Precise Experimental Test of Calculated Two-Electron Lamb Shifts in Helium

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We have measured the wave number of the ${}^{4}\text{He} 2{}^{1}S{}^{-3}P$ transition by Doppler-free spectroscopy in a metastable helium beam. The result, 19931.924794(45) cm⁻¹, represents an order of magnitude improvement over previous measurements and provides a sensitive test of two-electron QED effects. Our result shows an unexpectedly large deviation from theoretical prediction. We derive improved term values for a number of low-lying helium levels. All levels with n=2 show significant deviations from theory that are correlated with the size of calculated two-electron contributions to the Lamb shifts.

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The search for new physics beyond standard quantum electrodynamics (QED) has motivated increasingly accurate calculations and experiments in atomic and elementary particle physics. As a result, QED is the most stringently tested dynamical theory in physics. Most tests of QED involve the interaction of a single particle with the vacuum, as in the case of the one-electron Lamb shift in hydrogen. Recent advances in atomic theory, however, have made it possible to study more subtle contributions to the Lamb shift which are important when more than one electron is present in an atom.¹

The main new QED effect in helium can be described as a reduction of the hydrogenic Lamb shift due to screening of the nucleus by the second orbital electron.² In addition, there is a qualitatively new contribution to the Lamb shift ($\Delta E_{L,2}$) which becomes large when the two electrons come close together.³ Several precise experiments⁴⁻⁶ have been carried out on helium triplet states and have reported good agreement with theoretical predictions.⁷

In the singlet states of helium, two-electron QED effects are predicted to be larger than in the triplet states, yet no high-precision experiment has been reported. Enhancement of some QED effects in the singlet states can be understood in terms of the permutation symmetry of the electronic wave function: In the spatially symmetric singlet states the electrons spend more time close together, thus increasing two-electron proximity effects. In fact, some two-electron effects are entirely excluded from the triplet states by symmetry considerations alone. It is therefore particularly important to carry out precise measurements in the singlet states to fully test the theory.

In this Letter, we report a measurement of the $2^{1}S$ - $3^{1}P$ transition by Doppler-free laser techniques. The best previous measurements of this transition were made using emission sources in the late 1950s.⁸⁻¹⁰ Our result, with an uncertainty conservatively estimated to be an order of magnitude smaller than any previously reported for this line, reveals a discrepancy with theoretical calculations¹¹ that is significantly larger than the expected

theoretical uncertainty (Fig. 1). The experimental result is more than an order of magnitude more accurate than the calculation.

Most of the Lamb shift in this transition comes from the $2^{1}S$ level where the screening parameter is nearly 50% larger than in the corresponding triplet state.² In addition, the $\Delta E_{L,2}$ term is an order of magnitude larger in the singlet than in the triplet state.⁷ Its contribution of -0.011 cm⁻¹ to the $2^{1}S$ binding energy is 5 times larger than for any other excited state of helium.¹¹ Our measurement thus allows the first precise experimental test of this effect.

A schematic diagram of our apparatus is shown in Fig. 2. Light from a single-frequency cw laser intersects a beam of metastable helium atoms at a right angle. The laser frequency is scanned over the $2^{1}S-3^{1}P$ transition. Metastable atoms excited by the laser to $3^{1}P$ decay rap-



FIG. 1. Comparison of our result for the ⁴He $2^{1}S-3^{1}P$ wave number with the best previous measurements (Refs. 8–10) and the most recent theoretical results (Ref. 11). The uncertainty estimates of Martin, Series and Field, and this experiment represent at least 95%-confidence intervals. The uncertainty estimate given by Terrien is not clearly defined but appears to be less conservative. The uncertainty shown for the theory represents an estimate of possible contributions from higher-order terms neglected in the calculation (Ref. 11).

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FIG. 2. Schematic diagram of the experimental apparatus.

idly to the ground state, producing a drop in metastable flux at the end of the beam line proportional to the transition rate. During the scan we record the metastable flux and simultaneously determine the absolute wavelength of the laser using a high-precision Fabry-Pérot wave meter.¹² The resulting metastable depletion curve is fitted by a Voigt profile to determine the line center and width.

Our metastable helium source is modeled after that of Fahey, Parks, and Schearer.¹³ The beam is tightly collimated to yield a residual Doppler width (~ 10 MHz) much smaller than the natural width of the transition (92 MHz). At low laser powers (4 mW/cm²) typical linewidths of 105 MHz were observed. The metastable flux is monitored by electron ejection from a stainless-steel plate.¹⁴

The actively stabilized dye laser that probes the atomic transition has a linewidth of about 1 MHz. A computer scans the laser frequency across the transition in 3.5-MHz steps, covering a range of several atomic linewidths (about 450 MHz) over a period of about 100 s. After each step, a settling time of five lock-in time constants is allowed before the metastable flux is read and recorded by the computer. To remove line shifts due to residual electronic and laser settling times, each scan is immediately repeated in the reverse direction and the line centers determined from fits to the separate curves are averaged.

At frequency intervals of approximately 40 MHz the absolute wavelength of the laser is measured using a Fabry-Pérot wave meter developed in our laboratory.¹² Results from the wave meter were corrected for phase dispersion on reflection from its aluminized mirrors¹⁵ using data from Doppler-free measurements of a series of Te₂ lines that will be detailed in a subsequent publication. The Fabry-Pérot wave meter has been extensively tested and has reproduced the results of independent measurements in Te₂ (Ref. 16) and I₂ (Ref. 17) at the MHz level.

In order to eliminate small (\sim 3 MHz) errors due to

inaccuracy in aligning the laser normal to the atomic beam, our optical arrangement was designed to permit reversal of the direction of propagation of the laser beam with an accuracy of better than 5 arc sec. We repeated each scan with forward and reversed laser paths and averaged the line centers determined from fits to the separate scans. In tests of this averaging procedure we found that scans made with large intentional misalignments gave values in agreement with our most careful alignments.

Forty-one independent measurements based on 164 scans were made under a variety of conditions to test possible sources of systematic error. Each measurement is the average of four successive scans including all combinations of scan direction and direction of laser propagation. No systematic errors significant at the level of our quoted uncertainty were found. Experimental parameters that we investigated as possible sources of systematic error include laser power level, atomic beam flux, drift in laser power or atomic beam flux, charged impurity content of the atomic beam, slave-laser offset frequency, laser beam alignment, and electronics settling time.

Our result for the $2^{1}S-3^{1}P$ transition energy is 19931.924794(45) cm⁻¹. This is the equally weighted mean of all 41 measurements. The uncertainty quoted is equivalent to 1.4 MHz, of which 0.4 MHz is attributable to the uncertainty in the determination of the phase correction. Other quantifiable sources of systematic error such as photon recoil, second-order Doppler shift, and uncertainty in the reference laser frequency are negligible at our level of accuracy. The individual measurements scatter about the mean by less than 1.6 MHz, and over 90% of the points are within 1.0 MHz. The standard deviation of the mean is 0.6 MHz, and the standard error, assuming Gaussian statistics, is 0.1 MHz. Our final estimate of the experimental uncertainty has been increased to 1.4 MHz to allow for the possibility of sub-MHz systematic errors in the laser wavelength determination. Our result is compared with theory¹¹ and the best available previous measurements⁸⁻¹⁰ in Fig. 1.

Determination of Lamb shifts from experimental data is made possible by the tremendous progress that has been made in calculating the non-QED contributions to the helium energy levels.¹⁸ By taking the difference of accurately calculated relativistic (non-QED) term values and subtracting the result from the measured wave number, one obtains a measure of the true QED contribution to the transition energies, which can then be compared to QED calculations.

In Table I we list total transition Lamb shifts obtained from several experiments and from the most recent relativistic term values calculated by Drake.¹¹ The numerical precision of the calculations is better than the last digit shown. Neglected higher-order terms, however, can reduce the accuracy considerably. Since the overall un-

Transition	Experiment	Non-QED Theory ^a	Experimental Lamb Shift	Calculated Lamb Shift ^a	$\Delta E_{L,2}^{a}$	Deviation
2 ¹ S ₀ -3 ¹ P ₁	19931.924794(45) ^b	19932.017905	- 0.093111(45)	- 0.089981	0.010364	- 0.003130(45)
$2^{3}S_{1} - 2^{3}P_{0}$	9231.85650(9) ^c	9232.033865	- 0.17737(9)	- 0.177484	0.000288	+ 0.00011(9)
$2^{3}S_{1}-2^{3}P_{1}$	9230.86850(14) ^c	9231.045743	- 0.17724(14)	- 0.177492	0.000288	+ 0.00025(14)
$2^{3}S_{1}-2^{3}P_{2}$	9230.79208(8) ^c	9230.969258	- 0.17718(8)	- 0.177508	0.000288	+ 0.00033(8)
$2^{3}S_{1} - 3^{3}D_{3}$	26245.5719(5)d	26245.707749	- 0.13585(50)	- 0.135436	0.001149	- 0.00041(50)
$2^{3}S_{1} - 4^{3}D_{1}$	31588.52639(6) ^e	31588.661926	- 0.13554(6)	- 0.135139	0.001196	- 0.00040(6)
$2^{3}S_{1}-5^{3}D_{1}$	34061.18709(8)e	34061.322539	- 0.13545(8)	- 0.135017	0.001212	- 0.00043(8)

TABLE I. Term intervals. Units are cm^{-1} .

^aDrake (Ref. 11).

^bThis work.

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^cZhao et al. (Ref. 6).

certainty in the calculation is difficult to estimate precisely, the uncertainty listed in the deviation column is that due to the experiment only. Any remaining deviation is a measure of effects not fully accounted for in the calculation. For the transition reported in this Letter, the remaining deviation is unexpectedly large: 10 times larger than for any of the previously measured transitions and significantly larger than the estimated upper bound for neglected higher-order terms in the calculation.¹¹ The experimental uncertainty is much smaller than the observed deviation from theory (Fig. 1).

In addition to providing an independent and more accurate direct determination of the $2^{1}S-3^{1}P$ transition energy, our measurement can be combined with other recent results^{6,11} to update the experimental term values (ionization energies) of some low-lying helium levels following the procedure of Martin.¹⁹ The results are shown in Table II. Because of the improved accuracy of recent measurements, the uncertainties of all levels can now be reported with respect to a single reference level, $2^{3}S_{1}$. Our current measurement was used to reevaluate the energy of $2^{1}S_{0}$, reducing its uncertainty by a factor of nearly 20.

^dGiacobino and Biraben (Ref. 4).

^eHlousek, Lee, and Fairbank (Ref. 5).

From the deviation column of Table II it is apparent that the theoretical calculations do not adequately predict the n=2 Lamb shifts. The deviation ranges from 5.7 times the experimental uncertainty for $2^{3}P_{0}$ to 18.3 times the uncertainty for $2^{1}S_{0}$. The reported satisfactory agreement between theory and experiment for the transition measured by Zhao *et al.*⁶ was evidently due to a fortuitous cancellation of errors in the calculation of both the $2^{3}S$ and $2^{3}P$ Lamb shifts. The deviations for the $2^{3}P$ levels are seen to be 2.5-5 times larger than the deviations observed in the transitions. The analysis also shows that the poor agreement between theory and experiment seen in our measurement of the $2^{1}S$.

In general, the singlet levels show larger deviations and have larger two-electron screening parameters² and $\Delta E_{L,2}$ contributions than the corresponding triplet levels. Comparison between the deviation column and the explicit two-electron contribution to the Lamb shift

Level	Experimental Energy	Experimental Term Value ^a	Calculated Non-QED Term Value ^b	Experimental Lamb Shift	Calculated Lamb Shift ^b	Deviation	∆EL,2 ^b
2 ³ S1	159856.07767(ref)	38454.69467(6)	38454.829987	0.13532(6)	0.134877	-0.00044(6)	-0.001230
2 1So	166277.54366(17)	32033.22868(18)	32033.322463	0.09378(18)	0.090488	-0.00329(18)	-0.011019
$2^{3}P_{2}$	169086.869782(70)	29223.90256(9)	29223.860729	-0.04183(9)	-0.042631	-0.00080(9)	-0.001518
2 ³ P1	169087.946208(70)	29222.82613(9)	29222.784244	-0.04189(9)	-0.042615	-0.00072(9)	-0.001518
2 3P0	169087.934120(70)	29222.83822(9)	29222.796122	-0.04210(9)	-0.042607	-0.00051(9)	-0.001518
2 ¹ P1	171135.00000(13)	27175.77234(14)	27175.773406	0.00107(14)	-0.000088	-0.00116(14)	-0.002095
3 3D3	186101.64950(8)	12209.12284(10)	12209.122238	-0.00060(10)	-0.000559	0.00004(10) c	-0.000082
3 3D2	186101.65204(8)	12209.12030(10)	12209.119725	-0.00057(10)	-0.000559	0.00001(10) ¢	-0.000082
3 3 D1	186101.69622(8)	12209.07612(10)	12209.075524	-0 00060(10)	-0.000559	0.00004(10) c	-0.000082
3 1D2	186105.06984(11)	12205.70250(13)	12205.701902	-0.00060(13)	-0.000386	0.00021(13)	-0.000084
3 1P1	186209.46845(16)	12101.30389(17)	12101.304558	0.00067(17)	0.000507	-0.00016(17)	-0.000655
4 3D1	191444.60406(6)	6866.16828(9)	6866.168061	-0.00022(9)	-0.000262	-0.00004(9) c	-0.000035
5 3D1	193917.26476(8)	4393.50758(10)	4393.507448	-0.00013(10)	-0.000141	-0.00001(10) C	-0.000018
5 1 D2	193918.39320(10)	4392.37914(10)	4392.378964	-0.00018(10)	-0.000112	0.00007(10) ¢	-0.000019

TABLE II. QED contributions to low-lying levels of He. Units are cm⁻¹.

^aBased on an ionization energy of 198 310.772 34(6) cm⁻¹ referred to $2^{3}S_{1}$ at 159 856.077 67 cm⁻¹.

^bDrake (Ref. 11).

^cLevel used with theoretical term value (Ref. 11) in determining the ionization energy.

 $(\Delta E_{L,2})$ reveals a strong correlation. For the n=2 levels the deviation between theory and experiment ranges from 30% to 55% of $\Delta E_{L,2}$. There is no apparent correlation between the deviations and the size or sign of the total calculated Lamb shift.

In summary, we have presented the results of an accurate absolute wavelength measurement for the $2^{1}S-3^{1}P$ transition of helium which allows us to make the first precise test of some predicted two-electron QED effects. Our result shows significant disagreement with the theoretical prediction.

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FIG. 2. Schematic diagram of the experimental apparatus.