Direct Laser Measurement of the Lamb Shift of $2^{3}S - 2^{3}P$ Transitions in Helium

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For the first time, Lamb shifts of the $2^{3}S_{1}-2^{3}P_{0,1,2}$ transitions in helium have been measured directly with an LNA laser. The absolute wavelengths have been measured to a precision of 1 part in 10⁸, or 10^{-4} cm⁻¹. The Lamb shifts have been determined to 6 parts in 10⁴. Our results agree well with previous, indirect measurements, but represent a fivefold improvement in precision. The experimental results are compared with recent precision calculations. The obtained experimental Lamb shifts are 1 order of magnitude more precise than current theory.

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In order to test QED theory in two-electron systems, calculations and measurements of the Lamb shift in helium and heliumlike ions have drawn attention from both experimental and theoretical physicists over the course of the last two decades.¹⁻¹⁹ The low-lying S and P states in helium are especially interesting. Not only are their Lamb shifts larger than those of higher-lying levels, the nonrelativistic energies and relativistic corrections can be calculated to very high precision. Calculations of the Lamb shifts for these levels, however, are considerably less precise. Experimentalists may thus challenge theorists by making accurate measurements of transitions connecting these states so that precision experimental Lamb shifts can be deduced.

The optical transitions $2^{3}S_{1}-2^{3}P_{0,1,2}$ in helium have attracted considerable theoretical interest, ¹⁻¹⁵ because the QED contributions to the transition energies are larger than for any other transitions except for those connecting the ground state $1^{1}S_{0}$. The status of these transitions from both the theoretical and experimental point of view was summarized by Martin in 1987.⁸ Since then, there have been some improved nonrelativistic calculations with relativistic corrections of these transitions by Drake^{13,14} and by Baker, Hill, and Morgan,¹⁵ and also improved QED calculations (Lamb shifts) by Drake.^{13,14}

The experimental Lamb shifts \mathscr{S} of these transitions are defined in the following way:

 $S = \sigma_{\rm expt} - \sigma_n ,$

where σ_{expt} is the absolute transition energy measured by the experiment and σ_n is the theoretical calculation of the non-QED parts of the interval 2^3S_{1} - $2^3P_{0,1,2}$.

Until recently, no laser capable of operating at this transition wavelength existed. The best experimental measurements of these transitions were obtained by combining the measurements of the $2^{3}S-3^{3}D$ two-photon transitions¹⁶ and the $2^{3}P-3^{3}D$ single-photon transi-

tions,^{17,18} with a combined uncertainty of $\pm 5 \times 10^{-4}$ cm⁻¹. Now a direct measurement has become possible because of a newly developed solid-state laser^{20,21} employing as the active medium a crystal of LNA (La_{1-x}Nd_xMgAl₁₁O₁₉), which operates at the exact transition wavelength— 1.083 μ m.

In this Letter, we report the first direct precision measurement of the transitions $2^{3}S_{1}-2^{3}P_{0,1,2}$ in ⁴He. Doppler-free signals were obtained using saturated absorption spectroscopy. A magnetically shielded sealed cell containing ⁴He of high purity (99.99%) was excited with a weak rf discharge. The optical frequency of the LNA laser was dithered at 2 kHz and the laser was locked to the center of one of the lines by means of a lock-in amplifier. A portion of the laser light was sent into a traveling Michelson interferometer (wave meter), where the wavelength was compared to that of a Zeeman-stabilized He-Ne laser (633 nm).

The LNA laser is homemade, in a ring configuration. The LNA crystal is made by Airtron. It is pumped by a dye laser with rhodamine-6G dye tuned to 5830 Å. A lens of 5 cm focal length focuses the pump beam onto the LNA crystal. The absorption of the crystal is 99.8% at this wavelength. The laser cavity is comprised of the following: (1) four flat mirrors-three high reflectors and one output coupler with 90% reflection; (2) two lenses, both of 5 cm focal length; (3) one optical diode; (4) one birefringent filter; (5) one solid thin etalon of 0.2mm thickness, no coating; and (6) one solid thick etalon of 3 mm thickness and R = 45% coating on both sides. The laser is stabilized to an external super-invar confocal cavity (L=10 cm) by the reflected polarization method.²² With 1.5 W of pump power from the dye laser, the LNA laser puts out 130 mW single mode at 1.083 μ m and has a bandwidth of 200 kHz.

The Zeeman-stabilized laser is based on the design of Baer, Kowalski, and Hall.²³ It was calibrated by observing the beat note between it and a National Institute of Standards and Technology (NIST) iodine-stabilized He-Ne laser.²⁴ The beam was expanded with a telescope to a radius of ~ 2.5 mm to minimize diffraction. The correction due to diffraction of the Gaussian beams is then less than 5 parts in 10¹⁰. The wave-meter design follows Bennett and Gill.²⁵ By using a 48-MHz local oscillator to do fringe interpolation, it achieves a resolution of 3 parts in 10⁹.

Preliminary scans showed a large signal and a symmetric Doppler-free line shape. To minimize power broadening, we attenuated the laser beam with a neutral-density filter. We obtained linewidths of ~ 25 MHz, which presumably arose from collisions;²⁶ the natural linewidth is 1.6 MHz. With a 300-ms time constant on our lock-in amplifier, the signal-to-noise ratio was ~ 800 . We found it advantageous to keep the rf power as low as possible; otherwise absorption of the probe beam severely limited our signal.

We investigated the possibility of pressure shifts by using two cells of 0.27 and 0.78 Torr and maintaining the alignment of our wave meter for successive data sets. The pressure shift was found to be no greater than 2 MHz/Torr. These measurements were limited by the resolution of our wave meter. Consideration of other data²⁷ suggests that the pressure shift is actually lower by an order of magnitude, or less than 2 parts in 10^{10} in the 0.27-Torr cell used for most of our measurements.

Since our wave meter was not operated *in vacuo*, the raw data must be corrected for the dispersion of air.²⁵ For the dispersion of dry air at standard conditions, we use the work of Peck and Reeder,²⁸ which they claimed to be an improvement over the earlier work of Edlen²⁹ in the near ir. The effect of this choice on the final wavelength measurement is only 3 parts in 10^9 . This must then be corrected for temperature, pressure, and water vapor content, where we follow Edlen. The water-vapor correction is an extrapolation of data for visible light into the ir.

In practice, the wave meter was realigned prior to each run of the experiment. The LNA laser was locked to the saturated absorption line, and 100 measurements of the wavelength were taken over a period of about 10 min. The typical standard deviation of each run is 10 MHz (3 parts in 10⁸). Corrections for the dispersion of air were made, and the final vacuum wavelength was displayed on the computer screen. In all, fifty such sets of measurements were made, each with a statistical uncertainty σ/\sqrt{N} of about 4 parts in 10⁹.

The most significant source of error is imperfect alignment of the wave meter. Typically consecutive sets of 100 measurements using the same alignment would be consistent to 1-2 MHz, but as the instrument was realigned for each run the results fluctuated by considerably more than this. The ${}^{3}P_{0}$, ${}^{3}P_{1}$, and ${}^{3}P_{2}$ lines were measured 18, 12, and 20 times, respectively, yielding standard deviations of the mean of from 2.7 to 4.6 parts in 10⁹.

The Zeeman reference laser was calibrated against an NIST standard $^{127}I_2$ -stabilized He-Ne laser, accurate to 1.6 parts in 10¹⁰. Short-term fluctuations were of the order of 3 MHz, but these were averaged over the 10-min duration of each set of wavelength measurements. Five repeated averages for 5 min each over 2 days showed a standard deviation of 1.3 MHz. The mean of these averages was calculated and taken as our calibration, with a standard deviation of the mean of 0.6 MHz (1.3 parts in 10⁹).

In order to deduce experimental Lamb shifts \mathscr{S} from measurements, one must be clear about the definition of the non-QED part of the transition energy, σ_n . The total energy of an atomic state is

$$E = E_{\rm nr} + \Delta E_{\rm rel} + \Delta E_{\rm nuc} + \Delta E_{\rm QED} \, .$$

Here E_{nr} is the nonrelativistic energy, ΔE_{rel} are the relativistic corrections of $O(\alpha^2)$ and $O(\alpha^2 \mu/M)$, ΔE_{nuc} is the finite-nuclear-size correction, and ΔE_{OED} is the sum of all quantum-electrodynamic (QED) and relativistic corrections of $O(\alpha^3)$ and higher. We call the first three terms the non-QED parts. We attribute all $O(\alpha^4)$ terms to ΔE_{OED} , even though some of them arise from the Dirac equation. In a one-electron system, it is easy to separate the QED and non-QED parts, but in a twoelectron system there is no unique way to distinguish them. Here we accept Drake's classification³⁰ and include $O(a^4)$ terms as uncalculated parts of ΔE_{OED} . The $O(a^4)$ corrections, which are difficult to calculate and have not yet been done, include the mixture of $2^{3}P$ with higher levels $(n \ge 3)$ that arises from the second-order relativistic perturbation.

Very recently, high-precision calculations were carried out independently by Drake^{13,14} ($2^{3}S_{1}$ and $2^{3}P_{0,1,2}$) and Baker, Hill, and Morgan¹⁵ ($2^{3}S_{1}$ only). In addition to the nonrelativistic energy, all of their non-QED corrections of order α^{2} , μ/M , (μ/M)², and $\alpha^{2}\mu/M$ are calculated to a precision of better than 10^{-6} cm⁻¹ and agree exactly to that digit. Their results are listed in Table I. The difference between their numbers is only 3×10^{-7}

TABLE I. Calculated term values for the $2^{3}S_{1}$ and $2^{3}P_{0,1,2}$ levels by Drake. The σ_{n} are the non-QED parts, with accuracies better than 10^{-6} cm⁻¹. The Δ_{L} are the QED parts. Baker, Hill, and Morgan's value for the non-QED parts of $2^{3}S_{1}$ is -38454.8299864 cm⁻¹. All numbers are converted using the newly measured Rydberg constant: 109737.315709(18). Units are cm⁻¹.

Term	σ_n	Δ_L	
$2^{3}S_{1}$	-38454.8299867	0.1349(5)	
$2^{3}P_{0}$	-29 222.796 120	-0.0426(10)	
$2^{3}P_{1}$	-29 223.784 243	-0.0426(10)	
$2^{3}P_{2}$	-29 223.860 729	-0.0426(10)	



FIG. 1. Comparison of our results with previous measurements (Refs. 16-18) and theory (Refs. 13 and 14) for the Lamb shifts of the $2^{3}S_{1}-2^{3}P_{0,1,2}$ transitions in ⁴He. The Lamb shift is the difference between the measured interval and the non-QED parts of the calculated interval. Table II gives details.

cm⁻¹, or 9 kHz. Here we used the most recent value for the Rydberg constant³¹- 109737.315709(18) cm⁻¹.

The QED energy shifts ΔE_{QED} in the 2³S and 2³P states according to Drake's paper^{13,14} are also listed in Table I. The uncertainties are hard to estimate. The quoted uncertainties result from discrepancies with experiments, which according to Drake could be attributed to uncalculated relativistic and QED effects in terms of $O(\alpha^4)$. Because of this, the combined uncertainties are added linearly.

Our new experimental Lamb shifts of the interval $2^{3}S_{1}-2^{3}P_{0,1,2}$ in ⁴He are listed in Table II and illustrated in Fig. 1. The experimental statistical uncertainty, taken to be the sum in quadrature of the standard deviation of the mean from repeated measurements and the standard deviation of the mean of our reference calibration, is 3-5 parts in 10⁹. The estimated systematic uncertainty, due

to the extrapolation of the contribution of water vapor to the dispersion and possible long-term drifts of the Zeeman laser, is less than 6 parts in 10^9 . The quoted uncertainties in Table II are three times the statistical uncertainties. Our results agree very well with theory. The experimental value has an order of magnitude better precision.

As a check on the reliability of our results, we may compare the fine structure of the $2^{3}P$ states determined optically in our experiment to the very precise results obtained by Hughes and co-workers using the opticalmicrowave atomic-beam-magnetic-resonance technique.^{32,33} Table III shows this comparison. The numbers agree very well within the quoted uncertainties.

In conclusion, we have made the first direct precision measurement of the transitions $2^{3}S_{1}-2^{3}P_{0,1,2}$ in ⁴He and have obtained the most precise experimental Lamb shifts for these transitions. Our results agree with the most recent QED calculations. They agree with previous indirect measurements and are more precise by a factor of 5. The precision can be improved by at least an order of magnitude if an iodine-stabilized He-Ne laser and a high-quality interferometer are used. To go further, the cell would have to be replaced by a helium beam to eliminate possible shifts due to the cell. Then it would be possible to measure these transitions to a precision of 2 parts in 10¹⁰, i.e., 50 kHz, the limit of frequency measurement in this region. This will provide a greater challenge to theorists because this experimental Lamb shift will be 3 orders of magnitude more precise than current theory. Alternatively, if the QED calculations can be done to sufficient precision, these measurements could be interpreted as a new measurement of the Rydberg constant. The experimentally limited precision would be 2 parts in 10^{10} , the same as that from measurements in hydrogen.^{31,34} It would be interesting to see how well the values of the Rydberg constant measured by means of these two simplest atoms agree with each other to such high precision.

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TABLE II. Lamb shifts of the $2^{3}S_{1}-2^{3}P_{0,1,2}$ intervals. Previous results (σ_{prev}) are from Refs. 16-18. The experimental term intervals σ_{expt} are our results. The calculated on-QED intervals σ_{n} are from Table I. The experimental Lamb shifts, $\Im \equiv \sigma_{\text{expt}} - \sigma_{n}$, are compared with the calculated Lamb shifts Δ_{L} , from Table I. The bottom line shows the averaged Lamb shifts of the $2^{3}P$ states. Units are cm⁻¹.

Separation	σ _{prev}	σ _{expt}	σ_n	S	Δ_L
$2^{3}S_{1}-2^{3}P_{0}$	9231.8565(5)	9231.856 50(9)	9232.033867	-0.177 37(9)	-0.1775(15)
$2^{3}S_{1}-2^{3}P_{1}$	9230.8686(5)	9230.868 50(14)	9231.045744	-0.17724(14)	-0.1775(15)
$2^{3}S_{1}-2^{3}P_{2}$	9230.7922(5)	9230.79208(8)	9230.969 258	-0.17718(8)	-0.1775(15)
Average	•••	•••	•••	-0.177 26(10)	-0.1775(15)

TABLE III. Comparison of the fine structure obtained in this work to that obtained by precision microwave measurements by Hughes and co-workers (Refs. 32 and 33). Units are MHz.

Interval	Hughes and co-workers	This work	
$2^{3}P_{0}-2^{3}P_{2}$	31908.040(20)	31910.5(36)	
$2^{3}P_{0}-2^{3}P_{1}$	29616.844(21)	29619.5(50)	
$2^{3}P_{1}-2^{3}P_{2}$	2291.196(5)	2291.0(48)	

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 27 Reference 16 shows the pressure shift in the transition ${}^{23}P_{0}$ - ${}^{33}D_{1}$ to be ~150 kHz/Torr, and those for ${}^{23}S_{1}$ - ${}^{53}D_{1}$ and ${}^{23}S_{1}$ - ${}^{43}D_{1}$ to be ~7.2 and ~1.2 MHz/Torr, respectively. From Ref. 17, the pressure shifts increase with principal quantum number *n*, and so we estimate the shift in the transition ${}^{23}S_{1}$ - ${}^{33}D_{1}$ to be less than 0.3 MHz/Torr. The shift in the transition ${}^{23}S_{1}$ - ${}^{23}P_{0,1,2}$ of interest in this work should thus be of the order of 150 kHz/Torr.

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