First Pure Frequency Measurement of an Optical Transition in Helium: Lamb Shift of the 2^3S_1 Metastable Level

F. S. Pavone, F. Marin, P. De Natale, and M. Inguscio

Department of Physics and European Laboratory for Nonlinear Spectroscopy (LENS), University of Florence, Largo E. Fermi, 2 I-50125 Firenze, Italy

F. Biraben

Laboratoire "Kastler-Brossel" de l' Ecole Normale Supérieure, Université Pierre et Marie Curie, Paris, France

(Received 19 January 1994; revised manuscript received 8 April 1994)

The 2^3S_1-3^3P_0 ^{4}He transition at $\lambda_0 = 389$ nm is measured with respect to a previously frequency calibrated ^{87}Rb two-photon transition at $2\lambda_0 = 778$ nm. The ^{4}He absolute frequency is 770 732 839 058 (190) kHz, with an accuracy of 2.4 parts in 10^{10}. A Lamb shift value of +4057.61 (79) MHz, with 600 kHz uncertainty arising from the theoretical position of the 3^3P_0 energy level, is extracted for the 2^3S_1 level. This value is more than 2 orders of magnitude more accurate than the current best theoretical predictions.

PACS numbers: 32.30.Jc, 12.20.Fv, 31.30.Jv

Laser spectroscopy of simple atomic systems is a powerful tool for fundamental studies. Meaningful experiments have been performed on two-body systems and, for instance, the spectroscopy of hydrogen provides the best values of the Rydberg constant [1–3]. Advances in the theory of two-electron systems have stimulated experiments on helium as well [4–11]. At present, nonrelativistic wave functions of helium may be calculated with accuracies much higher than current experimental precision and the relativistic corrections are sufficiently well known to make this prototypic many-body system a candidate for quantum electrodynamics (QED) tests [12]. Helium energy levels so far have been obtained from wavelength measurement. Direct absolute frequency measurements are highly desirable since their accuracy does not suffer from the limitations of wavelength interferometry, such as phase shifts on the optics and diffraction. In this Letter, we report the first pure frequency measurement of an optical transition of helium, which also yields the best accuracy reported to date. The absolute frequency of the 2^3S_1-3^3P_0 transition at 389 nm is measured to 2.4 parts in 10^{10}. The Lamb shift of the metastable 2^3S_1 level, extracted with an accuracy of 1.9 parts in 10^4, is likely to stimulate further improvements in the theory.

In our experiment, the radiation at 389 nm is produced, in Ref. [10] for the wavelength determination and in Ref. [13] for the isotope effect investigation, by frequency doubling Ti-doped sapphire laser at $\nu = 385$ THz ($\lambda = 778$ nm).

The two-photon transitions $5S_{1/2}$-$5D_{3/2,5/2}$ of rubidium can be excited at nearly the same wavelength. The absolute optical frequencies of the Rb hyperfine components, excited by a Ti: sapphire laser, were measured to 1.3 parts in 10^{11} [14]. For this measurement it was convenient to use the close coincidence with the difference in frequency between the iodine stabilized HeNe laser and the methane stabilized HeNe laser, at 473 THz (633 nm) and 88 THz (3.39 μm), respectively. Therefore, the measured Rb transitions provide new references in the visible. Indeed, for our experiment on He we use for the first time a line narrowed diode laser locked on Rb.

The general scheme of the apparatus is shown in Fig. 1. ^{4}He is prepared in the 2^3S_1 metastable state in a low-pressure radio frequency discharge. Two probe beams at 389 nm pass through the cell and only one of them interacts with the counter propagating pump beam. By detecting the difference in the intensities of the two transmitted probe beams, the Gaussian contribution to the absorption profile of the 2^3S_1-3^3P_0 transition is eliminated. Derivative signals obtained by frequency modulation and first harmonic detection, as those shown in Fig. 2(a), are used to frequency lock the lasers at the line center. For the frequency standard at 385 THz, we use a low power

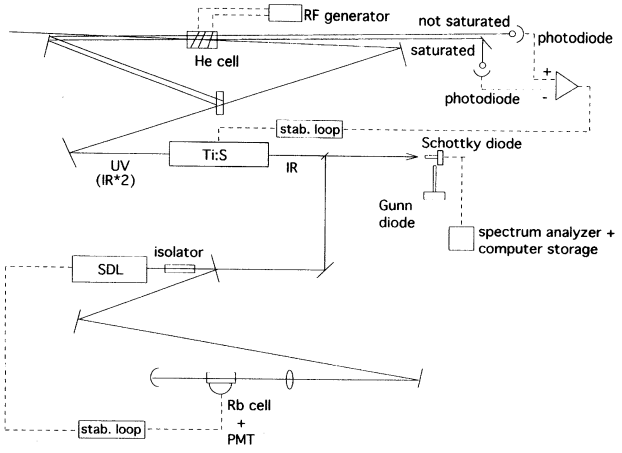

FIG. 1. Scheme of the experimental apparatus.

0031-9007/94/73(1)/42(4)$06.00

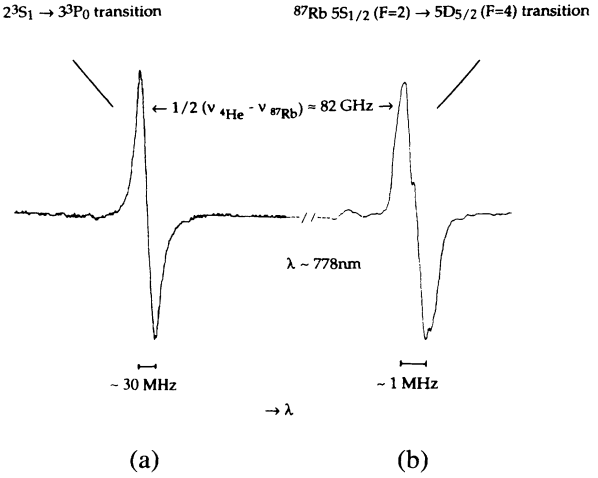

FIG. 2. Typical derivative signals on which the frequency-doubled Ti-S and the AlGaAs diode lasers were locked. (a) Saturated absorption of the 2^3S_1-3^3P_0 ^{4}He transition (the detection bandwidth is about 250 Hz). (b) Doppler-free two-photon $5S_{1/2}(F = 2)$–$5D_{5/2}(F = 4)$ ^{87}Rb transition (1.6 mW laser power, detection bandwidth ~80 Hz).

diode laser that is locked to the $5S_{1/2}$-$5D_{5/2}$ two-photon transition in Rb. While broadband diode laser excitation of two-photon transitions in rubidium has been very recently reported [15], in our experiment the 778 nm AlGaAs laser is mounted in a grating feedback scheme [16] for line narrowing. This allows the full resolution of the hyperfine atomic structure. The laser beam is focused into the same Rb cell used in Ref. [14] for the absolute frequency measurement, by means of a lens ($f = 40$ cm) and then retroreflected by a spherical mirror with a radius of curvature of 80 cm. The two photon transitions are observed by detecting the fluorescence from the $6P$-$5S$ decay at 420 nm. A derivative recording of the ^{87}Rb $5S_{1/2}(F = 2)$–$5D_{5/2}(F = 4)$ transition is shown in Fig. 2(b). The measurement of this transition recently performed in Paris [17] in a pure frequency scheme provides us with an absolute frequency reference at $\nu = 385\,284\,566\,366.3$ (6.0) kHz. After compensation of the residual magnetic field, the Rb resonance width has been reduced to about 1 MHz.

The comparison of the frequencies of the two lasers, one locked on He and the other one on Rb, is possible thanks to the use of a fast nonlinear detector. For this purpose, we have used a Schottky-barrier diode formed by the contact between an electrolytically sharpened tungsten wire and a GaAs semiconductor base [18]. For further analysis, the beat note frequency is lowered by mixing with radiation from an intermediate frequency oscillator. In particular, we have used a Gunn diode oscillating at 82 GHz, phase locked to a low noise quartz oscillator.

Low frequency beat notes between the Gunn diode frequency and the frequency differences of the lasers were observed with a typical signal to noise ratio of 20 dB in a 300 kHz bandwidth. Several independent measurements were performed at different pressures, as shown in Fig. 3. From these data, the zero pressure beat note frequency value has been extrapolated by a linear fit with an uncertainty of 70 kHz (1 standard error). The uncertainty due to the discharge perturbation effect has been estimated by performing measurements at different values of the discharge current. A systematic frequency shift is also caused by a slight line asymmetry due to the presence of a residual magnetic field in combination with a not perfect linear polarization of the laser light. A correction has been estimated by studying the Zeeman structure induced by an applied magnetic field. Furthermore, the presence of the $F = 2$–3 component of Rb, only 14 MHz far from the one ($F = 2$–4) used as reference, causes a frequency shift (overmodulation correction) because of the laser frequency modulation used to obtain derivative signals. Two in-phase modulations with depths of about 7 MHz, respectively for Rb and He signals, have been chosen to avoid modulation broadening of the beat note.

After the corrections for systematic reported in Table I, and the use of the reference frequency, the value of 770 732 839 058 (190) kHz is obtained for the 2^3S_1-3^3P_0 ^{4}He transition. In a previous experiment, Adams *et al.* [10] measured the wavelength of the crossover resonance among the 2^3S_1-3^3P_2 and 2^3S_1-3^3P_1 transitions. Combining their data with the existing 3^3P fine structure values [19], one can deduce a value of 770 732 839.8 (5) MHz for the 2^3S_1-3^3P_0 component, using the latest value for the frequency of the iodine stabilized HeNe [20]. The more accurate and direct frequency determination of the present work is in marginal agreement (1.4σ) with the wavelength measurement. More interesting, our

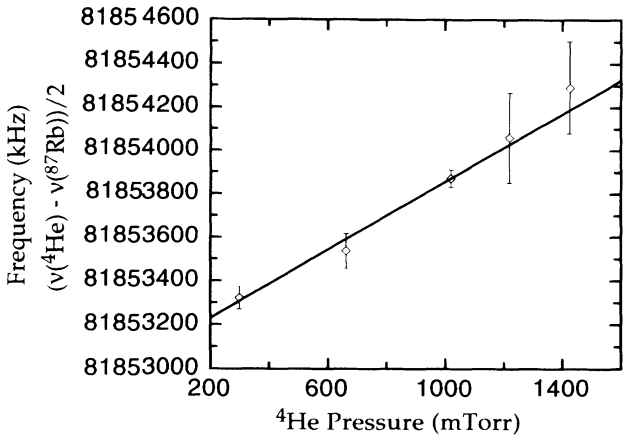

FIG. 3. Dependence on the helium pressure of the difference between half the 2^3S_1-3^3P_0 ^{4}He frequency and the Rb frequency standard. Error bars reflect the standard error of 12 independent measurements for each pressure.

TABLE I. Different contributions to the frequency measurement of the 2^3S_1-3^3P_0 ^{4}He transition.

	Value (kHz)	Uncertainty (kHz)[a]
^{4}He(2^3S_1-3^3P_0) $-^{87}$Rb[$5S_{1/2}(F=2) \rightarrow 5D_{5/2}(F=4)$] Measured separation at 778 nm	81 853 071	70
Uncertainty due to discharge effect	0	41
^{87}Rb$5S_{1/2}(F=2) \rightarrow 5D_{5/2}(F=4)$ frequency	385 284 566 366.3[b]	6
Correction for the light shift on Rb	−10	5
Correction for the overmodulation on Rb	+20	10[c]
Sum	385 366 419 437	92
Sum ×2	770 732 838 895	184
Correction for the light shift on ^{4}He	+4	
Correction for the residual magnetic field on ^{4}He	+151	51
Second order Doppler shift on ^{4}He	+8.4	
^{4}He $2^3S_1 \rightarrow 3^3P_0$ final result	770 732 839 058	190

[a]Uncertainties reported are 1 σ statistical errors.
[b]See Ref. [17].
[c]Not a statistical error.

measurement provides a precise determination of the 2^3S_1 energy after a proper comparison with theory. The non-QED contributions to the energy of the levels involved in the transition can be calculated with very high accuracy, essentially limited by the knowledge of the Rydberg constant [12]. Uncertainties of 18 and 8 kHz for $n = 2$ and $n = 3$ levels, respectively, are found when the most recent Rydberg value is used [3]. QED corrections are predicted with less accuracy, in particular for $n = 2$, and precise measurements are of crucial importance, as recently shown for the singlet states [8,9,21]. As for the triplet states, the Lamb shift, and hence the total energy, of the 3^3P_0 level, is predicted with an accuracy of 600 kHz while the value for the 2^3S_1 level is much less accurate [12]. The accurate frequency measurement performed in this work for the separation of the two levels can then be used to set the 2^3S_1 energy level, relative to He^+ ($1s$), at −1 152 842 742.50 MHz with an uncertainty of 600 kHz from theoretical calculation of the 3^3P_0 QED correction and 190 kHz from the present experiment. By subtracting the non-QED contributions, we obtain a value, for the 2^3S_1 Lamb shift, of +4057.61 (79) MHz with a theoretical uncertainty of 600 kHz and an experimental uncertainty of 190 kHz. A similar procedure was applied by Drake [12] to the early wavelength measurements [5] of the two-photon transitions to $(4-5)^3D$ states whose QED terms are well known. The Lamb shift, recalculated with the latest Rydberg value [3] and the new HeNe/I_2 laser frequency value [20], amounts to 4056.1 (2.4) MHz. Our result, 3 times more accurate, confirms the significant deviation from the theoretical value (4043 MHz). This should stimulate further developments in the theory, in particular for the one electron Lamb-shift term that is the main source of uncertainty.

In conclusion, we have performed the first direct frequency measurement of an optical transition on helium measuring the 2^3S_1-3^3P_0 ^{4}He separation. A frequency standard based on a two-photon Rb transition has been used as reference. Our result is in agreement with the less accurate value that can be indirectly obtained from previous wavelength measurements. By using the accurate theoretical prediction for the 3^3P_0 level, the energy of the metastable 2^3S_1 level is obtained with 3 times improved precision. The 2^3S_1 Lamb shift, determined with an accuracy of 1.9 parts in 10^4, is challenging for theorists because it is 3 orders of magnitude more accurate than theory. Further experimental improvements will come from the replacement of the cell by a helium beam to eliminate perturbations mainly due to collisions and electric field effects in the discharge. Also, measurements will be extended to other fine structure components and to ^{3}He. Besides, when QED calculations will be improved, our measurements will allow the quantitative evaluation of other contributions to the atomic energy, possibly leading to a direct determination of the ^{4}He nuclear radius (R_n). This could eventually allow a comparison with data extrapolated ($R_n = 1.671 \pm 0.014$ fm) from high energy scattering experiments [22], or with the more precise value ($R_n = 1.673 \pm 0.001$ fm) obtained in a μ-^{4}He nucleus bound system [23,24].

Finally, when more precise measurements are made for other transitions starting from the 2^3S_1 level, for example, the 2^3S-2^3P at 1.08 μm, our data will allow extracting the energy of the upper levels involved.

We would like to thank G. W. F. Drake for many useful discussions and comments, G. Tofani, V. Natale, and M. Catarzi of the Osservatorio Astrofisico di Arcetri, Firenze, for the kind loan of the Gunn diode. Laboratoire "Kastler-Brossel" de l'Normale Supérieure is associeé

au CNRS URA 18. This work was performed in the frame of a ECC Science Program Cooperation, Contract No. SC1CT92-0816.

[1] T. Andreae, W. König, R. Wynands, D. Leibfried, F. Schmidt-Kaler, C. Zimmermann, D. Meschede, and T. W. Hänsch, Phys. Rev. Lett. **69**, 1923 (1992).
[2] F. Nez, M. D. Plimmer, S. Bourzeix, L. Julien, F. Biraben, R. Felder, O. Acef, J. J. Zondy, P. Laurent, A. Clairon, M. Abed, Y. Millerioux, and P. Juncar, Phys. Rev. Lett. **69**, 2326 (1992).
[3] F. Nez, D. Plimmer, S. Bourzeix, L. Julien, F. Biraben, R. Felder, Y. Millerioux, and P. De Natale, Europhys. Lett. **24**, 635 (1993).
[4] E. Giacobino and F. Biraben, J. Phys. B **15**, L385 (1982).
[5] L. Hlousek, S. A. Lee, and W. M. Fairbank, Jr., Phys. Rev. Lett. **50**, 328 (1983).
[6] T. J. Sears, S. C. Foster, and A. R. W. Mc Kellar, J. Opt. Soc. Am. B **3**, 1037 (1986).
[7] P. Zhao, J. R. Lawall, A. W. Kam, M. D. Linsday, F. M. Pipkin, and W. Lichten, Phys. Rev. Lett. **63**, 1593 (1989).
[8] C. J. Sansonetti, J. D. Gillaspy, and C. L. Cromer, Phys. Rev. Lett. **65**, 2539 (1990).
[9] W. Lichten, D. Shiner, and Z.-X. Zhou, Phys. Rev. A **43**, 1663 (1991).
[10] C. S. Adams, E. Riis, A. I. Ferguson and W. R. C. Rowley, Phys. Rev. A **45**, R2667 (1992).
[11] K. S. E. Eikema, W. Ubachs, W. Vassen, and W. Hogervorst, Phys. Rev. Lett. **71**, 1690 (1993).
[12] G. W. F. Drake, in *Long-Range Casimir Forces: Theory and Recent Experiments on Atomic Systems*, edited by F. S. Levin and D. A. Micha (Plenum, New York, 1993).
[13] F. Marin, F. Minardi, F. Pavone, and M. Inguscio, Phys. Rev. A **49**, 1523 (1994).
[14] F. Nez, F. Biraben, R. Felder, and Y. Millerioux, Opt. Commun. **102**, 432 (1993).
[15] R. E. Ryan, L. A. Westling, and H. J. Metcalf, J. Opt. Soc. Am. B **10**, 1643 (1993).
[16] G. M. Tino, L. Hollberg, A. Sasso, M. Inguscio, and M. Barsanti, Phys. Rev. Lett. **64**, 25 (1990).
[17] O. Acef, R. Felder, and Y. Millerioux (private communication).
[18] H. U. Daniel, B. Maurer, and M. Steiner, Appl. Phys. B **30**, 189 (1983).
[19] D. H. Yang, P. Mc Nicholl, and H. Metcalf, Phys. Rev. A **33**, 1725 (1986).
[20] O. Acef, J. J. Zondy, M. Abed, D. G. Rovera, A. H. Gerard, A. Clairon, Ph. Laurent, Y. Millerioux, and P. Juncar, Opt. Commun **97**, 29 (1993).
[21] G. W. F. Drake, Adv. At. Mol. Opt. Phys. (to be published).
[22] C. R. Ottermann, G. Kobschall, K. Maurer, K. Rohrich, Ch. Schmitt, and V. H. Walther, Nucl. Phys. **A436**, 688 (1985).
[23] G. Carboni, G. Gorini, L. Palffy, F. Palmonari, G. Torelli, and E. Zavattini, Nucl. Phys. **A278**, 381 (1977).
[24] E. Borie and G. A. Rinker, Phys. Rev. A **18**, 324 (1978).