancy, and they were sufficiently uncertain about the calculation so that they would not say how much of the discrepancy was due to quantum electrodynamics and how much was due to the proton structure. The value of α derived from the hydrogen hyperfine structure (HHS) is

$$\alpha^{-1}(\mathrm{HHS}) = 137.0382(1 - \tfrac{1}{2}\delta_p). \tag{3}$$

Here δ_p is the structure correction factor for the proton. If we take δ_p to be 35 ± 3 ppm, as calculated by Iddings, we obtain for α the value

$$\alpha^{-1}(\mathrm{HHS}) = 137.0359(4) \quad (3\ \mathrm{ppm}). \tag{4}$$

Secondly, Ruderman[9] has pointed out that the value of the ratio of the muon magnetic moment to the proton magnetic moment used by Cleland *et al.* in their determination of the value of α from the muonium hyperfine structure has not been properly corrected for diamagnetic shielding in the water sample in which the g-factor measurements were made. Because of its lower mass and higher zero-point energy, a μ^+ meson can form a type of bond between water molecules which is considerably stronger than the usual hydrogen bond. Ruderman estimates that the chemical shift of the μ^+ is 15–20 ppm less than that of the proton with which it is compared. When Ruderman makes this correction and then uses the data of Cleland *et al.* to determine the fine-structure constant, he obtains

$$\alpha^{-1}(\mathrm{MR}) = 137.0377(13) \quad (10\ \mathrm{ppm}). \tag{5}$$

Thirdly, Parker, Taylor, and Langenberg[10] have used the ac Josephson effect to measure $2e/h$, and from this have calculated a new value for α. They obtain

$$\alpha^{-1}(\mathrm{J}) = 137.0359(4) \quad (3\ \mathrm{ppm}). \tag{6}$$

This value of α differs by 21 ± 5 ppm from the value of Dayhoff *et al.*; it agrees very well with the value derived from the hydrogen hyperfine structure using Iddings's value for the proton-structure correction.

Fourthly, Metcalf, Brandenberger, and Baird[11] have recently reported a new measurement of the fine-structure constant using an optically detected level crossing (LC) in atomic hydrogen. They obtain

$$\alpha^{-1}(\mathrm{LC}) = 137.0353(8) \quad (6\ \mathrm{ppm}). \tag{7}$$

This value agrees with the Josephson-effect measurements, and disagrees with the value Dayhoff *et al.* obtained for α^{-1}.

Thus at present it is attractive to believe that the measurements due to Dayhoff *et al.* are somewhat in error, that the correct value of the fine-structure constant is given by the Josephson-effect measurement, and that the calculated value for the proton-structure correction to the hydrogen hyperfine splitting is correct. This leaves somewhat in doubt the value of the fine-structure constant derived from the muonium measurements. Are these measurements in error? Is there an unknown correction to the structure of the muon? The purpose of this paper is to suggest an alternative method for analyzing the muonium data,[12] which, when combined with the Ruderman correction for the ratio of the muon magnetic moment to the proton magnetic moment, gives for the fine-structure constant the value

$$\alpha^{-1}(\mathrm{MR2}) = 137.0367(10) \quad (8\ \mathrm{ppm}). \tag{8}$$

This value agrees with the value from the measurements of the ac Josephson effect.

APPROACH TO THE PROBLEM

To determine the muonium hyperfine structure interval, Cleland *et al.* measured one of the muonium Zeeman transitions in a high magnetic field and in the presence of argon at high pressure. The argon perturbed the muonium atoms, and produced a change in the muonium hyperfine interval which was proportional to the argon density. To determine the hyperfine splitting of the free muonium atom, they made measurements at several pressures and extrapolated to zero pressure, a procedure which has been used in many optical pumping measurements of hyperfine intervals.[13] Cleland *et al.* found for muonium a zero-field hyperfine splitting of

$$\Delta\nu(\mathrm{M}) = 4463.15(6)\ \mathrm{Mc/sec} \quad (13\ \mathrm{ppm}), \tag{9}$$

and a fractional pressure shift $(1/\Delta\nu)d\Delta\nu/dP$ for muonium in argon at 0°C of

$$(-4.05 \pm 0.49) \times 10^{-9} (\mathrm{mm\ Hg})^{-1}. \tag{10}$$

This value for the fractional shift is somewhat less than the values found in optical pumping measurements on the hydrogen isotopes. These values are[14]

$$\frac{1}{\Delta\nu}\frac{d\Delta\nu}{dP}(\mathrm{hydrogen}) = (-4.77 \pm 0.12) \times 10^{-9}\ (\mathrm{mm\ Hg})^{-1}, \tag{11}$$

$$\frac{1}{\Delta\nu}\frac{d\Delta\nu}{dP}(\mathrm{deuterium}) = (-4.52 \pm 0.40) \times 10^{-9}\ (\mathrm{mm\ Hg})^{-1}, \tag{12}$$

$$\frac{1}{\Delta\nu}\frac{d\Delta\nu}{dP}(\mathrm{tritium}) = (-5.05 \pm 0.15) \times 10^{-9}\ (\mathrm{mm\ Hg})^{-1}. \tag{13}$$

On theoretical grounds,[15, 16] one expects the fractional pressure shift for muonium to be the same as the fractional pressure shifts for the hydrogen isotopes. From the standpoint of the pressure shifts, muonium is only a light hydrogen atom; the variation in atomic mass from muonium to hydrogen is only three times the variation from hydrogen to tritium. Theory indicates that the pressure shifts are, to first order, independent of the reduced mass of the colliding hydrogen-argon atoms. By making spin-exchange optical pumping measurements on a bulb containing hydrogen, tritium, and argon, one can measure the fractional pressure shift for hydrogen-like atoms and study the isotopic dependence of this pressure shift.

One may then use the more precise value for the fractional pressure shift found in the hydrogen spin-exchange optical pumping experiments to reanalyze the muonium data. This eliminates one of the unknowns and permits a more precise extrapolation to zero pressure. This paper reports a spin-exchange optical pumping measurement of the fractional pressure shifts of hydrogen and tritium, and the values of the muonium hyperfine splitting and the fine-structure constant obtained when this value of the pressure shift is used to extrapolate the muonium measurements to zero pressure.

APPARATUS

The spin-exchange optical pumping spectrometer shown diagrammatically in Fig. 1 has been described in some detail in previous publications.[17] The most essential feature not employed in the earlier experiments on hydrogen is the use of three concentric high-permeability magnetic cylinders to shield the sample from external magnetic disturbances.[18] A specially constructed solenoid powered by a mercury cell was used to produce an axial field of 200 mG at the sample. A World War II surplus APT-5 transmitter was used to drive the radio-frequency transitions of hydrogen and tritium. This radio-frequency source was stabilized by phase-locking it to the output of a Gertsch FM-4 which was driven by a Gertsch AM-1.

The 1-Mc/sec frequency standard for the Gertsch AM-1 was derived from oscillators maintained and monitored by J. A. Pierce of the Harvard Depart-

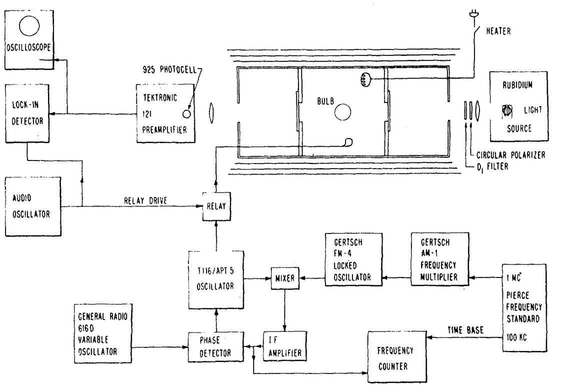

FIG. 1. A block diagram of the optical pumping apparatus. The magnetic field is produced by a precision solenoid which is surrounded by three concentric cylindrical magnetic shields.

ment of Applied Physics. The primary reference frequency was generated by a General Radio 1120 AH 1000 frequency standard, the oscillator of which was locked to the average frequency of a cesium-beam Atomichron and of a Varian optically pumped rubidium frequency standard. The General Radio frequency was continuously compared with the Loran C and National Bureau of Standards standard frequencies. These frequencies are maintained constant each year with reference to atomic standards of frequency, but they are offset to keep the time pulses in close agreement with the UT2 scale, which is used for navigation. The atomic or A1 time scale is such that the Cs^{133} hyperfine splitting is exactly 9 192 631 770 cps. All of the frequencies quoted in this paper are expressed in terms of the A1 time scale.

PREPARATION OF ABSORPTION BULBS

The following procedure was used to prepare 300-cm^3 spherical absorption bulbs containing rubidium metal, an isotope of hydrogen, and an accurately known pressure of argon. Figure 2 shows a schematic diagram of the vacuum and gas-handling system. Each bulb was provided with two glass-covered tungsten electrodes which were used together with a radio-frequency source to dissociate the molecular hydrogen. The system was constructed of glass and a small Varian Vac Ion pump was used to evacuate the system. A Texas Instruments Co. quartz bourdon-tube pressure gauge was used to measure the gas pressure. The calibration of this gauge was checked against a precise barometer.

The entire system was initially pumped out to 10^{-8} mm Hg, and the bulb was outgassed at high temperature. Gentle heating of the rubidium reservoir was then used to distill gradually the rubidium metal into the absorption bulb. As soon as the rubidium reservoir was empty, it was sealed off and removed. By heating the upper surface and simultaneously cooling the lower tip of the bulb, the sides of the bulb were cleaned off and the rubidium was moved to the bottom of the bulb.

Gas was then admitted to the system and the ratio of the volume of the gas manifold and the bulb was measured so that the proper corrections could be made when the argon and hydrogen were mixed and when the bulb was heated and sealed off. After the completion of the volume measurements the bulb was filled with a small amount of hydrogen and the desired amount of argon. The best signals were obtained with a 2 mm Hg partial pressure of hydrogen. The bulbs containing tritium and hydrogen were prepared by attaching to the main absorption flask a small sealed-off ampoule containing the desired amount of tritium. A breakoff seal was provided so that the tritium could be admitted into the flask after it had been filled with rubidium, argon, and hydrogen and sealed off from the vacuum system. The ampoule and breakoff seal were then sealed off and removed from the system. Bulbs were prepared with argon and hydrogen, and with argon, hydrogen, and tritium. The bulb pressures quoted in this paper are the pressures corrected to 0° C; the pressure-shift measurements were made at temperatures from 30 to 45° C.

MEASUREMENT PROCEDURE

The following procedure was used to take the data. The desired absorption flask was inserted into the oven inside the shielded solenoid system, and the bulb was heated. The $\Delta F = 0$, $\Delta m = \pm 1$ Rb^{87} Zeeman transitions were measured and the magnetic shield system was demagnetized until the full width at half maximum of the Rb^{87}(700 cycles/mG) Zeeman transitions was less than 50 cps. The discharge was then turned on, and the hydrogen $\Delta F = 0$, $\Delta m = \pm 1$ Zeeman transitions were observed. The discharge was adjusted so as to optimize the signal to noise ratio, and minimize the linewidth. The high-frequency signal source was then connected up, and the $\Delta F = 1$, $\Delta m = \pm 1$ hydrogen hyperfine transitions were located. When these transitions were found, the linewidth was measured and the temperature of the bulb and the discharge were varied to optimize the signal-to-noise ratio and minimize the linewidth. Measurements were then started. During the measurements, the linewidth of the hyperfine transitions was usually about 150 cps.

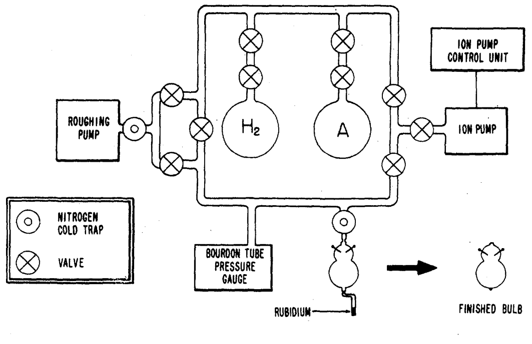

FIG. 2. A diagram of the vacuum system used to prepare the absorption bulbs. The pressures were measured with a Texas Instruments Co. quartz bourdon-tube pressure gauge.

TABLE I. This table gives all the measurements of the bulb containing hydrogen and 115.3 mm Hg of argon. Each measurement was obtained by averaging two observations of the $\Delta F = 1$, $\Delta m = +1$ hyperfine transition and two measurements of the $\Delta F = 1$, $\Delta m = -1$ hyperfine transition. The error in the bulb average was determined by treating each of the data sets as a separate measurement and then using the standard formula to calculate the standard deviation.

Evening on which measurements were made	Circular polarization of rubidium light	Measured frequencies (cps)	Average frequency for data set (cps)	Standard deviation of average
1	+	1 420 404 976 77 71 67 65	1 420 404 971	5
1	+	1 420 404 960 62 62 54 60	1 420 404 960	3
2	+	1 420 404 979 79 80 79 84	1 420 404 980	2
2	+	1 420 404 966 68 65 68 70	1 420 404 967	2
2	–	1 420 404 982 78 80 76 72	1 420 404 978	3
2	–	1 420 404 970 66 59 65 63	1 420 404 965	4
3	–	1 420 404 984 80 79 75 74 80 79	1 420 404 979	3
3	–	1 420 404 987 81 80 75 78	1 420 404 980	4
3	–	1 420 404 974 75 79 85 78	1 420 404 978	4
3	–	1 420 404 985 89 85 89 84	1 420 404 986	2
Normal average for all data sets			$\bar{\nu}_n = 1\,420\,404\,974(8)$	
Weighted average for all data sets			$\bar{\nu} = 1\,420\,404\,976(8)$	

In the weak field of the solenoid, the triplet hyperfine component of the hydrogen ground state is resolved into three Zeeman levels. Only the $\Delta F = 1$, $\Delta m = \pm 1$ transitions can be observed using the spin-exchange optical pumping technique. Measurements were made of the $\Delta F = 1$, $\Delta m = +1$ transition and of the $\Delta F = 1$, $\Delta m = -1$ transition, and an average was taken to determine the zero-field hyperfine splitting. The difference of the two frequencies was used to calculate the second-order correction due to the field-dependent behavior of the $F = 0$, $m = 0$ state.

Two people were used to take data; one observer used the lock-in detector to set the variable frequency oscillator at the center of the line; the other observer recorded the frequency of the oscillator. The observer first measured one of the hyperfine transitions twice, and then measured the other hyperfine transition twice. The four measurements were averaged to give one value for the hyperfine splitting. Five such consecutive values were taken at one time and grouped together as a data set. The two observers were then interchanged. Occasionally, when the two observers were interchanged, the relative orientation of the linear polarizer and quarter-wave plate was changed by 90°, so that the circular polarization of the rubidium light incident on the sample was changed from right to left or vice versa. This changed the sign of the rubidium polarization and served to average out any polarization-dependent spin-exchange frequency shifts. For each bulb at least four data sets were taken. Special precautions were taken not to take all the data for one bulb on the same evening and with the same magnetic field configuration.

This procedure was employed to average out systematic errors that might be caused by small asymmetries in the magnetic-field-dependent Zeeman lines. All the data sets for one bulb were weighted by their standard deviations and averaged to obtain the value of the hyperfine splitting for that particular bulb. Table I gives a summary of the mea-

TABLE II. A summary of the measured hyperfine frequencies for the bulbs containing hydrogen and argon. The * denotes the average of the various data sets weighted by the standard deviation of the individual data sets; the † denotes the straight average with no weighting. In each case the error in the average was determined by using each of the data sets as an independent measurement and then using the usual formula to compute the standard deviation.

Pressure (mm Hg) at 0°C	$\Delta\nu$ (cps)	σ (cps)	Number of data Sets
45.5 ± 0.5	1 420 405 441*	13.6	16
	1 420 405 445†	13.1	
90.4 ± 0.9	1 420 405 142*	10.0	19
	1 420 405 143†	10.0	
115.3 ± 1	1 420 404 976*	7.9	10
	1 420 404 974†	7.8	
208 ± 2	1 420 404 338*	8.6	15
	1 420 404 338†	8.6	
271 ± 3	1 420 403 893*	24.	4
	1 420 403 886†	23.	

surements for a typical bulb. The results of all the measurements are tabulated in Table II. Figure 3 gives histograms summarizing the measurements on the hydrogen bulbs.

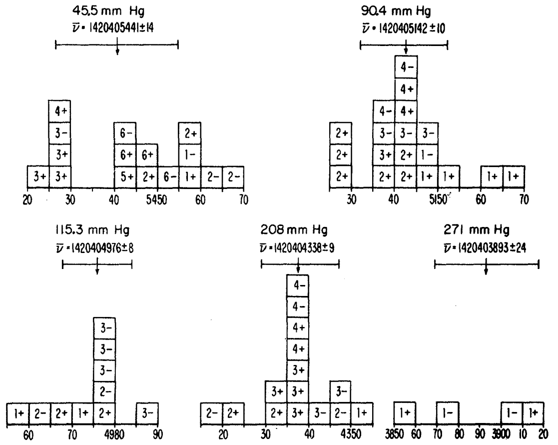

FIG. 3. Histogram of the data set averages for each of the hydrogen absorption bulbs. Each data set is the average of at least 20 separate measurements of the hyperfine splitting. The numbers in the boxes refer to the evening on which the measurements were made; the + or − refer to the circular polarization of the rubidium resonance radiation.

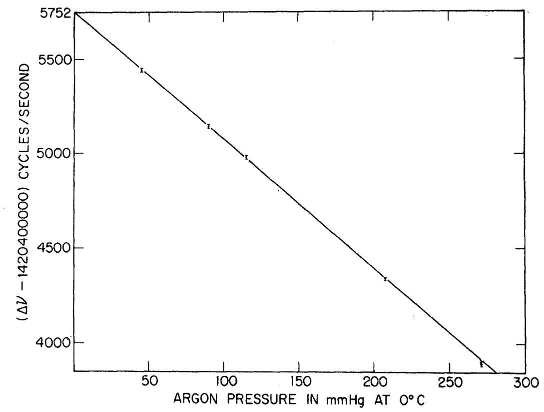

FIG. 4. A plot showing the measured hydrogen hyperfine splitting as a function of the argon pressure in the absorption bulb.

RESULTS FOR HYDROGEN ISOTOPES

Figure 4 shows the measured hydrogen hyperfine splitting plotted as a function of the argon pressure. There are two ways in which a least-squares procedure can be used to determine from the data the fractional pressure shift for hydrogen in argon. Crampton, Kleppner, and Ramsey[19] used a hydrogen maser to make a precise determination of the hydrogen hyperfine splitting. They found the value

$$\Delta\nu(\mathrm{H}) = 1\,420\,405\,751.800(28) \text{ cps}. \tag{14}$$

We can fit our measurements to a straight line of the form

$$\Delta\nu = \Delta\nu_0 + \alpha P \tag{15}$$

by either including the value due to Crampton *et al.* as the known zero-pressure value, or leaving the zero-pressure intercept as an unknown and then interpreting agreement between the value obtained by a least-squares fit and the value due to Crampton *et al.* as an indication of the reliability of our measurements. Table III gives the results

TABLE III. Results of using the method of least squares to fit the pressure-shift data to a straight line of the form $\Delta\nu = \Delta\nu_0 + \alpha P$. Here $\Delta\nu_0$ is the zero-pressure intercept and P is the pressure of argon in mm Hg at 0°C. Fit *B* uses only the data of this experiment; fit *A* includes with the data as the zero-pressure value the maser measurement by Crampton *et al*.

	Pressure shift α (cps/mm Hg)	Zero-pressure hyperfine splitting $\Delta\nu_0$ (cps)	Fractional pressure shift $(\alpha/\Delta\nu_0) \times 10^9$ (mm Hg) $^{-1}$
A	−6.79(4)	1 420 405 752(1)	−4.78(3)
B	−6.84(8)	1 420 405 760(13)	−4.82(6)

TABLE IV. The measured hyperfine frequencies for hydrogen and tritium in a bulb with an argon pressure of 90.8 ± 2.0 mm Hg at 0°C and the calculated value for the fractional pressure shift. The maser measurements of the zero-field hyperfine splitting of tritium [Phys. Rev. 158, 14 (1967)] and hydrogen [Phys. Rev. Letters 11, 338 (1963)] were used together with these data to determine the fractional pressure shift. The * denotes the weighted average of the various data sets; the † denotes the straight unweighted average.

Hydrogen isotope	Hyperfine splitting $\Delta\nu$ (cps)	Standard deviation σ (cps)	Fractional pressure shift $\frac{\Delta\nu-\Delta\nu_0}{P\Delta\nu_0}\times 10^9$
Tritium	1 516 700 821*	5.2	-4.72(4)
	1 516 700 819†	4.6	-4.74(3)
Hydrogen	1 420 405 146*	5.5	-4.70(4)
	1 420 405 147†	5.5	-4.69(4)

of fitting the data using these two hypotheses, showing that the data reported in this experiment yield a value of the hydrogen hyperfine splitting in agreement with the maser value and also that the value obtained for the fractional pressure shift is not sensitive to the mode of analysis. The best value for the fractional pressure shift for hydrogen in argon is

$$\frac{1}{\Delta\nu}\frac{d\Delta\nu}{dP}(\mathrm{H}) = (-4.78\pm0.03)\times10^{-9}\ (\mathrm{mm\ Hg})^{-1} \qquad (16)$$

at 0°C. This value is in good agreement with the values obtained in earlier optical pumping experiments [Eqs. (11), (12), and (13)]. The χ^2 for this fit is 1.63; one expects a χ^2 greater than this 80% of the time.

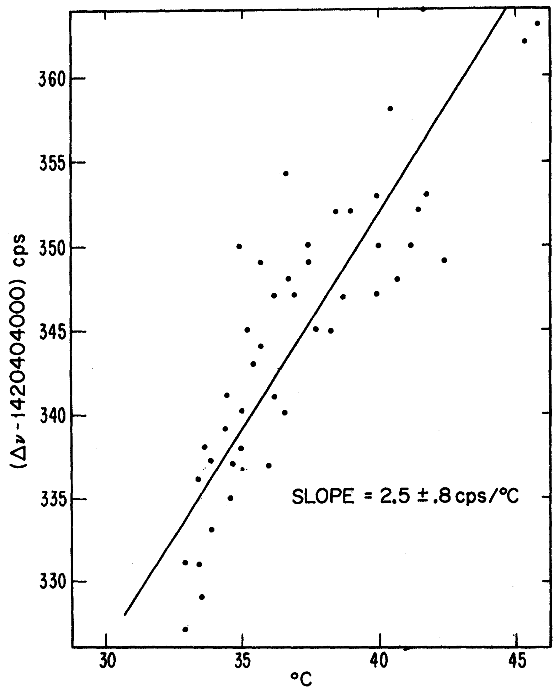

FIG. 5. A plot showing the hydrogen hyperfine splitting as a function of temperature for the bulb with an argon pressure of 208 mm Hg.

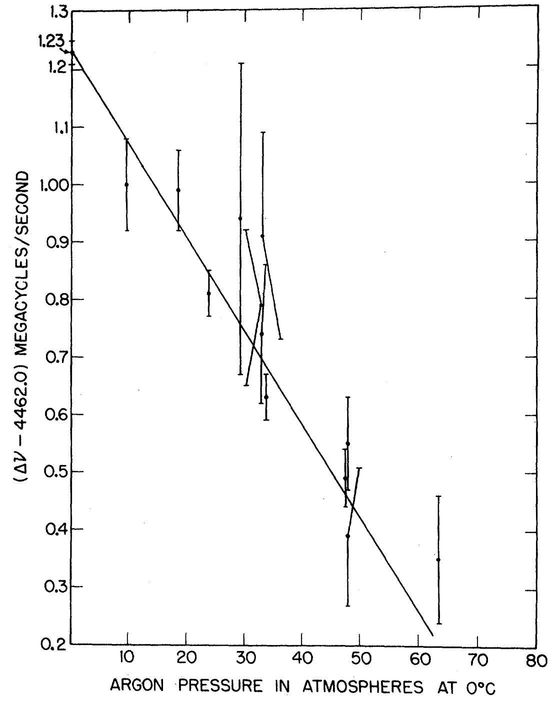

FIG. 6. A plot showing the muonium measurements of Cleland *et al.* as a function of argon pressure together with the straight-line fit to the data using the fractional pressure shift measured for hydrogen in this experiment.

A bulb containing hydrogen, tritium, rubidium, and 90.8 mm Hg of argon was used to measure the ratio of the fractional pressure shift for tritium to that for hydrogen. Table IV summarizes the results of these measurements. The maser measurement of the tritium hyperfine splitting[20] was used to calculate the tritium pressure shift. From these observations we conclude that the ratio of the fractional pressure shift for tritium to that for hydrogen is

$$1.007\pm0.012. \qquad (17)$$

The temperature dependence of the apparent hydrogen hyperfine interval was measured in the absorption bulbs containing 115.3 and 208 mm Hg argon. Figure 5 shows the data for the 208 mm Hg bulb. From these measurements we conclude that the temperature shift of the hydrogen hyperfine interval at constant argon density is

$$\frac{1}{P}\frac{\partial\Delta\nu(\mathrm{H})}{\partial T}\bigg|_{P_A} = 0.012\pm0.003\ \mathrm{cps}/{}^\circ\mathrm{K\ mm\ Hg}. \qquad (18)$$

APPLICATION TO MUONIUM

In this section we shall use the value of the fractional pressure shift found in this experiment to reanalyze the muonium measurements of Cleland *et al.* Figure 6 shows a least-squares fit to the muonium data,[3,21] using the result given in Eq. (16) for the fractional pressure shift. The χ^2 for this fit is 17.8; one expects a χ^2 greater than this 5% of the time. The zero-pressure muonium hyperfine interval determined by this extrapolation procedure is

$$\Delta\nu(\mathrm{M2}) = 4463.23(2)\ \mathrm{Mc/sec}\quad (5\,\mathrm{ppm}). \tag{19}$$

This is to be compared with the value

$$\Delta\nu(\mathrm{M1}) = 4463.15(6)\ \mathrm{Mc/sec}\quad (13\,\mathrm{ppm}), \tag{20}$$

found by Cleland *et al.* in a least-squares fit which assumed that both the zero-pressure intercept and the fractional pressure shifts were unknowns.

This alternative value for the muonium hyperfine interval can be used together with the Ruderman correction for the measured *g* factor of muon to determine an alternative value for the fine-structure constant. The theoretical expression for the muonium hyperfine splitting is [3,22]

$$\Delta\nu(\text{M-theo}) = \frac{16\alpha^2 c R_\infty}{3}\,\frac{\mu_\mu}{\mu_p}\,\frac{\mu_p}{\mu_e}\left(1+\frac{m_e}{m_\mu}\right)^{-3} \times (1+a_e)^2(1+\epsilon_1+\epsilon_2)(1-\delta_\mu), \tag{21}$$

where $a_e = \alpha/2\pi - 0.328\,\alpha^2/\pi^2$ (22)

$$\epsilon_1 = -(1-\ln 2)\alpha^2, \tag{23}$$

$$\epsilon_2 = -(8\alpha^3/3\pi)[(\ln\alpha)(\ln\alpha - \ln 4 + \tfrac{281}{480}) - 6.9], \tag{24}$$

and $\delta_\mu = (3\alpha/\pi)(m_e/m_\mu)\ln(m_\mu/m_e).$ (25)

The currently accepted values for the constants in Eq. (21) are[23,24]

$$R_\infty = 109\,737.31(1)\ \mathrm{cm}^{-1}\quad (0.1\,\mathrm{ppm}), \tag{26}$$

$$\mu_e/\mu_p = 658.2105(2)\quad (0.3\,\mathrm{ppm}), \tag{27}$$

$$c = 2.997\,925(1)\times 10^{10}\ \mathrm{cm/sec}\quad (0.3\,\mathrm{ppm}), \tag{28}$$

$$m_\mu/m_e = 206.765(3)\quad (13\,\mathrm{ppm}), \tag{29}$$

$$\mu_\mu/\mu_p = 3.183\,38(4)\quad (13\,\mathrm{ppm}). \tag{30}$$

When the Ruderman chemical-shift correction for the ratio of the measured muon and proton magnetic moments (taken as 16 ppm) is applied to Eq. (30), the ratio becomes

$$\mu_\mu/\mu_p = (3.183\,38)(1 - 16\times 10^{-6}) = 3.183\,329(41)\quad (13\,\mathrm{ppm}). \tag{31}$$

When we substitute the values from Eqs. (26), (27) (28), (29), and (31) into Eq. (21), we obtain for the muonium hyperfine splitting the expression

$$\Delta\nu(\text{M-theo}) = (8.381523\times 10^7)\alpha^2\ \mathrm{Mc/sec}\ (13\ \mathrm{ppm}). \tag{32}$$

This equation together with Eq. (19) gives for the fine-structure constant α the value

$$\alpha^{-1}(\mathrm{MR2}) = 137.0367(10)\quad (8\,\mathrm{ppm}). \tag{33}$$

This value for α agrees very well with the value determined from the measurements of $2e/h$ by means of the ac Josephson effect. The value of α which is found using the muonium hyperfine splitting determined by Cleland *et al.* [Eq. (20)] is [25]

$$\alpha^{-1}(\mathrm{MR1}) = 137.0379(12)\quad (9\,\mathrm{ppm}). \tag{34}$$

This value lies between the Josephson-effect value and the Lamb value.

DISCUSSION

In using the value of the fractional pressure shifts measured for hydrogen and tritium to analyze the muonium data, we are making three assumptions. First, we are assuming that there is no dependence of the pressure shift on the reduced mass of the hydrogen-like atom; second, we are assuming that the pressure shift is only linear and using measurements made at pressures in the range from 0.06 to 0.3 atm to analyze measurements made in the range from 10 to 65 atm; third, we are assuming that while the muonium atoms are being observed, they are sufficiently thermalized that we can use measurements made at room temperature to analyze the muonium data. We shall consider these assumptions one by one.

The theory of the pressure shifts developed by Clark[15] shows that the only dependence of the pressure shift on the mass of the interacting atoms comes from a quantum-statistical mass-dependent correction to the classical ensemble average. The theoretical expression for the pressure shift at constant volume and temperature which includes the first-order quantum-mechanical mass-dependent correction is

$$\left.\frac{\partial}{\partial P}\frac{\Delta\nu}{\Delta\nu_0}\right|_{T\ V} = \left(\frac{1}{kT}\right)\left\{\int\left(\frac{\Delta\nu(\vec{\mathrm{R}})}{\Delta\nu_0}\right)\exp\left(-\frac{U(\vec{\mathrm{R}})}{kT}\right)d\vec{\mathrm{R}} + \frac{1}{6(kT)^2}\left(\frac{\hbar^2}{2\mu}\right)\int\left(\frac{\Delta\nu(\vec{\mathrm{R}})}{\Delta\nu_0}\right)\exp\left(-\frac{U(\vec{\mathrm{R}})}{kT}\right) \times\left[\frac{1}{2kT}\left(\nabla U(\vec{\mathrm{R}})\right)^2 - \nabla^2 U(\vec{\mathrm{R}})\right]d\vec{R}\right\}. \tag{35}$$

The $U(\vec{R})$ is the potential describing the interaction between the two atoms; $\Delta\nu(\vec{R})$ gives the hyperfine splitting of the atom being considered as a function of its distance from the perturbing atom; μ is the reduced mass of the two colliding atoms; T is the absolute temperature; and k is the Boltzmann constant. Thus, in general, we can write

$$\frac{\partial}{\partial P}\left(\frac{\Delta\nu}{\Delta\nu_0}\right)\bigg|_{T,\,V} = A + \left(\frac{m_p}{\mu}\right)B. \tag{36}$$

The m_p is the mass of the proton. From the measurements on hydrogen and tritium reported in this paper, we obtain

$$A = (-4.83 \pm 0.06)\times 10^{-9}/\text{mm Hg}, \tag{37}$$

$$B = (0.05 \pm 0.08)\times 10^{-9}/\text{mm Hg}. \tag{38}$$

Equation (36) predicts for the muonium fractional pressure shift the value

$$\frac{\partial}{\partial P}\left(\frac{\Delta\nu}{\Delta\nu_0}\right)\bigg|_{T,\,V} (\text{muonium}) = (-4.39 \pm 0.84) \times 10^{-9}/\text{mm Hg}. \tag{39}$$

The error is sufficiently large so that this value encompasses the measured value of Cleland *et al.* This analysis shows that to make a completely justified extrapolation from the hydrogen to the muonium measurements, the ratio of the hydrogen and tritium pressure shifts must be measured to a tenth of 1% or better.

In order to understand the shift in the hyperfine interaction constant for hydrogen atoms trapped in an argon matrix, Adrian[26] calculated the fractional shift in the hydrogen hyperfine splitting as a function of the separation of the argon and hydrogen atoms. This calculation shows that the fractional shift in the hyperfine interaction constant is dominantly negative and due to the van der Waals interaction. Adrian found that he could satisfactorily understand the shifts in the solid by making calculations for hydrogen plus a single argon atom and then multiplying the result by the number of nearest-neighbor argon atoms. This assumes that the hyperfine shift is linear even at the equivalent pressures found in the solid. If the solid is treated as a high-pressure gas, the equivalent gas pressure at room temperature is 890 atm. If we now use this pressure and the hydrogen pressure shift reported in this paper to predict the shift for a hydrogen atom trapped in solid argon, we find an expected fractional shift of −0.32%. This is to be compared with the measured value −0.46%, reported by Foner, Cochran, Bowers, and Jen.[27] These calculations suggest that the nonlinear terms are not important. In any event, one expects the three-body terms to be dominated by the van der Waals interaction and to give an additional negative contribution. Thus if there is a pressure-dependent term which varies as the square of the pressure, it will most probably have a negative coefficient. In the case when one uses the correct linear term to analyze data in which there is a small negative quadratic component, one will obtain a value for the zero-pressure intercept which is too small rather than too large. If one fits the same data to a linear function of pressure, one will obtain a zero-pressure intercept which is too large rather than too small.

In the theory of the formation of muonium, it is usually assumed that the muon captures an electron when the muon velocity is comparable to the orbital velocity of an electron in the hydrogen atom. This corresponds to a muon kinetic energy of 2.5keV. The newly formed muonium atom then continues through collisions with the argon atoms to slow down to thermal velocities. Calculations using the simple slowing-down formulas of Fermi[28] show that it requires about a thousand collisions to bring the newly formed muonium down to thermal velocities. The corresponding time in argon at a pressure of 1 atm is about 10^{-7} sec. Since the mean life of a muon atom is 2.2μsec and the atoms are detected only when the muon decays and emits an electron, the muonium is well thermalized during most of the time it is being observed. Thus is seems justified to use data taken at room temperature on well-thermalized hydrogen atoms to analyze muonium.

CONCLUSIONS

In this paper we have reported measurements of the fractional pressure shift in argon of the hydrogen and tritium hyperfine splitting. These measurements give a more precise value for the hydrogen pressure shift, show that there is no strong dependence of the shift upon the mass of the hydrogen-like atom, and show that there is a weak dependence at constant argon density of the pressure shift on the temperature of the gas. When we use this value for the hydrogen pressure shift to reanalyze the muonium data of Cleland *et al.*, we obtain a somewhat larger value for the muonium hyperfine

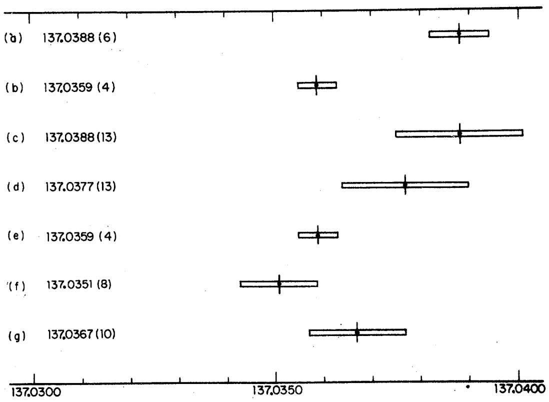

FIG. 7. A diagram showing the values of the fine-structure constant (α^{-1}) determined from the various independent sources. The sources are: (a) Ref. 1; (b) Ref. 19; (c) Ref. 3; (d) Ref. 9; (e) Ref. 10; (f) Ref. 11; (g) this paper.

splitting. The value of the fine-structure constant calculated from this alternative value for the muonium hyperfine splitting agrees very well with that found from the Josephson-effect measurements of $2e/h$. Figure 7 summarizes the values of α obtained from the different experimental sources. The trend of the data in Fig. 7 suggests that the old Lamb measurement is somewhat in error and that the correct value of α is that found using the Josephson effect. This also suggests that there is no discrepancy which is not understood in the proton structure correction to the hydrogen hyperfine splitting.

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[†] A preliminary report of this work has been given in an earlier publication: R. A. Brown, L. C. Balling, and F. M. Pipkin, Bull. Am. Phys. Soc. 11, 328 (1966). There is an error in this abstract. The sentence "For the bulb with... splitting *decreased* with an increase... at 35°C." should read "For the bulb with... splitting *increased* with an increase... at 35°C."

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