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Precision measurement of the $2\,^3S \rightarrow 3\,^3P$ transition in ^{4}He

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We report on a precision measurement of the $2\,^3S \rightarrow 3\,^3P$ transition in ^{4}He. Doppler-free spectra of the transition were observed by exciting metastable helium atoms, produced by a chopped rf discharge, using 389-nm radiation generated by a frequency-doubled Ti-doped sapphire laser. A frequency measurement of the Doppler-free lines was made by interferometric comparison of the Ti-doped sapphire frequency with that of an iodine-stabilized He-Ne laser using a 1-m evacuated Fabry-Pérot etalon. The measurement uncertainty was 2 parts in 10^9 (3 standard deviations), which gives an improvement of a factor of 60 in the term value of the $3\,^3P$ level with respect to earlier measurements. This result makes it possible to determine the one electron Lamb-shift correction to the $3\,^3P$ state with an accuracy of 0.8%.

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Since the first measurement of the Lamb shift in the $n=2$ level in hydrogen in 1947 [1], considerable effort has been devoted to testing the predictions of quantum electrodynamics (QED). While this theory applied to all atomic systems only the more simple ones are of interest for precision tests because of the difficulties involved in calculating the QED effects as well as the non-QED terms that typically have to be subtracted from experimentally measured quantities. Single leptons and hydrogenlike systems have been of particular interest [2] but heliumlike systems have also provided valuable tests [3]. This is partly due to improved laser spectroscopic techniques and partly due to the development of very accurate variational techniques for obtaining nonrelativistic wave functions. Thus Drake has recently calculated non-QED energies for several S, P, and D states in helium with an accuracy of better than 10 kHz [4].

The experimental work on helium has concentrated on the lower triplet levels which have rather long excited-state lifetimes and where the QED effects are largest. Precision measurements of the two-photon transition frequencies between the metastable $2\,^3S$ and $n\,^3S$ ($n=4$–6) as well as $n\,^3D$ ($n=3$–6) levels have been made using dye lasers [5,6]. The $2\,^3S \rightarrow 2\,^3P$ transition at 1083 nm has been measured with a $\mathrm{La}_{1-x}\mathrm{Nd}_x\mathrm{MgAl}_{11}\mathrm{O}_{19}$ (LNA) laser using saturation spectroscopy [7]. An absolute measurement has also been reported of the $3\,^3P \rightarrow 3\,^3D$ transition frequency using an infrared diode laser [8]. More recently, the separation between the singlet levels $2\,^1S$ and $3\,^1P$ has been determined [9] and accurate measurements of two-photon $2\,^1S \rightarrow n\,^1D$ ($n=3$–7) transition frequencies have been made using a dye laser [10]. A summary of the work in this field together with a comprehensive listing of experimental and calculated term values as of 1987 can be found in Ref. [11]. This listing is partially updated in Ref. [9].

During the last few years the single-frequency Ti-doped sapphire laser has been developed into a useful tool for high-resolution laser spectroscopy [12]. Its wide tuning range in the near-infrared region and the capability for efficient second-harmonic generation of frequencies not easily accessible with dye lasers [13] has opened up the possibility of exciting more levels directly from the metastable 2S levels. Two such examples are the $2\,^3S \rightarrow 3\,^3S$ two-photon transition at 855 nm and the $2\,^3S \rightarrow 3\,^3P$ transition at 389 nm. In this paper we report on a precision measurement of the $2\,^3S \rightarrow 3\,^3P$ transition using a frequency-doubled Ti-doped sapphire laser. The laser was locked to the center of one of the Doppler-free components of the transition and its frequency determined interferometrically relative to an iodine-stabilized He-Ne laser. The 3 standard-deviation uncertainty of our measurement is 1.5 MHz which is 60 times more accurate than the best previous determination of the $3\,^3P$ energy based on a measurement of the $3\,^3P \rightarrow 3\,^3D$ separation [8]. Since the fine structure of the $3\,^3P$ term has been determined accurately in a microwave experiment [14] it was sufficient to measure only one of the components of the $2\,^3S \rightarrow 3\,^3P$ transition.

The experimental arrangement used to obtain Dopper-free spectra of the $2\,^3S \rightarrow 3\,^3P$ transition is shown schematically in Fig. 1. The frequency-doubled Ti-doped sapphire system has been described in detail in previous work [13]. The Ti-doped sapphire laser was pumped by 6-W all blue-green lines from an argon-ion laser and produced a single-frequency output of typically 600 mW at 780 nm. The laser was locked to an Invar reference cavity and had a residual frequency jitter of < 50 kHz rms. The Ti-doped sapphire laser was frequency doubled using lithium triborate (LBO) in an external enhancement cavity. An acousto-optic (AO) modulator was used to prevent optical feedback from the enhancement cavity into the laser. The light incident on the enhancement cavity was the first order of the AO modulator. The diffraction efficiency to first order was 80%. The zero-order light was used for the preliminary wave-meter measurements and the inter-

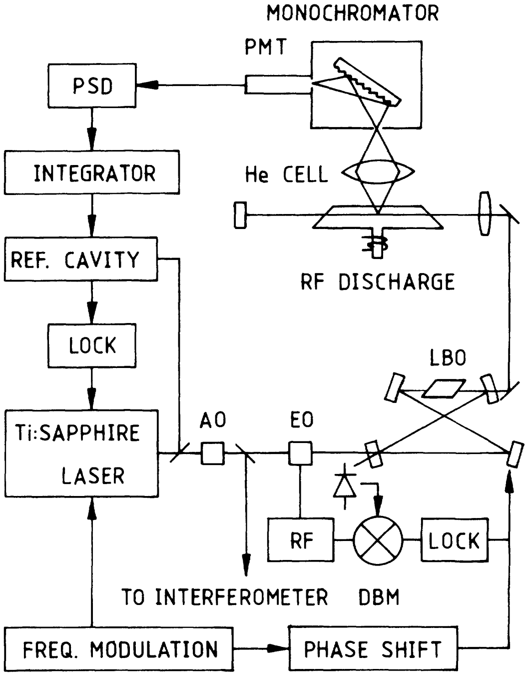

FIG. 1. Schematic diagram showing the experimental arrangement used to observe Doppler-free spectra of the $2\,^3S \rightarrow 3\,^3P$ transition in ^{4}He. The following abbreviations are used: phase-sensitive detector (PSD), photomultiplier tube (PMT), and double balanced mixer (DBM).

ferometric comparison with an iodine-stabilized He-Ne laser. The first-order light passed through an electro-optic (EO) modulator. The EO generated sidebands on the light allowing the enhancement cavity to be locked to the laser using the Pound-Drever technique [15]. With the cavity locked to the laser, the second-harmonic output at 389 nm was typically 5–10 mW. The uv radiation was weakly focused into a cell where ^{4}He atoms were excited into the metastable $2\,^3S$ level by a pulsed rf discharge. The discharge was operated with a continuous flow of helium. The fluorescence at 389 nm was monitored using a spectrometer and photomultiplier tube (PMT). The signals were detected in the afterglow to avoid perturbing effects of the discharge such as ac Stark shifts and to avoid a large background of discharge light. For the pressures and cell used in the experiment the metastable ^{4}He atoms have a lifetime of order 10 μs. Thus optimum signals were obtained by chopping the discharge at 20 kHz and observing the fluorescence during a 20-μs gate time delayed by 1 μs after the discharge was turned off. Metasable populations sufficient to observe good Doppler-free signals were obtained at helium pressures from 0.1 to 0.4 Torr.

Doppler-free spectra were observed using a saturated spectroscopy technique. The uv light was frequency modulated and retroreflected through the cell. The component of the chopped fluorescence signal at the modulation frequency was measured using a phase-sensitive detector (PSD). The frequency modulation was produced by a sinusoidal voltage applied to a piezo mounted mirror in the Ti-doped sapphire laser which was also used for fast correction in the laser servo loop. The modulation frequency was 50 kHz which was well above the unity gain frequency of the servo. The modulation amplitude was 1 MHz which was sufficiently large to observe Doppler-free lines with an acceptable signal to noise ratio but not so large that it deteriorated the fringe definition of the 1-m interferometer used for absolute wavelength comparison. The corresponding frequency modulation of the uv light was ±2 MHz. The enhancement cavity had a linewidth of 4–5 MHz. Therefore the frequency modulation generated a significant amplitude modulation on the uv output. The amplitude modulation was canceled by applying the same modulation with appropriate amplitude and phase to a similar piezo mounted mirror in the enhancement cavity.

A typical Doppler-free spectrum is shown in Fig. 2. The three components correspond to the $2\,^3S_1 \rightarrow 3\,^3P_{2,1}$ transitions and their crossover resonance. The splitting between the $J=1$ and $J=2$ lines is 658.5 MHz [14]. The $^3P_0 \rightarrow {}^3P_1$ interval is 8.1 GHz and therefore the $2\,^3S_1 \rightarrow 3\,^3P_{2,1}$ transition does not appear in Fig. 2. The observed linewidth [full width at half maximum (FWHM)] of the Doppler-free signals was 20 MHz. The Doppler broadened linewidth (FWHM) was measured to be 5.5 GHz. Thus the non-Doppler-free component within the observed signal was negligible.

The Ti-doped sapphire laser was locked to one of the Doppler-free signals. An absolute wavelength measurement was then made by interferometric comparison of the Ti-doped sapphire wavelength with the wavelength of an iodine-stabilized He-Ne laser at 633 nm using an evacuated 1-m plane-plane Fabry-Pérot cavity. A detailed account of the operation of the reference interferometer has been given elsewhere [16]. The precision associated with the interferometer measurement is dominated by the uncertainty in the definition of the 633 nm standard. This is typically an order of magnitude better than that achieved using a wave meter. The measurement procedure was as follows. The interferometer length was locked to a

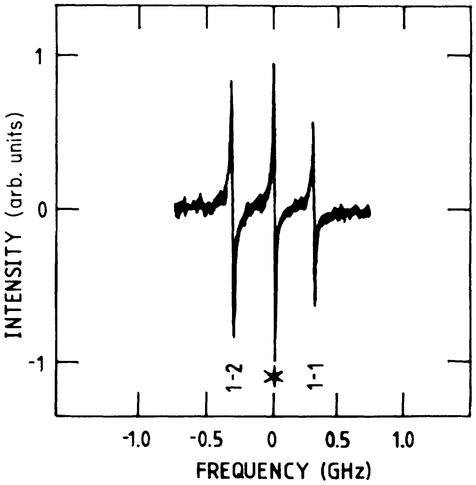

FIG. 2. Doppler-free spectra showing the $2\,^3S_1 \rightarrow 3\,^3P_1$ (1-1) and $2\,^3S_1 \rightarrow 3\,^3P_2$ (1-2) transitions and the associated crossover resonance.

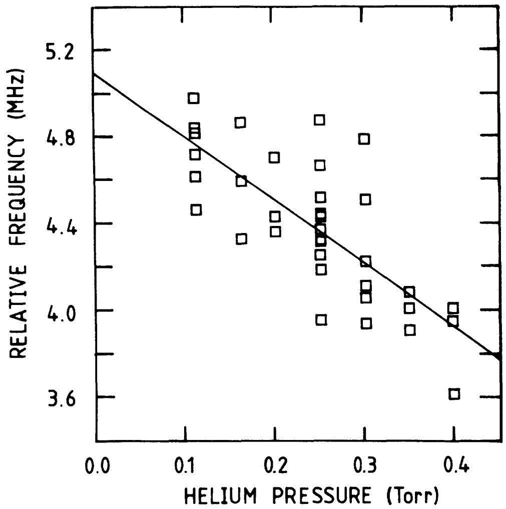

FIG. 3. Frequency measurement of the $2\,^3S \rightarrow 3\,^3P$ transition as a function of helium pressure.

TABLE I. Measurement uncertainties at 780 nm.

Source of error	Standard deviation (parts in 10^{11})
Phase-shift determination	5
Flatness and illumination uniformity	3
Prismatic imbalance (image shear)	6
Servo errors	2
Diffraction	2
633-nm reproducibility	2
633 nm relative to definition	34
Statistical reproducibility of results	29
AO frequency	5
Zero-pressure determination (±20 mTorr)	15
Doppler-free offset	45
Total (root sum of squares)	66 (0.25 MHz)

transmission maximum for the frequency of the Ti-doped sapphire laser. A He-Ne laser was tuned and also locked to a transmission peak of the interferometer. The beat frequency between the tunable He-Ne laser and a reference iodine-stabilized He-Ne laser was measured. The beat frequency measurements combined with the integer interferometer order numbers for the Ti-doped sapphire and He-Ne lasers gave an absolute value for the Ti-doped sapphire frequency. The free spectral range of the interferometer was 150 MHz allowing the order number of the Ti-doped sapphire laser to be deduced from the term values in Ref. [11] which are accurate to 30 MHz.

A total of 35 independent frequency measurements were made. Each measurement being an average of 200 beat frequency readings recorded at 1 s intervals. The effect of changing experimental conditions on the frequency measurement was investigated. A 50% reduction in the uv intensity produced no observable frequency shift. However, a small pressure shift was observed. The results of 35 observations plotted as a function of helium pressure are shown in Fig. 3. The straight-line fit indicated a pressure shift of -5.8 ± 1.0 MHz/Torr. The zero-pressure intercept was taken as the absolute frequency of the transition. The statistical repeatability of the result determined from the uncertainty in the zero-pressure intercept was 110 kHz. A complete listing of the measurement uncertainties is provided in Table I. Rows 1-5 refer to the operation of the 1-m interferometer. The phase shift at 780 nm was determined by extrapolation from previous measurements between 486 and 670 nm [17]. Row 7 is the uncertainty associated with the international specification of the 633 nm standard [18]. The last four lines refer to the Ti-doped sapphire laser reproducibility and measurement uncertainties associated with the zero-pressure determination and shifts of the measured frequency due to a residual dc background in the Doppler-free signal. The dc background arose primarily from imperfect cancellation of the amplitude modulation on the uv light. Although any systematic shift was randomized by periodic readjustment of the enhancement cavity feed forward modulation and recentering of the Doppler-free signal a possible frequency error equivalent to ±2% of the full signal (170 kHz) has been retained in the error budget. The total uncertainty (root sum of squares) was 0.25 MHz at the infrared wavelength. Thus the uncertainty in the $2\,^3S \rightarrow 3\,^3P$ measurement was 1.5 MHz (3 standard deviations).

We measured the frequency of the crossover resonance between the $2\,^3S_1 \rightarrow 3\,^3P_2$ and $2\,^3S_1 \rightarrow 3\,^3P_1$ transitions at 770724396.8(1.5) MHz. The fine-structure interval between the $J=1$ and 2 levels is 658.54 MHz [14], therefore the frequencies of the $2\,^3S_1 \rightarrow 3\,^2P_2$ and $2\,^3S_1 \rightarrow 3\,^3P_1$ transitions are 770724067.5(1.5) MHz [25708.58763(5) cm^{-1}] and 770724726.1(1.5) MHz [25708.60959(5) cm^{-1}], respectively. By comparing our measurement with the recent calculations of non-QED terms and the two-electron Lamb shift ($\Delta E_{L,2}$) by Drake [19], we are able to give an experimental value for the one-electron Lamb shift $\Delta E_{L,1}$ of $-346.5(2.8)$ MHz. The uncertainty of 2.8 MHz is dominated by an experiment correction of 12.7(2.4) MHz to the $2\,^3S$ energy derived from Ref. [6]. The one-electron Lamb shift calculated by Drake [19] is $-346.3(13.9)$ MHz.

In summary, we have performed Doppler-free saturated spectroscopy of the $2\,^3S \rightarrow 3\,^3P$ transition in ^{4}He using a frequency-doubled Ti-doped sapphire laser. By locking the laser to the helium signal and using interferometric comparison of the Ti-doped sapphire frequency with an iodine-stabilized He-Ne laser we have made a high-precision measurement of the $2\,^3S$ to $3\,^3P$ interval. The measurement uncertainty (3 standard deviations) was 2 parts in 10^9 (1.5 MHz) which provides an improvement by a factor of 60 in the term value of the $3\,^3P$ level with respect to previous measurements. By comparing our result with recent theoretical calculations we are able to determine the one-electron Lamb shift correction to the $3\,^3P$ term with an accuracy (3 standard deviations) of 0.8%.

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