Measurement of the He Ground State Lamb Shift via the Two-Photon $1\,^1S$-$2\,^1S$ Transition

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We have extended two-photon Doppler-free spectroscopy to the vacuum ultraviolet spectral region, to accurately measure the He 1^1S-2^1S transition at 120 nm. Our result is 4 984 872 315(48) MHz. This yields a ground state Lamb shift of 41 104(48) MHz, in fair agreement with theory and other experiments. This approach has the potential for significantly better accuracy once improvements in the laser and the wavelength metrology are implemented. [S0031-9007(98)05888-8]

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Doppler-free two-photon laser spectroscopy has proved to be an invaluable tool for precision atomic measurements. It has been employed in an impressive series of two-photon measurements in hydrogen, for example, yielding the best value for the Rydberg constant [1,2]. Extending such techniques to shorter wavelengths is technically challenging, due to the difficulty in producing narrow band radiation with power sufficient to excite two-photon transitions. We report here the first Doppler-free two-photon measurement in the vacuum ultraviolet (VUV) spectral region, observing for the first time the radiative transition between the 1^1S ground state of He and the 2^1S excited metastable state.

The He ground state provides an ideal testing ground for quantum electrodynamics (QED) calculations of the Lamb shift in two-electron, three-body systems. The "Lamb shift" refers to all QED and finite mass corrections beyond those of order $(Z\alpha)^2$. For hydrogenlike systems, the Lamb shift is primarily due to the electron self-energy term, which is the result of virtual photon emission and reabsorption, and the significantly smaller vacuum polarization term, which results from the creation and annihilation of e^--e^+ pairs. Both terms scales as n^{-3}, where n is the principle quantum number, and are strongest in the ground state, making high accuracy ground state measurements the most sensitive tests of the theory. The present status in H has evolved to the point at which the experimental uncertainty is 3 parts in 10^{13} (0.8 kHz) [2–4], and the theoretical uncertainty is 40 kHz. All of the experiments that exhibited this high accuracy were done via the Doppler-free two-photon $1S$-$2S$ transition at $\lambda = 243$ nm.

QED corrections for two-electron systems involve both electron-nuclear and electron-electron effects and are significantly more complicated to calculate. Helium is, of course, the simplest two-electron system and, for the purpose of testing QED calculations, has the additional advantage that the uncertainty in the size of the ^{4}He nucleus is even less than that for the size of the proton.

Given that the 2^1P and 2^1S energies are well known, an accurate measurement of either the 1^1S-2^1P or the 1^1S-2^1S transition frequency can be used to determine the ground state energy. A recent series of laser experiments [5–7] has chosen the former route, using a one-photon transition at $\lambda = 58.4$ nm. By providing careful attention to phase distortions in pulse amplification, the Doppler shift, the ac Stark shift (created by an ionizing laser pulse), and other small effects, the experimenters were able to measure the 1^1S-2^1P transition frequency to a relative uncertainty of $\sim 10^{-8}$. This measurement improved the ground state Lamb shift determination in He by a factor of 100 over a previous value [8] obtained using a grating spectrometer. Because of difficulties in further reducing errors associated with the Doppler effect and the Stark shift, these single-photon measurements are probably at their limit of accuracy [7].

Using the He 1^1S-2^1S two-photon transition eliminates errors from the Doppler effect and provides an extremely narrow natural linewidth (8 Hz). More importantly for the present case, however, the 19 ms lifetime of the 2^1S level [9] allows for a delay in the second ionizing laser pulse, thereby eliminating it as a source of Stark shifts. The main limitation to accuracy in our present experiment is optical phase perturbation occurring in the pulse amplification and nonlinear frequency mixing of the laser. The associated uncertainty can be reduced in the future by using a longer laser pulse and by devising better techniques to measure the optical phase evolution.

Doppler-free spectroscopy of the 1^1S-2^1S transition requires a coherent, narrow band source of photons at 120 nm. These VUV photons are generated by three stages of harmonic upconversion in crystals and gases. A cw Ti:Sapphire laser is pulse amplified in a five stage injection-seeded Nd:YAG pumped dye amplifier to a pulse energy of $\sim$50 mJ. Approximately 3 mJ of fourth harmonic is generated by doubling twice in successive beta barium borate (BBO) crystals. The fundamental (ω_1) and fourth harmonic (ω_4) are focused into a Kr cell generating

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~30 nJ of seventh harmonic (ω_7) at 120 nm in a $2\omega_4$-ω_1 process. The relatively high conversion efficiency in this mixing scheme exploits near-resonant energy levels in Kr. Adding Ar to the Kr improves the phase matching and increases the efficiency of seventh harmonic generation. We use a 4:1 volume ratio Kr/Ar mixture with a total pressure of 33 kPa (250 Torr). Each frequency up-conversion step compresses the temporal profile of the pulse, due to the nonlinear dependence on the electric field. Our estimated pulse duration at 120 nm, based on the degree of saturation of various frequency conversion steps, is 3 ns.

The exit window of the Kr/Ar cell is an off-axis MgF_2 lens, tilted so that the beams exiting the gas cell strike the curved surface near normal incidence. This lens acts as a prism to separate the ω_7 beam from the ω_1 and ω_4 beams, and the unwanted beams are blocked. The ω_7 beam is focused into the interaction region by a second MgF_2 lens. This optical arrangement minimizes the number of optical elements while achieving focusing with minimal aberration and high rejection of unwanted wavelengths.

The second lens focuses the 120 nm radiation to a 40 μm focal spot in a pulsed high density supersonic expansion of He. The integrated He density in the 1 cm interaction length is $\sim 3 \times 10^{14}$ cm^{-3}. The 120 nm radiation is retroreflected, and the focal spot is refocused by a 2.54 cm focal length concave mirror in a standard Doppler-free arrangement. Good overlap of the focus and its image in the He jet is required for optimal signal strength. The He atoms in the 2^1S level are ionized by a quadrupled Nd:YAG laser pulse at 266 nm, which is delayed 30 ns in time relative to the 120 nm pulse. The He ions are collected and counted by a time-of-flight mass spectrometer.

Figure 1 shows an average of three scans over the Doppler-free He 1^1S-2^1S transition at 120 nm. At the center of the line, we count an order of 50 ions per laser pulse. The structure in the He scan is a combination of shot-to-shot variability in the pulsed laser power and pointing stability and electrical noise in the signal recording electronics. The ion signal from the time-of-flight mass spectrometer is smoothed with a 3 sec time constant and sampled at 10 Hz. Also shown in the figure are fringes from a marker etalon. At 120 nm, the fringes appear every 525 MHz. The Doppler-free feature appears on top of a much broader Doppler pedestal (FWHM at 120 nm = 6 GHz). Because the natural width of the Doppler-free transition is only 8 Hz, the observed transition width is a direct measure of the frequency spectrum in our laser pulse. The solid line is a least-squares fit of a model line shape to the data. The observed and modeled FWHM are 180 and 164 MHz, respectively. The statistical uncertainty in finding the line center is approximately 1 MHz. The Fourier transform width of a 3 ns pulse is 146 MHz. To date, this is the narrowest observed laser-induced atomic transition in the vacuum ultraviolet with $\Delta\nu_{\mathrm{observed}}/\nu_{\mathrm{laser}} = 7 \times 10^{-8}$.

The optical phase of the cw laser is perturbed during the pulse amplification process. Unless properly handled, this

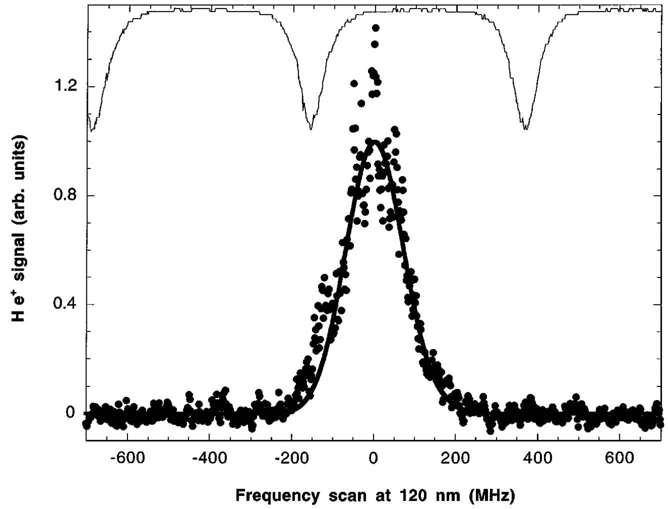

FIG. 1. Scan of Doppler-free 1^1S-2^1S He transition at $E(\omega_4) = 0.8$ mJ. The solid circles are data (FWHM 180 MHz). The heavy solid line is a model line shape (FWHM 164 MHz). The thin solid line shows fringes from a marker etalon. Each fringe corresponds to 525 MHz at 120 nm.

perturbation can limit the accuracy of pulsed-laser measurements [10]. Using an optical heterodyne technique, we measure the perturbation of the optical phase during pulse amplification, derive the instantaneous frequency, and incorporate these results into our determination of the He 1^1S-2^1S transition energy.

Our heterodyne technique is described in [11]. A portion of the cw beam before pulse amplification is shifted 400 MHz by an acousto-optic modulator. This cw beam is mixed with a portion of the fully amplified pulsed beam on a photodiode, giving a nominally 400 MHz beat note inside the pulse envelope. The beat note is Fourier analyzed to determine the instantaneous optical phase, which is differentiated to determine the instantaneous frequency. In addition to the analysis described in [11], we have carefully corrected the results for a systematic underestimate of phase perturbations, revealed by numerical tests. The optical phase perturbations are stable and reproducible from shot to shot. For all of the measurements reported here, we observe a monotonic chirp in the instantaneous frequency inside a symmetric laser pulse. The frequency at the center of the pulse is shifted typically 20 to 25 MHz higher than the cw frequency, and the slope of the chirp is typically +6 MHz/ns across the FWHM of the 842 nm laser pulse.

We now consider additional sources of optical phase perturbations in frequency doubling, four-wave mixing, and two-photon absorption in He. Second harmonic generation in BBO does not perturb the optical phase of the laser pulse when the phase matching condition is met. However, for small angular detuning, the perturbation and resulting frequency excursions can be large. Recent work [12] showed that a wave vector mismatch of only 0.2 mm^{-1} can produce frequency excursions in the 25 MHz range. At these wavelengths in BBO, in the low conversion efficiency limit, this wave vector mismatch corresponds to an angular detuning of 2 mrad. For a symmetric temporal laser profile, the resulting optical phase perturbation is bipolar (dispersion shaped). Therefore, any optical phase perturbation from the doubling and quadrupling process will most likely only broaden the frequency spread in the laser pulse without shifting either the mean frequency or the frequency at the peak of the laser pulse.

It is likely that the four-wave mixing in the Kr/Ar gas cell also introduces a frequency chirp. Our mixing scheme may be more susceptible to this kind of chirp because of the near resonance of several Kr energy levels. At present we have no way of investigating this directly. However, any resulting frequency shift should depend strongly on the peak laser intensity in the gas cell. We have measured the apparent He transition frequency as a function of laser intensity. Extrapolation to zero pulse energy will correct for the frequency chirp in the UV generation and four-wave mixing steps.

We point out that the He transition shown in Fig. 1 is reasonably symmetric and conclude that there are no pathological asymmetric shifts in frequency conversion steps between 842 and 120 nm. The observed width of 180 MHz (at 120 nm) is only slightly broader than the calculated line shape obtained from computer simulations using the results from our optical heterodyne measurements.

Using a 75 MHz Fabry-Perot cavity, we measure the relative frequency separation between the fundamental laser at the He transition and a Ne transition 8 GHz away, which serves as a transfer standard. The wave number of the Ne line is determined with respect to an iodine-stabilized 633 nm He-Ne laser in a separate measurement, using a high precision Fabry-Perot wave meter [13]. As a consistency check for the determination of the phase correction of the Fabry-Perot interferometer, we have measured two nearby Ne transitions in addition to our reference transition [14].

Our uncertainty budget for the He 1^1S-2^1S transition is presented in Table I. For consistency, all uncertainties are given in terms of their contributions to the 1^1S-2^1S transition energy. Doppler-free spectroscopy cancels the first order Doppler shift, and the second order Doppler shift is negligible. The peak intensity in the interaction region in the Doppler-free arrangement is 10^6 W/cm^2, giving a worst-case ac Stark shift of 0.7 MHz. The Ne calibration uncertainty contributes 5.6 MHz. The uncertainty in the Fabry-Perot free spectral range contributes 6.3 MHz. The possible error stemming from nonlinearity in the frequency scan is no larger than 10 MHz.

The uncertainties arising from the corrections to the optical phase perturbation during pulse amplification have a random component and a systematic component. Our estimate of the total systematic uncertainty in processing the optical phase perturbation contributes 13 MHz to the uncertainty in the He 1^1S-2^1S transition frequency.

Contributions to the random component of the uncertainty in the processing procedure include those stemming from processing parameters, the pulse effect on the instantaneous frequency, laser pulse fluctuation, line shape modeling, and statistical uncertainties in the phase perturbation algorithm. The sum of these random components contributes 29 MHz to each measurement of the two-photon transition energy.

TABLE I. Uncertainty in 1^1S-2^1S transition.

Source	Standard uncertainty (MHz)
ac Stark shift	0.7
Ne Reference transition	5.6
Free spectral range	6.3
Frequency scan	10
Optical phase perturbation	13
Power extrapolation	44
Combined standard uncertainty	48

We have measured the 1^1S-2^1S transition for several ω_4 laser pulse energies ranging from 2.0 to 0.55 mJ and find a weak dependence on pulse energy. Using the random contributions to the uncertainty as discussed in the previous paragraph, we make a linear least-squares fit of the apparent He transition frequency versus ω_4 pulse energy. The slope of the line is -85 ± 34 MHz/mJ. The uncertainty in the intercept is 26 MHz. However, it may be that the extrapolation overestimates the correction. In the absence of a viable model of the line center shift as a function of pulse energy, we have extended the uncertainty in the intercept to 44 MHz to cover the data points taken at the lowest ω_4 pulse energy, and included this a "power extrapolation" in Table I. This uncertainty, which dominates the overall uncertainty budget, reflects the unknown validity of a linear fit to the data and is much larger than the statistical uncertainty from fitting the data to a line.

Our final number for the 1^1S-2^1S transition frequency is 4 984 872 315(48) MHz. The reported uncertainty represents 1 standard deviation. We can now determine a new value for the binding energy and the Lamb shift in the 1^1S level. The binding energy of the 2^1S level has been previously determined to be $-960\,332\,041.01(15)$ MHz relative to He^+ [15,16]. From our measurement, we derive a new binding energy of the 1^1S to be $-5\,945\,204\,356(48)$ MHz. We can compare this ground state binding energy with a recent experimental determination of $-5\,945\,204\,236(45)$ MHz by Eikema *et al.* [7], who measured the 1^1S-2^1P transition frequency. These two results differ by 1.8 times the combined standard deviations.

The Lamb shift in the ground state is determined by comparing the calculated non-QED 1^1S energy level with our experimentally determined value. The non-QED calculation of the 1^1S binding energy from Drake [17] is $-5\,945\,245\,460$ MHz. The difference between this value and our measured binding energy is the Lamb shift: 41 104(48) MHz. This can be compared with 41 224(45) MHz from Ref. [7], which again differs from our result by 1.8 times the combined standard deviations. A full QED calculation of the Lamb shift as reported in [15,17] is 41 233(35) MHz, where the error bar reflects the estimated uncertainty due to α^5 terms in determining the α^4 terms. However, uncalculated relativistic corrections of order α^4 may contribute at the ± 300 MHz level. Further calculations are in progress [18].

Doppler-free two-photon spectroscopy in the VUV is an important advancement towards significantly improving high resolution VUV measurements. Although these measurements are presently at the same level of accuracy as the one-photon measurements of [7], the accuracy of the Doppler-free two-photon measurements can be greatly improved by better laser metrology. The first determination of the Lamb shift in the ground state of H using Doppler-free two-photon measurements of the $1S$-$2S$ transition at 243 nm had an uncertainty of 800 MHz [19]. However, advances in laser technology and experimental techniques have reduced the uncertainties to 800 Hz [2]. This has driven a corresponding improvement in the QED calculations of the H level energies, which are now accurate to 12 significant digits. It is likely that a similar improvement in theory will eventually be seen in He. In the near term, using a long-pulse Nd:YAG pump laser and an appropriate choice of dye concentration in the pulsed dye amplifier, we can reduce our uncertainty stemming from the optical phase perturbation from 35 to 7 MHz. Furthermore, straightforward techniques such as offset locking the cw laser to the Ne reference transition should greatly improve the repeatability and stability of these measurements. In the long term, development of a cw laser source at 120 nm [20] would dramatically improve the experimental accuracy.

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