Precision Wavelength Measurements and New Experimental Lamb Shifts in Helium

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Absolute wave numbers have been measured to 2–3 parts in 10^9 for the $2^3S \rightarrow 4^3S$, 5^3S, 4^3D, and 5^3D two-photon transitions and the $2^3P \rightarrow 3^3D$ one-photon transition in helium. With use of relativistic calculations, Lamb shift values for these energy levels are derived and compared to theory. With improvements of one to two orders of magnitude in the theoretical calculations, these measurements could provide a new value for the Rydberg constant with an accuracy of 2 parts in 10^9.

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Precise determinations of the spectra of hydrogenic atoms and ions have, historically, provided important tests for the fundamental theories of physics. There has been renewed interest in this field in the last decade as new techniques of Doppler-free laser spectroscopy have improved the precision of absolute wavelength measurements by two orders of magnitude. This has made possible new tests of quantum electrodynamics by Lamb-shift measurements[1,2] and better determinations of the Rydberg constant.[3] To date the spectroscopic data have agreed with QED predictions, but two of the recent nonlaser tests of QED show small discrepancies outside the quoted error limits.[4] Thus further tests on new systems are desirable.

With modern computational methods it is possible to do highly accurate theoretical calculations of the spectra of two-electron systems, as well. Accad, Pekeris, and Schiff[5,6] and Blanchard and Drake[7,8] have done nonrelativistic calculations with relativistic corrections for the low-lying S, P, and D states. In some cases the quoted accuracy is as good as 1 part in 10^9, or 10^{-4} cm^{-1}. Thus in principle, high-precision tests of theory are also possible in heliumlike atoms and ions. Unfortunately the Lamb-shift (QED contribution) calculations done to data for two-electron atoms are not nearly as good.[9,10] Typical results for the $2s$ and $2p$ states have error limits of 10^{-2} cm^{-1}.

In this paper we will present absolute wavelength measurements for five one- and two-photon transitions in helium made with Doppler-free laser spectroscopy. With use of current relativistic calculations, these measurements allow a determination of the Lamb-shift differences to an accuracy of 10^{-3} cm^{-1}. With further refinements in the theoretical calculations, the accuracy of these experimental Lamb shifts could be improved to 10^{-4} cm^{-1}, and an experimental value for the Rydberg constant could be derived to better than 1 part in 10^8.

In our experiments high-resolution spectra of transitions from the $n=2$ states populated in a helium discharge were taken by Doppler-free laser spectroscopy. The cw dye laser used was both intensity and frequency stabilized (Coherent model 599-21 and 301). Absolute wavelength measurement was done by comparison to simultaneous reference interferometer scans and spectra of iodine wavelength standard lines. Our helium cell was a simple positive-column discharge tube constructed out of 1-cm-diam glass tubing and copper electrodes. A slow flow of ultrahigh-purity helium insured a clean discharge without the need for a high-vacuum system. The iodine cell was designed for stabilizing He-Ne lasers and was filled with $^{127}I_2$ by Howard Layer of the National Bureau of Standards.

The conventional saturation spectroscopy setup was used to get Doppler-free one-photon spectra in iodine and helium. For the two-photon transition in helium it was necessary to build a power enhancement cavity around the helium discharge tube. A nearly concentric Fabry-Perot design was used so as to produce a small waist radius, about 35 μm, in the center of the discharge. The cavity mirrors were standard dye-laser mirrors of 10-cm radius of curvature mounted internally on piezoelectric tubes. The reflectivities were about 98% on the input and 99.8% on the output. A commercial frequency stabilizer unit (Lansing) kept the cavity transmission locked to maximum as the dye-laser frequency was scanned. By employment of a large dither amplitude, the laser intensity in the cavity was modulated at twice the dither frequency. The fluorescence generated by two-photon absorption could then be discriminated from the discharge fluorescence with a $\frac{1}{4}$-m monochromator and a lock-in amplifier. A polarizer and Faraday rotator isolated the dye laser from the enhancement cavity.

Two confocal interferometers, 0.5 and 1.0 m long, were used for the absolute wavelength measurements. These were constructed out of quartz tubes with a design somewhat similar to that of Goldsmith *et al.*[11] The four mirrors were quartz substrates coated simultaneously with evaporated aluminum. Each mirror had a reflectivity of 90% and a transmission of 0.5%. The interferometers were mounted in a double-walled insulated box, evacuated to less than 10^{-4} Torr, and temperature stabilized to ±0.001 °C.

The free spectral ranges of these interferometers were calibrated in stepwise fashion, with successively improved accuracy, by the method of exact fractions, as outlined by Goldsmith *et al.*[11] For the initial steps, the $^{127}I_2$ absorption wave numbers of Gerstenkorn and Luc were used.[12] The final calibration was done with the well-known iodine $R(127)11$-$5i$, $P(62)17$-$1o$, and $P(13)43$-$0s$ wavelength standards at 6328, 5761, and 5145 Å, respectively.[13] In the last case the saturation spectra were taken with a piezoelectrically tuned, single-mode argon-ion laser. Since the $R(127)11$-$5i$ standard line at 6328 Å has a weak saturation spectroscopy signal, it was more convenient to use the strong adjacent line, $P(33)6$-$3r$, as our experimental wavelength standard. We measured its wave number to be 15797.967 837(26) cm^{-1}, which is in good agreement with the updated results of Goldsmith *et al.*[11]

All the spectra were recorded on a dual-pen x-y recorder (Hewlett-Packard model 7046A) driven by the scan ramp of the dye or argon laser. Usually one He or I_2 spectrum was recorded simultaneously with the transmission of one of the two reference interferometers. Typically 6–10 scans were made at each wavelength in the following order: 5145 Å, 6328 Å, the He or I_2 line of interest, 6328 Å, and 5145 Å.[14] The fractional fringe position of the I_2 or He line in each scan was then measured carefully and the results averaged for each wavelength. From the measured numbers of fringes between the 5145- and 6328-Å standard lines, the free spectral ranges of the two interferometers were calculated. The absolute wave number of the He or I_2 line of interest was then determined from the fringe number of the He or I_2 line relative to the 6328-Å line.

Our measured wave numbers for four two-photon transitions from the 2^3S_1 metastable state in helium and the 2^3P_0–3^3D_1 one-photon transition are presented in Table I along with previous measurements for these lines. Our results are a factor of 6 to 30 more precise than the previous data. Except for the 2^3S_1 to 4^3S_1 transition, both sets of values agree within the quoted error limits. The differential Lamb shifts for these transitions can be obtained by subtracting from the experimental data the non-QED contributions to the energy levels, as calculated theoretically by Accad, Pekeris, and Schiff (nS and nP states),[5,6] Blanchard and Drake (nonrelativistic $3D$ and $4D$ states),[7] and Drake (relativistic and mass-polarization corrections for the $3D$ state).[8] The results are given in the fourth column of Table I. The small relativistic and mass-polarization cor-

TABLE I. Measured helium wave numbers and Lamb shifts. The experimental Lamb shifts are obtained by subtracting relativistic calculations, including mass-polarization corrections, from our experimental wave numbers. The error limits in the experimental Lamb shifts are due mainly to the uncertainties in the relativistic calculations.

He	Measured wave numbers (cm^{-1})		Lamb shift (cm^{-1})	
Transition	This experiment	Other measurements	Experiment	Theory
$2^3S_1-4^3S_1$	30442.13891(8)	30442.1398(6)[a]	−0.122(1)	−0.113(13)[c] −0.119(?)[d]
$2^3S_1-5^3S_1$	33491.01706(6)	33491.0177(8)[a]	−0.132(10)	−0.121(13)[c] −0.127(?)[d]
$2^3S_1-4^3D_1$	31588.52639(6)	31588.5262(5)[a]	−0.48(?)	−0.129(13)[c]
$2^3S_1-5^3D_1$	34061.18709(8)	34061.1876(5)[a]	⋯	⋯
$2^3P_0-3^3D_1$	17013.76210(3)	17013.7623(10)[b]	−0.038(23)	0.040(?)[d,e]

[a] Ref. 15.
[b] Ref. 16.
[c] Ref. 9.
[d] Ref. 10.
[e] Ref. 8.

rections for the $4D$ state were obtained by n^{-3} scaling of the $3D$-state corrections.[17] Theoretical Lamb-shift calculations of Suh and Zaidi[9] and Ermolaev[10] are presented in column 5 for comparison. Unfortunately no QED calculations have been done for the D states and the higher S states. In Table I we have approximated the small $4S$ and $5S$ Lamb shifts by n^{-3} scaling of the $2S$ Lamb shift. The D-state Lamb shifts are expected to be small and have been neglected in Table I.

The agreement between theory and experiment for the S states is quite good, well within the uncertainties in current theoretical calculations. There is a large discrepancy, however, for the D states, especially the $4D$ state. This indicates that errors on the order of tenths of an inverse centimeter exist in the latest nonrelativistic calculations for these states. Our experimental results for the Lamb shifts of the S states are presently an order of magnitude more precise than the QED calculations. With improvements in the nonrelativistic calculations for the $4S$ and $5S$ states, an additional order of magnitude can be obtained. Thus there is the potential for a very good check of QED theory for a two-electron system if better QED and nonrelativistic calculations can be done.

The error limits on the measurements of Table I arise from three major sources: the error limits on the wavelength standards used, uncertainties in reading the fractional fringes, and pressure-shift uncertainties. The contribution of each error source to the total error limit is listed in Table II. A final error limit was obtained by adding the individual errors in quadrature. The errors due to fractional fringe reading were assumed to be statistical except in the case of the 5145-Å line, where scan nonlinearity had to be included. The quoted limits correspond to three standard deviations of the mean.

The pressure-shift and pressure-broadening coefficients of all five helium lines were determined by recording the observed wave number as the pressure was varied from 0.1 up to 1.5 Torr. The results are given in Table III. In all cases the wave numbers of Table I were corrected for pressure shifts, and the residual error contributions quoted in Table II are due solely to the uncertainties in the pressure shifts.

The possible existence of wavelength-dependent phase shifts in the aluminum-coated mirrors of the interferometers was checked by comparing measurements made with the 1.0- and 0.5-m interferometers for various $^{127}I_2$ lines in this spectral region. In all cases the agreement was excellent, well within the uncertainties due to fringe reading. Thus we conclude that wavelength-dependent phase shifts are not observable in our apparatus. This is consistent with previous studies.[18]

The existence of Stark shifts was tested by varying the discharge current from 10 to 45 mA. Absolutely no shift was observed for the most sensitive transition, $2s-5d$. This agrees with our estimates of the dc field in the positive column, <3 V/cm, and the local fluctuating field due to the ion density, ≤ 2.5 V/cm. For the $2s-5d$ transition a 3-V/cm field produces only a 3×10^{-6}-cm^{-1} shift,[17] which is negligible in our experiments. There are also no significant ac Stark shifts due to the laser field.

If improvements can be made of an order of magnitude in the relativistic calculations for helium and two orders of magnitude in the QED contributions, our precision absolute wavelength measurements could be used to derive a value for the Rydberg constant accurate to 2 parts in 10^9. This would be not only an important check of the Rydberg value determined with hydrogen, but also a valuable check on the theory of two-elec-

TABLE II. Sources of error in the helium wave-number measurements. All values are given in units of 10^{-5} cm^{-1}.

	2^3S_1 to				2^3P_0 to
Error source	4^3S_1	5^3S_1	4^3D_1	5^3D_1	3^3D_1
λ standards	4.4	3.0	3.6	3.2	1.7
Fringe reading	6.4	5.0	4.4	6.8	2.7
Pressure shift	0.8	0.5	0.4	2.6	0.1
Total	7.8	5.8	5.7	7.9	3.2

TABLE III. Measured pressure-shift and pressure-broadening coefficients for helium atoms in our positive-column discharge. All values are given in units of 10^{-4} cm^{-1} Torr^{-1}. The error limits quoted are the sum of the standard deviations generated by a linear least-squares fit to the data and the 0.7% uncertainty in the pressure measurement.

Transition	Shift	Broadening
2^3S-4^3S	10.5 ± 0.6	41.7 ± 1.7
2^3S-5^3S	34.0 ± 0.5	54.2 ± 1.0
$2^3S-4^3D_1$	0.38 ± 0.29	22.7 ± 0.9
$2^3S-5^3D_1$	2.4 ± 1.4	26.2 ± 0.9
$2^3P_0-3^3D_1$	0.05 ± 0.05	9.8 ± 0.4

tron atoms.

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[14]An accepted standard at 5761 Å was not available when these measurements were made. Only in the case of the $3P-3D$ transition, where we happened to have also taken scans at 5761 Å, could 5761 Å be used later in place of 5145 Å as a standard.

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Observation of Quantum Diffractive Velocity-Changing Collisions by Use of Two-Level Heavy Optical Radiators

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Coherence-preserving collisions for optical radiator-perturber scattering are studied by means of a velocity-selective photon-echo technique, using ytterbium atoms as heavy, long-lived, two-level radiators. These experiments establish the quantum diffractive nature of the radiator velocity change by demonstrating quantitative agreement with calculations. Radiator-destruction cross sections obtained with use of both Lamb dip and echo techniques agree to within 10%.

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We report the study of coherence- (superposition-state) preserving optical radiator-perturber collisions by means of a velocity-selective photon-echo technique, using ytterbium atoms as heavy, long-lived, two-level radiators. Previous work with light, short-lived radiators has shown that (i) collisional velocity changes can affect optical radiators involving dissimilar states,[1] and (ii)

