Measurement of the Helium $2\,^3P_0$-$2\,^3P_1$ Fine Structure Interval

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(Received 7 July 1998; revised manuscript received 6 October 1998)

We report the most accurate measurement of the helium fine structure splitting. The $2\,^3P_0$-$2\,^3P_1$ energy splitting is 29 616 949.7 ± 2.0 kHz. Laser saturation spectroscopy, heterodyne pure frequency determination, and fluorescence detection were combined in a novel experimental approach. The absence of external perturbing magnetic fields, used in earlier experiments, lends confidence to our determined value and allows us to discriminate between contradictory results previously reported. This result, when combined with expected advances in theory, should yield a new value of the fine structure α, which may help clarify a presently puzzling experimental situation. [S0031-9007(99)08416-1]

PACS numbers: 32.10.Fn, 06.20.Jr, 42.62.Fi

Spectroscopy of simple systems is a powerful tool for fundamental studies, since the capability of high accuracy peculiar to frequency measurement combines with comparably precise theoretical predictions. Comparison with experimental results allows stringent tests for the theories describing energy spectra: for simple atoms, since the discovery of the Lamb shift, such theory has been QED. Nowadays, QED is very well established, therefore experiments are generally regarded as measurements of the physical constants entering the theoretical calculations as parameters (e.g., Rydberg constant R_∞, fine structure constant α, the proton charge radius, etc.). Obviously, this procedure is meaningful only provided that the experiments are paralleled by calculations of comparable precision.

The hydrogen atom has played a major role, as demonstrated by the work done in Paris and in Garching [1,2], which improved the accuracy of the Rydberg constant to better than one part in 10^{11}. Besides hydrogen, in the last two decades helium has been gaining the status of a "simple system," since an extension of the Dirac theory has been employed to deal with its spectrum. Helium, for instance, provides a unique playground to test the calculation of electron-electron Lamb shift, which is, of course, absent in hydrogen. Necessarily approximate, yet very precise, calculations have been performed mainly by means of variational techniques [3]. These have proved to be in good agreement with different experiments, investigating the metastable $2S$ states [4–8] and the ground $1S$ state [9,10].

With a view for measuring the fine structure constant α, helium was early recognized to be a better candidate than hydrogen, due to its larger fine structure energy splitting and longer lifetimes. Indeed, for the $2P$ state, the ratio of fine structure interval to natural linewidth is nearly 2 orders of magnitude higher in helium than in hydrogen.

This work provides a new measurement of the $2\,^3P_0$-$2\,^3P_1$ fine structure interval, ν_{01}, obtained as the difference between the $2\,^3S_1 \rightarrow 2\,^3P_0$ and $2\,^3S_1 \rightarrow 2\,^3P_1$ transitions, both measured with respect to a common reference (see the level scheme in Fig. 1). From our measurement, a value for α can be obtained with an accuracy comparable to other existing determinations. This "direct atomic physics" determination of α will be valuable, since, as for any fundamental physical constant, the consistency between measurements obtained with very different techniques supports the reliability of the value.

The fine structure of the $2\,^3P$ level has been measured at Yale by the group of Hughes [11] and, more recently, by Shiner *et al.* [7]. Two different approaches were used: direct measurement in the microwave domain and "subtraction" of optical transition wavelengths, respectively. For the $2\,^3P_0$-$2\,^3P_1$ interval, ν_{01}, reported uncertainties are 0.7 ppm [11] and 0.1 ppm [7]. However, the significant disagreement ($>5\sigma$) makes a new measurement highly desirable for the determination of α. Since the required accuracy, a few kHz, is about one thousandth of the $2\,^3P$ level natural width, systematic effects need to be carefully checked.

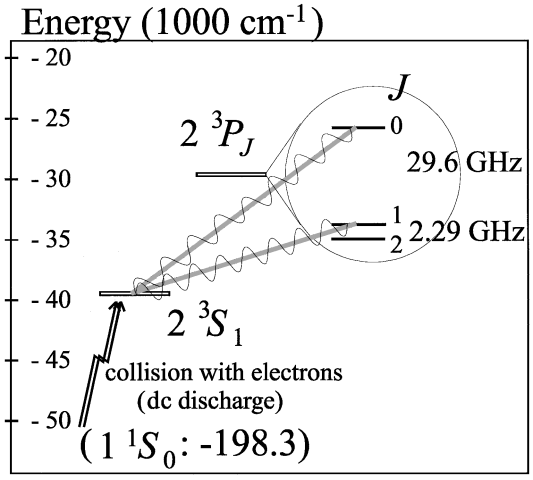

FIG. 1. The fine structure energy splitting is measured as the difference of two optical transition frequencies (λ = 1083 nm).

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Our experimental setup is based on the detection of saturated fluorescence from a beam of metastable helium atoms. We stress that detecting fluorescence instead of metastable atoms, though less efficient, enabled us to perform a measurement in the absence of any bias magnetic field.

Two lasers are employed: the first is the fixed-frequency reference oscillator (master laser) and the second is scanned across the He^* resonances (slave laser).

The laser sources are semiconductor diode lasers emitting at $\lambda = 1083$ nm, whose linewidth is reduced to less than 200 kHz using the extended cavity configuration described in [12]. We stabilized the master laser frequency on the saturated absorption signal of the $2\,^3S_1 \rightarrow 2\,^3P_2$ transition in a cell rf discharge. The slave laser is phase locked to the master [12], with a frequency offset provided by a microwave synthesizer: the scan across the resonance is accomplished by a personal computer simply by varying in steps the frequency offset. In this way, we control the frequency difference of the two lasers with a precision of 0.2 Hz in 1 s (root Allan variance): therefore, the uncertainty of the slave laser frequency scan is determined only by the stability of the master laser frequency (7 kHz in 1 s, root Allan variance).

The atomic sample is a beam of He atoms, excited to the metastable $2\,^3S_1$ level by means of a dc discharge, similar to that described in [13]. Though helium atoms are cooled to liquid nitrogen temperature before entering the discharge, they are subsequently heated by longitudinal electron collisions. Recording the Doppler profile of the $2\,^3S_1 \rightarrow 2\,^3P_2$, we have measured the longitudinal velocity distribution of the atomic flux, which is well fitted by a "generalized Maxwellian":

$$\frac{d\Phi}{dv} \propto (v/u)^{\beta} \exp[-(v/u)^2], \qquad (1)$$

with $\beta = 6.5(1)$ and $u = 1.56(7) \times 10^3$ m/s. The transverse velocity distribution gives rise to a typical Doppler broadened linewidth of 120 MHz (FWHM). An estimate of the flux of metastable $2\,^3S_1$ atoms is inferred from the detected fluorescence to be 10^{15} atoms/s $\cdot$ str.

The atomic beam is orthogonally probed by a linearly polarized standing wave obtained by retroreflection from a dielectric mirror. The power imbalance between the two counterpropagating waves is as low as 1%, since all of the optical elements in the return path are antireflection coated. The laser beam is given a Gaussian shaped intensity profile by spatial filtering with a pinhole. The laser power is 0.150(15) mW and the waist is $w = 2.12(14)$ mm, giving a peak intensity of 2.1 mW/cm^2 per beam. A photomultiplier monitors the resonance fluorescence collected over a solid angle of $0.3 \times 4\pi$ sterad, around the direction orthogonal to both the laser polarization and the atomic velocity. We employ third harmonic phase-sensitive detection (0.5 Hz bandwidth) to increase the signal-to-noise ratio and to reduce spurious signals arising from residual amplitude modulation and the Doppler pedestal.

We believe that a major improvement over previous measurements is our shielding the interaction region against magnetic fields by means of a 1-mm-thick high magnetic permittivity (mu-metal) cylinder together with a pair of internal Helmholtz coils. As discussed by Hughes and co-workers [14], with substantial magnetic fields significant corrections have to be introduced to take into account the quadratic Zeeman effect. In our experience, even with pure (better than 99.9%) π polarized laser beams, a magnetic field of 16 μT shifted the measured fine structure separation by tens of kHz. Such shifts cannot be accounted for by simply admitting a slight ellipticity in laser polarization: an exhaustive explanation probably requires taking into account simultaneously Larmor precession and optical pumping, as in Ref. [15], together with photon recoil. In our experimental situation, the residual field measured with a commercial magnetometer is lower than 0.1 μT in a 1 cm^3 volume centered around the interaction region. Final minimization of magnetic field is achieved *in situ* with the optical Hanle effect. This is a crucial point of difference with respect to previous measurements [7,11] that have been performed, respectively, with moderate (30 mT) and high (between 10 mT an 0.2 T) magnetic fields.

The collection optics have been carefully devised to guarantee that all source points in the interaction region uniformly illuminate the photocathode. A single acquisition is composed of 101 points, with an integration time of 0.5 s/pt. The line is scanned twice, increasing and decreasing the frequency. All of the lines have been fitted with the third harmonic component of a frequency modulated Lorentzian, allowing for an additive straight line which mimics the presence of the Doppler pedestal: the fitting uncertainty for the linecenter is 10 to 20 kHz.

We accumulated 191 measurements of the ν_{01} interval (see Fig. 2) over several months. The weighted average is

$$\nu_{01}(\text{stat}) = 29\,616\,931.3 \pm 1.8\,\text{kHz}\,.$$

We attribute the main source of statistical error to the reference frequency long-term instability and to fluctuations of the atomic beam intensity and velocity.

A significant effort was devoted to identifying and eliminating any source of systematic effects.

A mutual misalignment of the counterpropagating laser beams combined with the strongly anisotropic velocity distribution shifts each linecenter frequency by about 2.5 MHz/mrad. Randomization of this shift is accomplished by periodic realignment of the retroreflected beam; moreover, it does not affect the fine structure interval since it cancels in subtracting the transition frequencies. The same cancellation occurs for second order Doppler shifts [14.4(4) kHz for each transition frequency]. For each transition, ac Stark shifts (light shifts) are calculated from numerical solution of the optical Bloch equations,

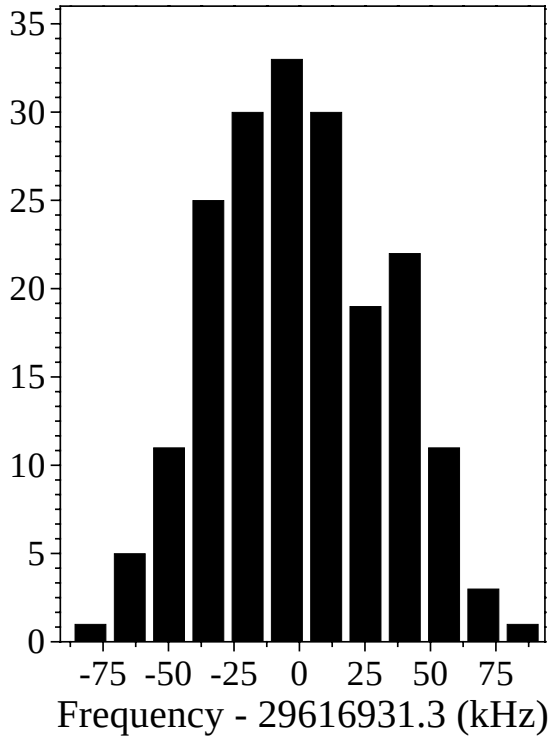

FIG. 2. Histogram showing the spread of the ν_{01} measurements around the statistical mean (15 kHz/bin).

more accurate than the simple formula derived from second order perturbation theory; we applied a correction of $-0.49(6)$ kHz to the measured interval. Experimentally, a check was done by doubling the laser power, and we did not find any appreciable difference. Thanks to the mu-metal shield, the error arising from the residual magnetic field is estimated to be ± 0.3 kHz.

The calibration of the microwave synthesizer time base has introduced the largest correction to the ν_{01} measured value: 2.44(1) parts in 10^7. However, it does not spoil the attained accuracy since it is exactly determined by comparison with a Rb frequency standard traceable to cesium primary standards with an accuracy of 1 part in 10^{11}. The uncertainty associated with this correction arises from the measured long-term stability of the synthesizer.

Momentum exchange between atoms and laser photons gives rise to radiation forces, whose effects are generically referred to as "laser cooling." Generally neglected in high precision spectroscopy, multiple atomic recoils have nonetheless proved to be relevant [16]. In helium, due to the low atomic mass, the transition linecenter can indeed be offset: on the closed $2\,^3S_1 \rightarrow 2\,^3P_2$ transition we have observed line shifts as large as 10% of the natural linewidth for interaction times tenfold longer than the $2\,^3P$ level lifetime ($\tau = 97.88$ ns) [17]. The "open" $2\,^3S_1 \rightarrow 2\,^3P_{0,1}$ transitions are much less affected by mechanical effects since, after a few absorption–spontaneous emission processes, atoms are rapidly pumped into "dark" Zeeman sublevels. Two qualitative arguments permit us at least to understand the origin of the shift. First, we can consider that the radiation pressure collimates (decollimates) the atomic beam for red (blue) detuning, i.e., $\omega_L > \omega_0$ ($\omega_L < \omega_0$). Second, we can consider that the dipole force partially localizes the atoms at the antinodes (nodes) of the laser standing wave, for red (blue) detuning. In both cases, the fluorescence is higher on the red side of the resonance than on the blue side. This causes a shift of the linecenter towards the blue.

We have studied this radiation-force induced shift from both the experimental and theoretical aspects. Experimentally, we have changed the interaction time between atoms and laser by varying the beam width along the direction of flight of the atoms. To this purpose, an adjustable slit was placed in the path of the laser beam and imaged onto the interaction region with a suitable optical setup avoiding wave front distortion. Then, we have remeasured the ν_{01} interval with a 0.95 mm wide laser beam and found a difference of $+9.4(2.1)$ kHz with respect to the value obtained without the slit. On the theoretical side, a simple 1D model based on the optical Bloch equations was solved by numerical integration: it confirmed the origin of the shift and proved that it is not spurious. The analysis was carried out for both experimental situations, with and without the slit. The calculated shifts of the ν_{01} interval are $-3.0(5)$ and $-11.7(9)$ kHz, respectively. Their difference, $+8.7(1.0)$ kHz, is in very good agreement with its measured value. Therefore, we have introduced a correction of $+11.7(9)$ kHz over the ν_{01} value obtained without the slit.

All of the above corrections are summarized in Table I. Our final value for the ν_{01} fine structure separation is

$$\nu_{01} = 29\,616\,949.7(2.0)\ \text{kHz}\,.$$

The 68 ppb accuracy is the best achieved so far in helium fine structure measurements.

In Table II, we compare with previously reported measurements and with the latest theoretical result [18]: our result is closer, yet not in agreement, with the slightly less accurate value of Ref. [7]. Also, we notice a mild agreement with an unpublished less precise measurement performed at Harvard, in a cell discharge [19].

Owing to many cancellations, fine structure energy splittings are calculated with a better absolute accuracy

TABLE I. Budget of corrections and uncertainties; all the values are in kHz.

Statistical ν_{01} value	29 616 931.3(1.8)
ac Stark shift	$-0.49(6)$
Residual magnetic field	<0.3
Second-order Doppler	<0.002
Residual amplitude modulation	<0.1
Synthesizer calibration	$+7.23(3)$
Recoil-induced shift	$+11.7(9)$
Final ν_{01} value	29 616 949.7(2.0)

TABLE II. Comparison of this work with previous measurements and the theoretical prediction.

	ν_{01} value (kHz)	Difference (kHz)
This work	29 616 949.7(2.0)	...
Shiner *et al.* [7]	29 616 962(3)	−12(3)
Hughes *et al.* [11]	29 616 844(21)	+106(21)
Theory [18]	29 616 974(10)	−24(10)

than energy levels. Early in 1974, Douglas and Kroll [20] identified all of the contributions of order $O(mc^2\alpha^6)$, but these are not sufficient to reach a precision of 1 kHz. Recently, a full derivation of terms $O(mc^2(\alpha^7 + \alpha^7 \log \alpha))$ has been carried out [21], but only terms $O(mc^2\alpha^7 \log \alpha)$ have been fully evaluated (i.e., their expectation values have been calculated). As soon as the evaluation of all terms $O(mc^2\alpha^7)$ is completed, the uncertainty of ν_{01} is expected to reduce to the 1 kHz level [18].

We can speculate that our measurement combined with a 1 kHz accurate theoretical prediction will yield a value for α with a 38 ppb uncertainty. In view of the forthcoming adjustment of physical constants, it is instructive to compare the above precision with that of alternative α measurements.

At present, the most accurate (3.8 ppb) value of α is inferred from the electron magnetic anomaly $g_e - 2$ [22]: $\alpha^{-1} = 137.035\,999\,93(52)$. The measurements of the von Klitzing constant $R_K = h/e^2$, via the quantum Hall, effect and of the Planck constant to neutron mass ratio, by means of neutron interferometry, provide α values with 24 ppb [23] and 39 ppb [24] accuracy, respectively, which are in agreement with the $g_e - 2$ value [25]. But a discrepancy as large as 0.25 ppm appears for the α value measured via the Josephson effect [26]. Our measurement, together with the foreseen improvement in theory, should lead to a new atomic determination of α which will hopefully clarify this situation.

We are indebted to Dr. A. Godone and Dr. F. Cordara from IEN–Galileo Ferraris, Torino, Italy, who kindly loaned us the Rb frequency standard. We are grateful to Prof. G. Gabrielse who communicated to us the results of the unpublished measurement from his group. This work was supported by the Istituto Nazionale di Fisica Nucleare (INFN) under project Alfelio.

Note added in proof.—A new microwave measurement has been recently published: $\nu_{01} = 29\,616\,966(13)$ kHz [C. H. Storry and E. A. Hessels, Phys. Rev. A **58**, R8 (1998)].

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