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Accurate wave-number measurements for the ^{4}He I $1s\,2p$-$1s\,3d$ transitions and comparisons of several term separations with theory

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We have measured the ^{4}He 2^3P-3^3D (5875 Å) and 2^1P-3^1D (6678 Å) lines by Doppler-free intermodulated fluorescence spectroscopy, and also the spin-forbidden $2^3P_{1,2}$-3^1D_2 (5874 Å) and 2^1P_1-3^3D_2 (6679 Å) lines as observed in Doppler-limited fluorescence spectra. The positive column of a low-pressure discharge was the source of the excited He atoms. The wave number of the tunable dye laser inducing the fluorescence was measured relative to an iodine-stabilized He-Ne laser by photographic Fabry-Perot interferometry. The results are $\sigma(2^3P_0\text{-}3^3D_1)=17\,013.762\,08(9)$ cm^{-1}, $\sigma(2^3P_1\text{-}3^3D_2)=17\,014.705\,78(9)$ cm^{-1}, $\sigma(2^3P_1\text{-}3^1D_2)=17\,018.123\,8(4)$ cm^{-1}, $\sigma(2^1P_1\text{-}3^3D_2)=14\,966.652\,04(10)$ cm^{-1}, and $\sigma(2^1P_1\text{-}3^1D_2)=14\,970.069\,84(10)$ cm^{-1}. The 2^3P-$3\,^3D$ results are compared with two other recent determinations. A value of 1325.4(2.0) MHz obtained for the 3^3D_1-3^3D_2 interval by combination of our results with the accurately known 2^3P_0-2^3P_1 interval is consistent with several previous measurements. The measurements of the spin-forbidden lines give a more accurate connection between the low triplet and singlet terms of this spectrum. We have reevaluated several term separations (directly from our measurements or by combining them with other recent results) for comparison with available calculations. The 2^3S-2^3P separation in combination with previous calculations gives an experimental value of $-0.177\,6(5)$ cm^{-1} for the Lamb-shift contribution to this separation, as compared with Ermolaev's theoretical value of -0.173 cm^{-1}. Comparison of our value for the 2^3P-2^1P intersystem separation with theory indicates that the absolute value of the Lamb shift of 2^1P is probably less than 0.01 cm^{-1}, this result being supported by a similar comparison for our 2^1P-3^1D measurement. Our value of 102 200(3) MHz for the nonrelativistic 3^3D-3^1D separation, combined with a previous result for the corresponding separation in ^{3}He, yields experimental values for the mass-polarization contributions to the 3^3D-3^1D separations in ^{4}He and ^{3}He: The values are $-243(18)$ and $-322(24)$ MHz, respectively. We give new parametric values for the other ^{4}He $1s\,3d$ interactions (three magnetic fine-structure interactions and the exchange energy) and for the singlet-triplet mixing coefficient.

I. INTRODUCTION

Most investigations of the He I energy structures carried out during the past thirty years or so have concerned level separations in the microwave and radio-frequency ranges.[1] Indeed, it seems likely that more such separations have been measured for He than for any other atom. The resulting data provide valuable tests of theoretical calculations of the relativistic and electrostatic fine structures, but the best calculations carried out thus far for most of these structures are much less accurate than the corresponding experimental determinations.

The situation with regard to basic term separations in the optical range is rather different. The electric-dipole single-photon transitions connecting the $2S$ or $2P$ terms with the $3S,P,D$ terms and some higher terms fall in a convenient range for optical interferometry, with the $2S$-$3P$, $2P$-$3,4,5S$, and $2P$-$3,4,5D$ transition arrays lying in the range 3888—7281 Å. With the exception of some measurements of several lines[2] in 1960, however, no interferometric determinations of any of these lines were published during the period from 1928[3] until the time we began the measurements reported here. Electron correlation effects are especially important in accurate calculations of these low terms. Using wave-function expansions including the interelectronic separation explicitly, Pekeris and co-workers[4] have calculated several of the lower $1sns$ and $1snp$ levels with estimated uncertainties of ± 0.001 cm^{-1} or less, exclusive of the Lamb shifts. The first experimental measurements of any lines of the above arrays with errors significantly smaller than 0.001 cm^{-1} were performed very recently; Juncar *et al.*[5] and Hlousek *et al.*[6] having measured the 2^3P_1-3^3D_2, 2^3P_2-3^3D_3 wave numbers and the 2^3P_0-3^3D_1 wave number, respectively, using Doppler-free laser spectroscopy. Giacobino and Biraben[7] made the first direct measurements of the 2^3S-$n\,^3S$ ($n=4$—6) and 2^3S-$n\,^3D_3$ ($n=3$—6) separations using Doppler-free two-photon spectroscopy, and Hlousek *et al.*[6] determined the 2^3S-$n\,^3S$ and 2^3S-$n\,^3D_3$ ($n=4,5$) separations with higher accuracy.

We report here new measurements of all previously observed lines of the $2P$-$3D$ array except for 2^3P_2-3^3D_3. Our results for the 2^1P-3^1D line and for the intercombination (spin-forbidden) lines are 20 to 100 times more accurate than the best previous measurements, and our wave number for the 2^3P_1-3^3D_2 line is an improvement over a recent Doppler-free determination.[5] We use these data directly or combined with previous experimental results to obtain term separations of the types 2^3S-2^3P, 2^3P-2^1P, 2^1P-3^1D, 2^1P-3^1P, and 3^3D-3^1D; these terms have been chosen as being of the most interest for comparison with available theoretical calculations. We discuss the $1s\,3d$ level structure in some detail and give experimental values for the exchange energy and for the mass-polarization contribution to the single-triplet separation. The experimental ^{4}He-^{3}He isotopic shift of this mass-polarization energy is consistent with calculations, as reported previously.[8]

II. EXPERIMENT

A. Doppler-free measurements

We observed Doppler-free spectra of the 2^3P-3^3D (5875 Å) and 2^1P-3^1D (6678 Å) lines of ^{4}He by intermodulated fluorescence spectroscopy.[9] This method has previously been used to measure fine and hyperfine structure and isotope shift in the 2^3P and 3^3D terms of ^{3}He and ^{4}He.[10,11] The experimental arrangement is shown in Fig. 1. Monochromatic radiation from an actively stabilized single-mode dye laser (linewidth 1 MHz) was split into two beams of equal intensity which counterpropagated through a He discharge. The two beams were chopped at approximately 2850 and 2517 Hz by a dual frequency chopping wheel. Fluorescence from the discharge was monitored through a narrow band interference filter with a silicon photodiode, and the signal at the difference frequency of 333 Hz was detected with a lock-in amplifier.

The He dc discharge was maintained in a large bore positive column tube of 1 cm diameter equipped with two anodes permitting a positive column length of 5 or 18 cm. Test spectra showed that a cleaner Doppler-free signal could be obtained in the shorter column. The discharge was operated with He pressures of 0.12 to 1.8 Torr and currents from 15 to 35 mA. A pressure of 0.3 Torr and current of 35 mA produced the best Doppler-free spectra. The high-purity He gas was circulated continuously through a liquid-nitrogen cooled charcoal trap by a mercury diffusion pump to produce a He discharge free of any observable impurity lines.

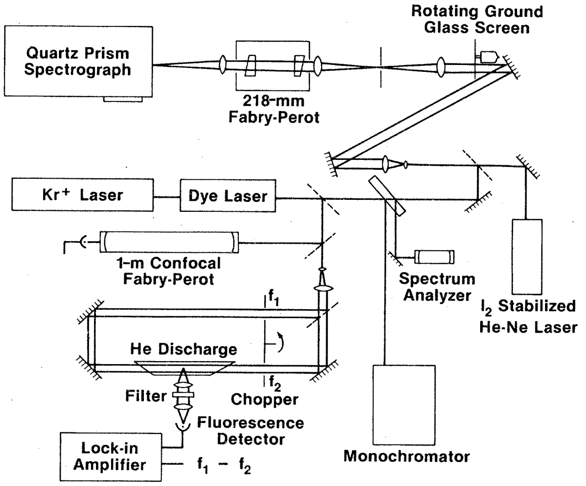

FIG. 1. Experimental arrangement for intermodulated fluorescence spectroscopy in a He discharge with measurement of the dye laser wave number by photographic Fabry-Perot interferometry. The 1-m confocal Fabry-Perot interferometer was used only to measure linewidths and check the linearity of the laser scan.

The single-mode dye laser pumped by a krypton ion laser produced an output power of about 120 mW at 5875 Å (dye Rhodamine-6G) and 90 mW at 6678 Å (dye DCM). The laser beam was expanded and collimated at a diameter of approximately 3 mm before being directed through the discharge. With this power density, strong saturation broadening was evident in the observed Doppler-free profiles. Attenuation of the laser intensity by a factor of 100 reduced the observed linewidth [full width at half maximum (FWHM)] of the 2^3P_0-3^3D_1 transition from 150 to 30 MHz. Most of our data was taken with about 2.5 mW in each of the counterpropagating beams.

The wavelength of the dye laser was determined by photographic Fabry-Perot interferometry. A detailed description of this method is given in a classic paper by Meissner.[12] Our interferometer had 10-cm quartz plates flat to approximately $\lambda/50$ separated by a 218-mm Invar spacer. The plates were coated with aluminum of relatively low reflectivity ($\sim 85\%$) providing broadband utility and high efficiency rather than high finesse. The interferometer was placed in a vacuum housing and maintained at a pressure of less than 0.01 Torr by a mechanical pump.

Illumination of the interferometer by the lasers was arranged as shown in Fig. 1. A portion of the dye laser output was combined collinearly with the output of an I_2-stabilized He-Ne laser (633 nm) which served as the wavelength standard.[13] The combined beams were expanded to a diameter of approximately 1 cm and scattered from a rotating ground glass screen which served as an extended source with sufficient angular distribution to fill the aperture of the interferometer. The interference fringes were projected onto the slit of a large quartz prism spectrograph by a 1-m achromatic lens and photographed. We measured eight fully developed rings for each pattern using a comparator with a photoelectric setting device. Computer reduction of these measurements yielded the fractional order number for the center of the fringe pattern with an uncertainty of less than 0.003 order.

The integer order of the interference (or, equivalently, the exact spacer length) was determined as follows. The approximate free spectral range was determined by physical measurement of the spacer and refined by setting the dye laser on Doppler-limited I_2 lines of successively increasing separations. Wave numbers for these I_2 lines were taken from the atlas of Gerstenkorn and Luc.[14] We then determined the exact order with the method described by Meissner[12] using the He-Ne reference laser and hyperfine components from each of the Na D_1 and D_2 lines[15] observed by Doppler-free intermodulated fluorescence. Fortunately, it proved necessary to determine the spacer length only once since the observed variation of the He-Ne laser order number was only 0.6 over a period of several months with no temperature stabilization of the interferometer.

In order to make accurate measurements by Fabry-Perot interferometry, one must take into account the wavelength-dependent phase shift on reflection from the interferometer coatings. We determined this phase shift by measuring emission lines of ^{198}Hg, Ar, and Ne using the same plates but a 5-mm spacer. With this short spacer, the phase dispersion dominates all other sources of error. Any deviation between the measured wavelengths and accurately known values[16] gives a direct measure of the phase shift. The fractional order correction due to phase dispersion was found to be 0.000 ± 0.003 throughout the region 5000—7500 Å. This result is in agreement with results of Barrell and Teasdale-Buckell[17] and of Hlousek *et al.*[6] for aluminum coatings. Our results thus include no correction for phase dispersion.

Because the He lines of interest are relatively broad (even if the instrumental effects could be kept negligible), we did not try to set the dye laser accurately to the line center. Instead, we chose to record the full line profile and determine its position by averaging the midpoints of 8 to 12 chords distributed over the upper two-thirds of the profile. The dye laser was scanned slowly enough to permit four exposures of the Fabry-Perot fringes to be made during the scan. The positions of the exposures were recorded with the line profile by using a superimposed event marker. The wave number of the line was then determined by a least-squares linear interpolation through the four measured points. A typical recording of the 2^3P_0-3^3D_1 transition is shown in Fig. 2.

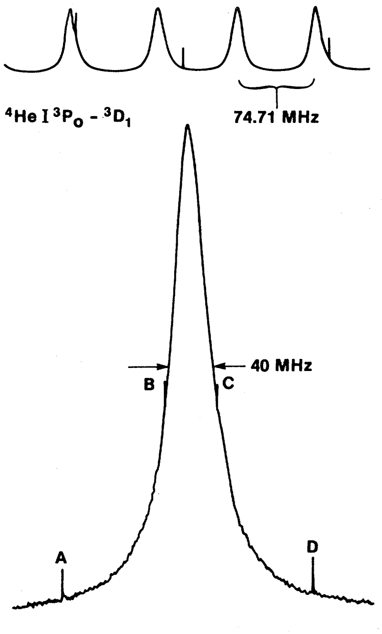

FIG. 2. Typical Doppler-free recording of the 2^3P_0-3^3D_1 transition. The observed linewidth of 40 MHz includes moderate saturation broadening. The spikes marked *A* through *D* on the trace are the points at which Fabry-Perot fringes were photographed to calibrate the dye laser wave number. The upper trace shows the interference fringes of the 1-m confocal Fabry-Perot interferometer. These were used for diagnostic purposes only and had no role in the measurement of absolute wave numbers.

Although we were able to observe all allowed transitions of the 1*s*2*p*-1*s*3*d* array, we have made Doppler-free measurements for only the 2^3P_0-3^3D_1, 2^3P_1-3^3D_2, and 2^1P_1-3^1D_2 transitions. These are the strongest transitions not perturbed by other components or crossovers. The strongest line of the multiplet, 2^3P_2-3^3D_3, was not measured because it was not fully resolved from 2^3P_2-3^3D_2 (only 76 MHz away) and the associated crossover. As will be seen, a combination of our data with previous measurements involving the 2^3P and 3^3D terms accurately determines the relative positions of all the levels.

B. Doppler-limited measurements

We were unable to observe the spin-forbidden 2^1P-3^3D and 2^3P-3^1D transitions by Doppler-free methods; however, by focusing a single laser beam into the He discharge good Doppler-limited fluorescence spectra of these lines were obtained. The transitions are mediated by singlet-triplet mixing in the 1*s*3*d* configuration, the observed lines being the single component 2^1P_1-3^3D_2 at 6679 Å and an asymmetric two-component blend $2^3P_{1,2}$-3^1D_2 at 5874 Å. The spectra were recorded as described above with four exposures of the Fabry-Perot fringes during the scan for calibration.

Because of the large Doppler width of these lines ($\sim$3.5-GHz FWHM) we could not accurately measure the line position directly from the recorded charts. Instead, each recorded spectrum was digitized from the chart by using a high-resolution digitizing table. The laser output power was also recorded and digitized. A computer was used to normalize the fluorescence signal to eliminate the influence of laser power variations and to fit model profiles to the recorded spectra with standard nonlinear least-squares techniques. Since the width of these lines was dominated by Doppler broadening, simple Gaussian profiles were assumed. We fit the 6679-Å line (2^1P_1-

TABLE I. Wave number measurements for He I 1*s*2*p*-1*s*3*d* transitions (cm^{-1}).

Transition	Present measurements	Other measurements
2^3P_0-3^3D_1	17 013.762 08(9)	17 013.762 10(3)[b]
2^3P_1-3^3D_2	17 014.705 78(9)	17 014.706 06(15)[c]
2^3P_2-3^3D_3		17 014.779 87(15)[c]
2^3P_1-3^1D_2	17 018.123 8(4)[a]	17 018.116(15)[d]
2^3P_2-3^1D_2	17 018.200 2(4)[a]	
2^1P_1-3^3D_2	14 966.652 04(10)	14 966.638(11)[d]
2^1P_1-3^1D_2	14 970.069 84(10)	14 970.069[e]

[a] The $2^3P_{1,2}$-3^1D_2 lines were observed as a single asymmetric feature because of Doppler broadening. The separation of the two unresolved components was fixed at 0.076 43 cm^{-1} (see text).

[b] Hlousek *et al.* (Ref. 6).

[c] Juncar *et al.* (Ref. 5).

[d] Herzberg (Ref. 21).

[e] Pérard (Ref. 3).

TABLE II. Some experimental and theoretical values for the $1s3d\ {}^3D_1$-${}^3D_2$ interval in ^{4}He I (MHz).

Frequency	Method
1325.4(2.0)	Present measurements combined with 2^3P_0-${}^3P_1$ interval[a]
1322(5)	Optical saturated-absorption spectroscopy[b]
1324.70(0.37)	Level-crossing spectroscopy[c]
1324.50(0.35)	Level-crossing spectroscopy[d]
1326.4(1.0)	Level-crossing spectroscopy[e]
1327.2(1.1)	Level-crossing spectroscopy[f]
1323.6(2.3)	Quantum-beat measurement[g]
1322	Theoretical[h]
1328	Theoretical[i]
1329.7	Theoretical[j]
1317.0	Theoretical[k]

[a] The 2^3P_0-${}^3P_1$ interval was taken from Kponou *et al.* (Ref. 18).

[b] Cahuzac and Damaschini (Ref. 23).

[c] Difference of 3^3D_1-${}^3D_3$ and ${}^3D_2$-${}^3D_3$ intervals obtained by Tam (Ref. 22).

[d] Perschmann *et al.* (Ref. 27).

[e] Anisimova and Semenov (Ref. 26) using Tam's experimental data.

[f] Kaul (Ref. 24).

[g] Astner *et al.* (Ref. 25).

[h] Chang and Poe (Ref. 30).

[i] Tam (Ref. 28).

[j] Cok and Lundeen (Ref. 29) using hydrogenic radial wave functions.

[k] Cok and Lundeen (Ref. 29) using the "extended adiabatic" approximation.

3^3D_2) with least-squares adjustment of the position, intensity, halfwidth, and background intensity. The asymptotic standard deviation of the position obtained from one of these fits was typically 0.000 02 cm^{-1} (0.7 MHz), which is only $\frac{1}{5}$ as large as the final uncertainty of the measurements. The standard error of the mean of 14 determinations, also 0.000 02 cm^{-1} (2×10^{-4} of the Doppler width), was comparable to the same quantity for the Doppler-free measurements. Thus, the large width of the line contributed almost negligibly to the total uncertainty and, because of other sources of uncertainty in our experiment, this Doppler-broadened line could be measured about as accurately as the Doppler-free lines.

We fit the 5874-Å line ($2^3P_{1,2}$-3^1D_2) with the sum of two Gaussian profiles whose separation was fixed at the known 2^3P_1-${}^3P_2$ interval[18] of 0.076 43 cm^{-1}. The position and intensity of the stronger component, the intensity ratio, the halfwidth, and the background intensity were adjusted to a least-squares fit. The wave number of this line was not determined as accurately as the 6679-Å line because of the strong correlation between some of the parameters in the fit. The asymptotic standard deviation for a single fitted position was typically 0.000 38 cm^{-1}, and the standard error of the mean of five determinations was 0.000 13 cm^{-1}; therefore, the statistical contribution dominates the uncertainty of this measurement. The intensity ratio of the ${}^3P_2$-${}^1D_2$ and ${}^3P_1$-${}^1D_2$ components obtained from our fitting procedure was 0.52(2) for the only discharge conditions (0.3 Torr, 35 mA) used for this line. Since the singlet-triplet mixing coefficient[19] for $1s3d$ is some 60 times greater than that for $1s2p$, the theoretical statistical intensity ratio for these components should be equal to the ratio for ${}^3P_2$-${}^3D_2$ and ${}^3P_1$-${}^3D_2$, or $\frac{1}{3}$. None of several possible explanations of the discrepancy that occur to us would be likely to affect the wave-number measurement significantly, and we have not investigated the matter further.

We tested the fitting procedures for both the 6679- and 5874-Å lines by examining computer plots of the raw and smoothed residuals between the recorded spectra and fitted profiles. The models represented the spectra with excellent fidelity, no systematic deviations being found in the residuals.

III. RESULTS AND COMPARISONS WITH THEORY

A. Measurement results

The wave numbers from our measurements are presented in Table I with the estimated uncertainties of the final digits given in parentheses. The wave number for each transition represents an average of 12 to 16 independent measurements, except that only five measurements were made for the $2^3P_{1,2}$-3^1D_2 line. All determinations are based on the "*e*" and "*g*" components of the ${}^{127}I_2$ transition 11-5, $R(127)$, used as standards. The ${}^{127}I_2$ wavelengths given in the 1982 Comité Consultatif pour la Définition du Metre (CCDM) recommendations[20] correspond to wave numbers 15 798.008 069(16) and 15 798.007 183(16) cm^{-1}, respectively, for these components. We assumed these values since the small corrections for deviations from the temperature, power, and other conditions included in the CCDM recommendations were insignificant for our measurements.

We obtained the uncertainties by adding in quadrature the possible systematic error due to dispersion of reflection phase change ($\pm$0.000 07 cm^{-1}), the uncertainty of the ${}^{127}I_2$ standards (taken as $\pm$0.000 02 cm^{-1}), and the statistical uncertainty (three times the standard error of the mean). The statistical uncertainty was less than 0.000 05 cm^{-1} for the two 2^3P-3^3D lines and less than 0.000 07 cm^{-1} for the 2^1P-3^1D and 2^1P-3^3D_2 lines. In retrospect, it would have been desirable to determine the possible correction for phase-change dispersion more accurately by using several spacer lengths.

We tested for systematic deviations of the measured wave numbers due to He pressure, discharge current, laser power, and direction of scan (increasing or decreasing wavelength). For the Doppler-limited 2^1P_1-3^3D_2 line, we checked for Doppler shifts by reversing the direction of the laser propagation along the axis of the dc discharge. No statistically significant variation was observed with respect to any of these parameters, and we believe any effects due to them are small compared to the uncertainties in Table I. Our observations regarding variations with pressure and current are consistent with the results Hlousek *et al.*[6] obtained for the 2^3P_0-3^3D_1 transition.

Our measurement of the 2^1P_1-3^3D_2 intercombination line in Table I is consistent with, but more accurate than, the preliminary value of 14 966.652 2(5) cm^{-1} reported earlier.[8] Subtraction of the new wave number for this line from the 2^1P_1-3^1D_2 wave number in Table I gives a value of 3.417 80(9) cm^{-1} for the 3^3D_2-3^1D_2 separation, with only the statistical errors contributing significantly to the uncertainty. (A statistical error of 0.000 06 cm^{-1} was obtained for each of the two lines involved, representing in each case three times the standard error of the mean of 14 or 15 measurements.) A less accurate but consistent value of 3.4180(6) cm^{-1} is given by the difference of our wave numbers for the 2^3P_1-3^1D_2 and 2^3P_1-3^3D_2 lines.

B. Comparisons with previous measurements, including the 3^3D_1-$^3D_2$ interval

The third column in Table I gives for each transition the previous measurement having the smallest estimated uncertainty.[3,5,6,21] Hlousek *et al.*[6] measured the 2^3P_0-3^3D_1 line of this array, and Juncar *et al.*[5] measured the 2^3P_1-3^3D_2 and 2^3P_2-3^3D_3 lines. Our value for the 2^3P_0-3^3D_1 wave number is seen to agree very well with the determination by Hlousek *et al.*, which is apparently the most accurate measurement made so far for any of these lines. Our value for the 2^3P_1-3^3D_2 wave number is 0.000 28 cm^{-1} less than the value measured by Juncar *et al.* The wave number for this transition can be independently determined by combining the 2^3P_0-3^3D_1 wave number of Hlousek *et al.* with the accurately known 2^3P_0-$^3P_1$ interval[18] and a value of 1324.6(0.3) MHz for the 3^3D_1-$^3D_2$ interval (discussed below); the result, 17 014.705 83(3) cm^{-1}, is 0.000 05 cm^{-1} above our value, well within the uncertainties. A similar prediction of the 2^3P_2-3^3D_3 wave number by combination of the above measurement of Hlousek *et al.* with the 2^3P_0-$^3P_2$ and 3^3D_1-$^3D_3$ separations[18,22] gives a value of 17 014.779 72(3) cm^{-1}, which is also less than the value measured by Juncar *et al.*, in this case by exactly their estimated uncertainty (0.000 15 cm^{-1}).

The difference of our wave numbers for the 2^3P_1-3^3D_2 and 2^3P_0-3^3D_1 lines is 0.943 70(6) cm^{-1}, the uncertainty being the combined statistical uncertainty of the two separate measurements (the other contributors to the uncertainties of the absolute wave numbers do not significantly affect this difference). Combining this difference with the 2^3P_0-$^3P_1$ interval[18] [29 616.864(36) MHz], we obtain a value of 0.044 21(6) cm^{-1} for the 3^3D_1-$^3D_2$ interval. This value, expressed as 1325.4(2.0) MHz, is listed with a number of other experimental[22–27] or theoretical[28–30] values in Table II. The lack of agreement (within the quoted errors) of some of the experimental values is evident. The two determinations with the smallest quoted errors (Refs. 22 and 27) agree very well, however, and the values obtained for the 3^3D_1-$^3D_3$ separation in these same two investigations were 1400.67(0.29) MHz (Ref. 22) and 1400.65(0.37) MHz (Ref. 27). We adopt averages from these results and use the values 3^3D_1-$^3D_2 = 1324.6(0.3)$ MHz and 3^3D_1-$^3D_3 = 1400.66(0.23)$ MHz wherever the experimental splittings are required in this paper.

C. 2^3S-2^3P, 2^3P-2^1P, 2^1P-3^1D, and 2^1P-3^1P separations: Comparisons with theory; Lamb shifts

1. 2^3S-2^3P separation

The experimental 2^3S-2^3P wave numbers in Table III were obtained by combining our 2^3P-3^3D measurements with the 2^3S_1-3^3D_3 separation [26 245.5719(5) cm^{-1}] observed by Giacobino and Biraben[7] and using the 2^3P and 3^3D fine-structure data as above. The 2^3S_1-2^3P_2 wave number, for example, is a combination of our 2^3P_0-3^3D_1 measurement, the 2^3S_1-3^3D_3 separation,[7] and the 2^3P_0-$^3P_2$ and 3^3D_1-$^3D_3$ fine-structure intervals.[18,22,27] The theoretical values given for the 2^3S-2^3P separations are from Accad *et al.*[4] but with the 2^3P_1 level and the 2^3P fine-

TABLE III. $1s2s\ ^3S$-$1s2p\ ^3P$ and $1s2p\ ^3P_1$-$1s2p\ ^1P_1$ separations in He I (cm^{-1}).

Separation	Experimental	Theoretical[e]	Expt. − Theor.	Lamb shift (theory)[f]
2^3S_1-2^3P_0	9231.856 5(5)[a]	9232.0342	−0.1777(5)	−0.173
2^3S_1-2^3P_1	9230.868 7(5)[b]	9231.0462	−0.1775(5)	−0.173
2^3S_1-2^3P_2	9230.792 2(5)[c]	9230.9698	−0.1776(5)	−0.173
2^3P_1-2^1P_1	2048.053 74(14)[d]	2048.011	0.043	

[a] Value obtained by combination of our 2^3P_0-3^3D_1 wave number, the 2^3S_1-3^3D_3 wave number (Ref. 7), and the 3^3D_1-$^3D_3$ interval (Refs. 22 and 27).

[b] Value obtained by combination of our 2^3P_1-3^3D_2 wave number, the 2^3S_1-3^3D_3 wave number (Ref. 7), and a value of 76.1 MHz (0.002 54 cm^{-1}) for the 3^3D_2-$^3D_1$ interval.

[c] Value obtained by combination of our 2^3P_0-3^3D_1 wave number, the 2^3S_1-3^3D_3 wave number (Ref. 7), and the 2^3P_0-$^3P_2$ (Ref. 18) and 3^3D_1-$^3D_3$ (Refs. 22 and 27) intervals.

[d] Difference of our values for the 2^3P_1-3^3D_2 and 2^1P_1-3^3D_2 wave numbers.

[e] Accad *et al.* (Ref. 4). Their results have been adjusted to a more accurate value for the Rydberg constant (see text), and the 2^3P_1 and 2^1P_1 energies include small singlet-triplet mixing contributions (Ref. 31). The Lamb shift is not included.

[f] Ermolaev (Ref. 33).

structure intervals corrected to include the effect of the singlet-triplet interaction.[31] All wave numbers given by Accad *et al.* have here been adjusted to a value of 109 722.2731 cm^{-1} for the ^{4}He Rydberg constant, based on the values[32] $R_\infty = 109\,737.315\,29(11)$ cm^{-1} and $m_e/M_\alpha = 1.370\,933\,7 \times 10^{-4}$.

Accad *et al.*[4] included relativistic and mass-polarization corrections but no QED corrections in their calculations of the $n\,^3S_1$ and $n\,^3P_1$ levels. Thus, the difference between the experimental value for the 2^3S_1-2^3P_1 separation and their calculated value should be due mainly to the Lamb shift of this transition. This difference of $-0.1775(5)$ cm^{-1} is compared in Table III with the value -0.173 cm^{-1} calculated by Ermolaev[33] as arising from a downward shift of the 2^3P_1 level by 0.038 cm^{-1} and an upward shift of the 2^3S_1 level by 0.135 cm^{-1}. In their calculations of the fine-structure displacements of the $^3P_0$ and $^3P_2$ levels from the $^3P_1$ level, Pekeris and his co-workers[4,31] included the QED corrections of order $O(mc^2\alpha^5)$. Since the sum of the remaining fine-structure corrections through $O(mc^2\alpha^6)$ is only a few MHz for either of the 2^3P intervals,[34] the deviations of the calculated 2^3S_1-2^3P_0 and 2^3S_1-2^3P_2 separations from the observed values are equal to the corresponding 2^3S_1-2^3P_1 deviation to within 0.0002 cm^{-1} (fourth column of Table III); thus, Ermolaev's result can properly be compared with all three deviations to well within the accuracy of his calculation. Although his value for the shift is within 3% of the "experimental-theoretical" difference, the discrepancy is an order of magnitude greater than the experimental uncertainty.[35]

2. $2\,^3P$-$2\,^1P$ separation and the intersystem connection

The difference between our measurement of the 2^3P_1-2^1P_1 separation and a value calculated without QED corrections is also given in Table III. Combining this difference of 0.043 cm^{-1} with Ermolaev's value of 0.038 cm^{-1} for the (downward) Lamb shift of the 2^3P_1 level, we obtain formally an upward Lamb displacement of 0.005 cm^{-1} for the 2^1P_1 level. This result should be accurate enough to establish that the absolute value of the 2^1P_1 shift is much smaller than the 2^3P_1 shift,[33] probably being of the order of 0.01 cm^{-1} or less.

TABLE IV. Theoretical ionization energies for the He I $1s2p\,^1P$ and $1s3d\,^1D$ terms, excluding Lamb shifts. Units are cm^{-1}.

	2^1P	3^1D
Nonrelativistic term	27 176.6895[a]	12 205.6680[d]
Relativistic correction	0.4676[b]	0.0322[e]
Mass polarization	−1.3852[c]	0.0072[f]
Ionization energy	27 175.7719	12 205.7074

[a] Accad *et al.* (Ref. 4). We have adjusted their result to a more accurate value for the Rydberg constant (see text).

[b] Value from Ref. 4, adjusted to include the small singlet-triplet mixing contribution (Ref. 31).

[c] Reference 4.

[d] Sims *et al.* (Ref. 39). This term value in cm^{-1} corresponds to $-2.055\,620\,74$ a.u. for the "estimated exact" nonrelativistic total energy. One unit in the last decimal place of the value in a.u. corresponds to 0.0022 cm^{-1}.

[e] Cok and Lundeen (Ref. 29).

[f] Drake (Ref. 19).

The relative positions of the ^{4}He I singlet and triplet systems adopted[36] in 1960 and also used in the 1973 compilation[37] was based primarily on an average obtained from Herzberg's measurements of two of the intercombination lines in Table I. The resulting value[37] for the 2^3P_1-2^1P_1 separation is 0.0063 cm^{-1} greater than the new value in Table III, which means that the best-determined triplet levels in the compilation[37] are systematically too low with respect to the basic 2^1P_1 level by about this amount. This correction is within the uncertainty of ±0.01 cm^{-1} estimated for the previous intersystem connection.[37,38]

3. $2\,^1P$-$3\,^1D$ separation

A relatively small Lamb shift for the 2^1P_1 level, as deduced above, is also indicated by comparison of the experimental 2^1P-3^1D wave number with a value for this separation derived from calculated energies[4,39,29,19] omitting QED contributions. The difference of the calculated 2^1P and 3^1D ionization energies in Table IV, 14 970.0645 cm^{-1}, is only 0.0053 cm^{-1} less than the measured wave number. If this difference could be regarded as significant and as due to the 2^1P_1 Lamb shift (negligible shift for 3^1D_2), the sign would correspond to a downward displacement of 2^1P_1, i.e., a shift about equal to that obtained above but in the opposite direction. The apparent disagreement does not necessarily indicate a significant 3^1D_2 shift, since the combined uncertainty of the calculated energies may be of the same order as the difference. Fischer and Cheng[40] also compared theory and experiment[37] for the 2^1P-3^1D transition by including relativistic and mass-polarization corrections; with the Lamb contribution excluded, their result is equivalent to a difference of 0.013 cm^{-1} between the experimental and calculated transition wave numbers. An accurate calculation of the Lamb shifts would be required to test the nonrelativistic term energies[4,39] to the desired accuracy (±0.001 cm^{-1} or better), and more accurate values of some of the other corrections in Table IV would probably also be necessary.

TABLE V. He I $1s2p\,^1P$-$1s3p\,^1P$ separation (cm^{-1}).

2^1P-3^1P	Source
15 074.467(2)	Combination of 2^1P-3^1D and 3^1D-3^1P wave numbers[a]
15 074.471	Adopted levels (1973)[b]
15 074.47	Theory[c]

[a] Present measurement of 2^1P-3^1D combined with 3^1D-3^1P value from Ref. 41.

[b] Reference 37.

[c] Reference 4.

TABLE VI. He I $1s3d\,^3D_2$-$^1D_2$ separation (MHz).

3^3D_2-3^1D_2	Source
102 463(3)	Present result[a]
102 499(300)	Adopted levels (1973)[b]
102 360(200)	Level anticrossing spectroscopy[c]
102 000	Theory[d]

[a] From the difference of our values for the 2^1P_1-3^1D_2 and 2^1P_1-3^3D_2 wave numbers in Table I.

[b] Reference 37.

[c] Derouard *et al.* (Ref. 42).

[d] Chang and Poe (Ref. 30).

4. $2\,^1P$-$3\,^1P$ separation

The most accurate experimental value for the 2^1P-3^1P separation should be given by combination of our 2^1P-3^1D wave number with a measurement of the 3^1D-3^1P (laser) line by Mathias *et al.*[41] [vacuum wavelength 95.788(2) μm, equivalent to 104.3972(22) cm^{-1}]. The resulting 2^1P-3^1P separation of 15 074.467(2) cm^{-1} agrees fairly well with the best previous value,[37] 15 074.471 cm^{-1}, and also agrees with the most accurate available calculated value,[4] 15 074.47 cm^{-1} (Table V). The latter agreement indicates that the absolute value of the Lamb shift for this separation is $\lesssim 0.01$ cm^{-1}, consistent with the above discussion.

D. Energy parameters for the $1s3d$ configuration

Our value for the 3^3D_2-3^1D_2 separation is given together with some previous values[37,42,30] in Table VI. The most accurate theoretical value calculated so far was obtained by Chang and Poe[30] using a perturbation method that included the second-order exchange interaction.

We base our discussion of the $1s3d$ level structure on the energy matrix for $1snd$ in the single-configuration approximation as given by Bessis *et al.*[43] This matrix includes the nonrelativistic energies $E_0(n\,^3D)$ and $E_0(n\,^1D)$ and the elements from the Breit Hamiltonian involving the magnetic-interaction radial integrals $\mathfrak{z}_0(nd)$, $\mathfrak{z}_1(1snd)$, and $\mathfrak{z}_2(1snd)$. We evaluated the parameters $E_0(3^1D)$-$E_0(3^3D)$ (denoted here as E_{st}), $\mathfrak{z}_0$, $\mathfrak{z}_1$, and $\mathfrak{z}_2$ for $1s3d$ by fitting the three independent experimental separations $^3D_2$-$^1D_2=102\,463(3)$ MHz, $^3D_1$-$^3D_2=1324.6(0.3)$ MHz, $^3D_1$-$^3D_3=1400.66(0.23)$ MHz, and adopting the experimental result $\frac{1}{2}(\mathfrak{z}_0+\mathfrak{z}_1)=650(1)$ MHz obtained for ^{3}He $1s3d$ by level anticrossing spectroscopy.[44] The resulting values of the magnetic-interaction (fine-structure) parameters and E_{st} are included in Tables VII and VIII. The errors given with the fitted parameter values reflect only the assumed experimental uncertainties; these values may also be distorted by absorption of any significant interactions not included in the matrix (including configuration interactions).

Some other values of $\mathfrak{z}_0$, $\mathfrak{z}_1$, and $\mathfrak{z}_2$ as derived from previous experimental and theoretical results are given in Table VII. Since the magnetic interactions have been expressed by different three-parameter sets in several pertinent references, we give equations connecting four such parameter sets in the Appendix. The parameter values obtained from an analysis of Tam's level-crossing data[22] by Anisimova and Semenov[26] do not agree with our values to within the uncertainties. Inspection of the pertinent integral indicates that any value of $\mathfrak{z}_2$ calculated with physically realistic wave functions will be positive[43] in contrast with the negative value obtained by the fitting procedure in Ref. 26. The theoretical value of $\mathfrak{z}_2$ calculated with the "extended adiabatic" wave functions[29] agrees best with our fitted value.

The mixing of the $^3D_2$ and $^1D_2$ wave functions induced by the off-diagonal matrix element $-(\frac{3}{2})^{1/2}(\mathfrak{z}_0+\mathfrak{z}_1)$ can be expressed by the resulting expansion coefficient[19] $\sin\theta$ or by the ratio of the expansion coefficients $\omega=\tan\theta$ (θ is the mixing angle, ω the mixing coefficient). The value of $\omega=\tan\theta$ for $1s3d$ is equal to the expansion coefficient $\sin\theta$ to four significant figures. We used our result $E_{st}=102\,200(3)$ MHz in obtaining the values of ω in Table VII. The disagreement between the values we ob-

TABLE VII. Values of the magnetic-interaction radial integrals $\mathfrak{z}_0$, $\mathfrak{z}_1$, and $\mathfrak{z}_2$ (in MHz) and the singlet-triplet mixing coefficnent ω for He I $1s3d$.

Method	$\mathfrak{z}_0$	$\mathfrak{z}_1$	$\mathfrak{z}_2$	ω
Experimental, ^{4}He and ^{3}He data[a]	868.6(2.0)	431.43(0.20)	9.4(1.8)	0.015 54(3)
Experimental, ^{4}He data[b]	852.0(1.2)	432.78(0.12)	−8.1(0.4)	0.015 36(2)
Theoretical, hydrogenic wave functions[c]	866	430	14	0.015 50
Theoretical, hydrogenic wave functions[d]	866.0	431.3	11.9	0.015 51
Theoretical, extended adiabatic wave functions[d]	861.2	428.4	9.7	0.015 42

[a] Present results, see text.

[b] Anisimova and Semenov (Ref. 26) as derived from their analysis of Tam's level-crossing data (Ref. 22).

[c] Bessis *et al.* (Ref. 43).

[d] Cok and Lundeen (Ref. 29).

TABLE VIII. Frequencies corresponding to some energy differences and the exchange parameter for ^{4}He and ^{3}He $1s3d$. The $^3D_{cg}$-$^1D_{cg}$ separation refers to the term baricenters. The nonrelativistic single-triplet separation E_{st} is evaluated parametrically by fitting the observed $1s3d$ level structure (including, for ^{3}He, the hyperfine structure). The exchange separation $\frac{2}{5}G^2$ and the mass-polarization contribution ϵ_{Mst} have been evaluated by assuming E_{st} to be equal to the sum of these quantities. The values of ϵ_{Mst} from two theoretical calculations are also listed. Units are MHz.

	^{4}He	^{3}He
$^3D_{cg}$-$^1D_{cg}$	102 233(3)[a]	102 340(5)[f]
E_{st}	102 200(3)[a]	102 116(5)[g]
ϵ_{Mst}	−243(18)[b]	−322(24)[b]
	−239[c]	−318[c]
	−284[d]	−377[d]
$\frac{2}{5}G^2$	102 443(20)[e]	102 438(20)[e]

[a] Present result.

[b] Mass-polarization contribution to the singlet-triplet separation as derived from the difference of the experimental E_{st} values for ^{4}He and ^{3}He.

[c] Value calculated by Drake (Ref. 19).

[d] Value calculated by Cok and Lundeen (Ref. 29).

[e] Exchange separation obtained from the experimental E_{st} and ϵ_{Mst} energies.

[f] From the data of Derouard *et al.* (Ref. 44).

[g] Reference 44.

tain for the fine-structure parameters and those derived in Ref. 26 leads to a corresponding disagreement between the two "experimental" values of ω. The theoretical value of ω obtained from the hydrogenic—wave-function values of $\mathfrak{z}_0$ and $\mathfrak{z}_1$ agrees best with what we believe to be the more accurate experimental value, 0.015 54(3).

The main interaction separating the ^3D and ^1D terms is, of course, the exchange energy[45] $\frac{2}{5}G^2(1s3d)$. We assume this energy to be 4.6 MHz larger for ^{4}He than for ^{3}He, due to the difference of the reduced electron mass for the two isotopes (normal mass effect). The difference between our value of E_{st} for ^{4}He and the value obtained by Derouard *et al.*[44] for ^{3}He is 84(6) MHz (Table VIII); one or more additional interactions must contribute 79.4(6.0) MHz to this difference. We assume the additional interaction to be the mass-polarization (specific mass) contribution ϵ_M, so that[8]

$$E_{st}=\tfrac{2}{5}G^2+\epsilon_{Mst}\ ,$$

with ϵ_{Mst} denoting the difference of the mass-polarization energy for the 3^1D and 3^3D terms. By differencing this equation for the two isotopes and using the inverse relation between ϵ_M and the nuclear mass M, i.e.,

$$\epsilon_M(^3\text{He})/\epsilon_M(^4\text{He})=M(^4\text{He})/M(^3\text{He})=1.3272\ ,$$

we obtain the experimental values of ϵ_{Mst} and $\frac{2}{5}G^2$ given for each isotope in Table VIII.

The values of ϵ_{Mst} from two theoretical calculations[19,29] are included in Table VIII. Correlation between the momenta of the two electrons is necessary for a nonvanishing mass-polarization energy,[46] so that the calculated ϵ_M values are a test of the accuracy of the correlation corrections included in the wave functions. The ϵ_{Mst} values from Drake's calculation[19] agree with the experimental values very well. The values calculated by Cok and Lundeen[29] are outside the experimental values by about twice the estimated experimental errors. We emphasize, however, that these errors include no allowance for effects on the experimental ϵ_{Mst} values that might arise from omission of any additional significant interactions in the parametric single-configuration treatment of $1s3d$.

Differences ΔE_{st} between the ^{4}He and ^{3}He $1snd$ triplet-singlet separations have also been determined experimentally[44,47] for higher n (up to $n=17$), although less accurately than for $n=3$. As noted previously,[8] the existing experimental ΔE_{st} values for $n=4$—7 agree to within the errors with values predicted by Cok and Lundeen's calculations of the ϵ_M energies,[29] and the calculated ϵ_{Mst} values for still higher n would contribute substantially to some of the corresponding observed ΔE_{st} values. The $1snd$ mass-polarization contributions should also be included in the analysis of accurate isotope-shift data for other separations involving $1snd$ terms (such as, for example, the 2^3S-$n\,^3D$ isotope-shift measurements by de Clercq *et al.*[48]).

IV. CONCLUSION

In this experiment we have combined the well-known methods of Doppler-free intermodulated fluorescence spectroscopy and photographic Fabry-Perot interferometry to make accurate measurements of the allowed $1s2p$-$1s3d$ transitions of ^{4}He. Our results confirm the accuracy of a recent measurement of the 2^3P_0-3^3D_1 transition[6] and provide a result for the 2^1P_1-3^1D_2 transition that is 20 times more accurate than the best previous determination. We have also used Doppler-limited fluorescence spectroscopy to measure the $1s2p$-$1s3d$ intercombination lines providing an improvement in the singlet-triplet separation by a factor of 100.

The method of photographic Fabry-Perot interferometry has been highly developed as the most accurate means of wavelength measurement for incoherent emission sources. It is worth noting that it also has significant advantages for laser sources where high accuracy is required. With an optical arrangement such as we have used, the interferometer is illuminated by an extended monochromatic source with sufficient angular distribution to fill its aperture. This eliminates the critical dependence on laser alignment and collimation which are the most serious sources of systematic error in high-precision wave meters. Although the accuracy of our measurements was limited by our spacer length and by the uncertainty in our determination of the dispersion of reflection phase change, these limitations are not fundamental. Both scale down in direct proportion to the spacer length. With the great coherence length of laser sources, much longer spacers are practical and could produce correspondingly higher accuracies.

Our comparisons of experiment and theory show that the calculated separations of a number of low He I terms are in error by less than 0.01 cm^{-1}. These results include singlet-triplet separations based on a reevaluation of the intersystem connection. In the case of the S and P terms, the comparisons apparently constitute a test of the accuracy of calculated Lamb shifts ($2^3S, 2^3P$) or give limits for their magnitude (2^1P).

The wave functions calculated for several low terms of He I are presumably the most accurate involving correlations ever obtained.[4,39] The predicted term energies cannot be tested against experiment to within the expected errors, however, because of a lack of sufficiently accurate calculations of smaller contributions (mainly Lamb shifts). The testing of Lamb-shift calculations for two-electron systems is of considerable interest in itself, of course. We have not compared the measured 2^3P-3^3D separation with theory because of a lack of any calculation of the nonrelativistic 3^3D term energy accurate enough to make the comparison interesting (errors less than 0.01 cm^{-1}). Calculations of the low $n\,^3D$ and $n\,^3F$ terms at least as accurate as the best results of Sims *et al.*[39] for the singlets are needed. The most accurate available calculations of the electrostatic and magnetic structure for $1s3d$ also fail to reproduce the experimental results to within their accuracies (Tables VI and VII). The effort required to calculate more accurately some of the contributions to term energies discussed here would appear to be justified by the importance of this spectrum for atomic theory and the improved experimental results now becoming available.

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APPENDIX

The magnetic fine-structure interactions in the Breit Hamiltonian for a two-electron atom have frequently been expressed by three parameters A, $\mathscr{A}$, and b representing the spin-orbit coupling, an off-diagonal combination of spin-orbit type interactions, and spin-spin coupling, respectively.[49,42,44,11] Cok and Lundeen[29] denote their equivalent set of parameters by $h_{\rm so}$, $h_{\rm off}$, and $h_{\rm ss}$. The values of these parameters[50] for $1snd$ in terms of the integrals $\mathfrak{z}_0$, $\mathfrak{z}_1$, and $\mathfrak{z}_2$ defined by Bessis *et al.*[43] are

$$A = h_{\rm so} = \tfrac{1}{2}(\mathfrak{z}_0 - 3\mathfrak{z}_1) - \tfrac{3}{5}\mathfrak{z}_2\ ,$$

$$\mathscr{A} = -h_{\rm off} = \tfrac{1}{2}(\mathfrak{z}_0 + \mathfrak{z}_1)\ ,$$

$$7b = 16h_{\rm ss} = 8\mathfrak{z}_1\ .$$

In this paper we have quoted all results involving these parameters in terms of the basic integrals $\mathfrak{z}_0$, $\mathfrak{z}_1$, and $\mathfrak{z}_2$. Anisimova and Semenov[26] use the equivalent set $\xi_2 = \mathfrak{z}_0$, $S_2 = \frac{1}{2}\mathfrak{z}_1$, and $S_4 = -\frac{1}{2}\mathfrak{z}_2$.

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