Accurate Measurement of the 2^3S_1-3^3D_1 Two-Photon Transition Frequency in Helium: New Determination of the 2^3S_1 Lamb Shift

C. Dorrer,* F. Nez, B. de Beauvoir, L. Julien, and F. Biraben

Laboratoire Kastler Brossel, Ecole Normale Supérieure et Université Pierre et Marie Curie, Laboratoire associé au CNRS URA18, 4 Place Jussieu, Tour 12 E01, 75252 Paris Cedex 05, France

(Received 10 February 1997)

We have performed a precise measurement of the 2^3S_1-3^3D_1 two-photon transition frequency in ^{4}He at 762 nm. The 2^3S_1-3^3D_1 frequency is 786 823 850.002(56) MHz, with a relative uncertainty of 7.1×10^{-11}. The deduced 2^3S_1 Lamb shift is 4057.276(60) MHz. This result, the most accurate at the present time, reduces the uncertainty in the 2^3S_1 Lamb shift by 1 order of magnitude and is 100 times more precise than the theoretical prediction [4062.3(8.0) MHz]. [S0031-9007(97)03086-X]

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

In simple atomic systems, calculations, as well as experimental measurements, have reached an impressive accuracy. The hydrogen atom is the most significant case. Its properties have been calculated very precisely, with a relative accuracy of order 10^{-11} [1,2], and, at the same time, experimental measurements have been performed at the same level of precision [3–6]. Because it is the simplest multielectron atom, atomic helium plays an important role. The helium level energy is conventionally expressed as the sum of three terms: the nonrelativistic energy, the lowest order relativistic correction, and the Lamb shift, which includes the quantum electrodynamics corrections (QED) and the high order relativistic terms. As the theoretical uncertainties of the first two terms are well below the experimental accuracy [7], precise measurements in helium provide an important test of QED calculations. Thanks to a better evaluation of the Bethe logarithm for the $1S$ and $2S$ states [8], the theoretical uncertainties are now 180 MHz for the $1S$ state and 8 MHz for the $2S$ and $2P$ states [9]. For the D states, the theoretical uncertainties are much smaller, for instance, 20 kHz for the $3D$ states. Consequently, it is possible to use the D states as points of reference in the interpretation of the spectroscopic results. For example, very precise measurements of $2S$-nD transitions in helium provide accurate determination of the 2^1S and 2^3S Lamb shifts.

Helium has been studied experimentally for many years. Optical spectroscopy involving the ground state is very difficult since the lowest excited level lies about 20 eV above it. Recently, a beautiful experiment has been performed in Amsterdam, by Hogervorst *et al.* who have measured the frequency of the 1^1S-2^1P transition in both ^{4}He and ^{3}He at 58.4 nm [10]. The 1^1S Lamb shift, in agreement with the theoretical one, was deduced with a relative uncertainty of 4.2×10^{-3}. Most other spectroscopic studies use the 2^1S and 2^3S metastable states as lower level [11–18]. For instance, the combination of the Lamb shifts involved in the 2^3S-2^3P transition has been measured by an interferometric method with a relative uncertainty of 1.3×10^{-5} [12]. In Florence, the group of Inguscio has realized the first pure frequency measurement of an optical transition in helium and deduced the 2^3S Lamb shift with an accuracy of 790 kHz [13]. The 2^3S-n^3D two-photon transitions were extensively studied during the 1980's [16,17] and, more recently, the 2^1S-n^1D transitions (with n in the range 7–20) were investigated at Yale [18]. In this Letter, we report a very precise measurement of the 2^3S_1-3^3D_1 two-photon transition at 762 nm in ^{4}He and we deduce an improved value for the 2^3S_1 Lamb shift.

The heart of our measurement apparatus is a very stable Fabry-Pérot reference cavity, labeled FPR. It consists of a 50 cm zerodur rod and two silver coated mirrors, one flat and one spherical (60 cm curvature radius). The finesse is about 100 at 760 nm. The FPR cavity is scanned with a piezoelectric translator and locked to an iodine stabilized helium-neon laser (He-Ne/I_2) at 633 nm. The 762 nm radiation used for the two-photon excitation in helium is provided by a homemade titanium sapphire laser (TiSa) described in Ref. [19]. This laser is actively stabilized on a second cavity by the sideband technique, the frequency jitter being reduced below 10 kHz. The laser is then locked on a fringe of the FPR cavity. This second servo loop transfers the long term stability of the He-Ne/I_2 standard to the TiSa laser. The laser frequency scanning is made with an acoustooptic modulator placed between the FPR cavity and the laser. The FPR cavity is also used to determine the TiSa frequency. Frequency measurements with a Fabry-Pérot cavity are difficult, owing to the reflective and Fresnel phase shifts. The reflective phase shift is influenced by the mirror coatings and is wavelength dependent. To eliminate this effect, a first method is to change the length of the Fabry-Pérot cavity. We used this technique in our group in 1989 to measure precisely some hydrogen frequencies, but this method is long and difficult to implement [20]. In the present experiment, the length of the FPR cavity was fixed and we simply used the FPR cavity to make a linear interpolation between two, very

 0031-9007/97/78(19)/3658(4)$10.00

well known reference frequencies. The first is provided by a new frequency standard, a laser diode stabilized on the $5S$-$5D$ two-photon transition of rubidium at 778 nm, whose frequency has been measured recently with an uncertainty of only 2 kHz [21]. The second reference frequency is the $2S_{1/2}$-$10D_{5/2}$ two-photon transition in deuterium at 760 nm. We have chosen this transition because it lies close to the 2^3S-3^3D transition in helium (about 2 nm). Taking advantage of our apparatus used for the Rydberg constant measurement [3,20], we have made several recordings of this line. Using the recent experimental value of the $2S_{1/2}$-$8D_{5/2}$ two-photon frequency in deuterium [3], and the theoretical value of the $8D_{5/2}$-$10D_{5/2}$ splitting, we are able to know the frequency of the closest FPR fringe with an uncertainty of 10 kHz. To evaluate the effect of the reflective phase shift, we have used a third reference frequency, the 633 nm radiation of the He-Ne/I_2 laser. Immediately after the helium experiment, we have measured the frequency of our He-Ne/I_2 standard with an uncertainty of 4 kHz, using the same scheme as described in Ref. [22]. If we simulate the effect of the reflective phase shift by a slight, linear variation of the FPR free spectral range versus the frequency, we can make a quadratic interpolation between these three reference frequencies. The result, for the FPR fringe close to the helium line, differs from the linear interpolation by only 0.8 kHz. Consequently, the variation of the reflective phase shift between the two reference frequencies at 760 and 778 nm is negligible. This is due to the fact that we use silver coated mirrors which are very broad band. Finally, we estimate the uncertainty on the frequency of the FPR fringe used to stabilize the TiSa laser close to the helium line at 10 kHz. In addition, we have to take into account the accuracy of the servo loop of the TiSa laser on the FPR fringe, which is also estimated to be 10 kHz.

The apparatus used for the helium experiment is the following. The cell is a sphere (6 cm in diameter) with Brewster windows. A large spherical cell is necessary to avoid quenching of the metastable state on the wall. The cell is filled with ^{4}He gas. A radio-frequency discharge, at 14 MHz, populates the 2^3S_1 metastable states. The Earth's magnetic field is compensated by a system of three Helmoltz coils. To enhance the optical power seen by the atoms and to obtain two counterpropagating waves, the cell is placed in a built-up cavity formed by two spherical mirrors (77 and 104 mm curvature radius, 150 mm apart). This cavity maintained in resonance with the TiSa laser using a lock-in amplifier. The two-photon transition is detected by monitoring the 3^3D_1-2^3P fluorescence at 587 nm which is collected with a large lens (150 mm of diameter and 100 mm of focal length) and detected with a photomultiplier. To avoid parasitic light due to the discharge, the discharge is pulsed at 10 kHz and detection carried out in the afterglow regime. The data acquisition is driven by a computer. The radio frequency is switched on only during 5 μs, then we look at the photomultiplier signal for 40 μs starting 40 μs later. This signal is sent in an integrator which is read once every second by the computer. In addition, this delay reduces the effect of parasitic electric field produced by the discharge: no significant frequency shift was observed when the radio-frequency power was varied by a factor of 2. Ten scans on the line are combined to obtain the signal shown in Fig. 1.

To study the pressure shift, we filled the cell with seven different gas pressures, in the range 0.05–0.5 Torr. For each pressure, the two-photon transition was recorded for various optical powers (typically ten different one-way powers from 9 to 26 W inside the cavity). A theoretical line shape is fitted on each experimental signal (see Fig. 1). We employ a convolution of a Lorentzian line shape, which takes into account the natural width and the pressure broadening, and of a double exponential curve, which describes the broadening due to the finite transit time [23]. The parameters of the fit are the parameters of the Lorentzian shape. The fit provides the width of the Lorentzian shape and the frequency of the experimental line. For each pressure, linear extrapolations of these data versus the light power give the width of the Lorentzian shape and the frequency of the 2^3S_1-3^3D_1 transition without light shift. In a second step, we make linear extrapolations versus pressure [see Figs. 2(a) and 2(b)]. Figure 2(a) shows the pressure broadening of the line which is evaluated to be 35.6(1.7) MHz/Torr. The width for zero pressure is 11.33(19) MHz, in very good agreement with the natural width of the 3D level (11.25 MHz [9]). On the other hand, the pressure shift is small [0.4(3) MHz/Torr]. The extrapolation of Fig. 2(b) gives the frequency of the line corrected for the light and pressure shifts. This frequency is also

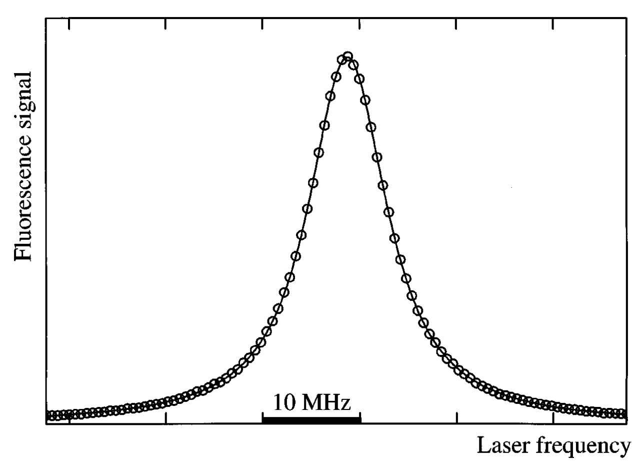

FIG. 1. Line shape of the 2^3S_1-3^3D_1 two-photon transition in ^{4}He. Optical power inside the Fabry-Pérot cavity is 11 W in each direction; the pressure in the cell is 0.151 Torr. The experimental points are fitted with a theoretical curve which takes into account the natural width, the pressure broadening, and the transit time broadening.

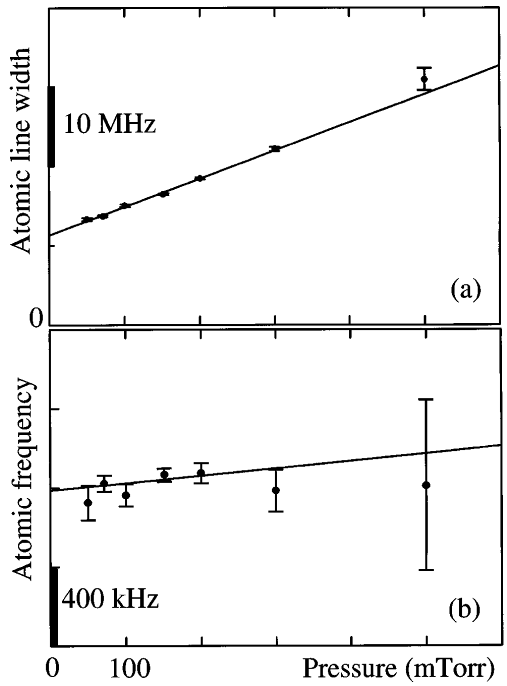

FIG. 2. Extrapolations of the Lorentzian width (a) and of the line position (b) versus the pressure.

shifted by the second order Doppler effect. At 300 K, the correction to the atomic frequency is about 8.1 kHz. Finally, the frequency of the 2^3S_1-3^3D_1 transition in ^{4}He is 786 823 850.002(56) MHz. This uncertainty arises from the interferometric measurement (28 kHz) and the statistics of the extrapolations (in optical power and pressure) of the line center (48 kHz). This result is in good agreement with a previous measurement of the 2^3S_1-3^3D_3 transition [786 822 451(13) MHz [16]] using the theoretical value of the 3^3D_1-3^3D_3 fine structure splitting [1 400.455(24) MHz] given in Ref. [7].

Using the theoretical value of the 3^3D_1 ionization energy [366 018 892.857(20) MHz from Refs. [7,9] after correction of the recent value of the Rydberg constant [3]], we obtain the ionization energy of the 2^3S_1 level. By subtracting the nonrelativistic energy, the first relativistic corrections and the finite nuclear size correction calculated in Ref. [7] for the 2^3S_1 state [1 152 846 800.135(3) MHz after correction for the Rydberg constant], we can deduce the 2^3S_1 Lamb shift. We obtain the value $L(2^3S_1) = 4057.276(60)$ MHz. The uncertainty is essentially due to the experimental determination of the two-photon frequency. Contributions to this uncertainty are listed in Table I. Our precision surpasses by more than 2 orders of magnitude that of the theoretical prediction [4062.3(8.0) MHz]. This result should stimulate new developments on the theoretical side. We compare our result to previous measurements in Fig. 3, where we have extracted the Lamb shift values from other measurements by using the theoretical

TABLE I. Uncertainty budget of the Lamb shift determination.

Frequency determination (experimental)	56 kHz
Lamb shift of the 3^3D_1 level (theoretical)	20 kHz
Nuclear size	3 kHz
Uncertainty in Rydberg constant	8 kHz
Overall uncertainty	60 kHz

predictions and the conservative uncertainties given in Ref. [9]. Our result, 10 times more precise, is in very good agreement with the latest measurement made by Inguscio *et al.* [4057.61(79) MHz] [13]. Our measurement is also in good agreement with the precise result deduced from the 2^3S_1-2^3P_0 and 2^3P_0-3^3D_1 measurements [4056.7(9) MHz] [12,17].

We can also combine our measurement with various measurements starting from the 2^3S_1 state to eliminate the uncertainty due to the 2^3S_1 Lamb shift and so get an accurate determination of the Lamb shift of the higher level. These results are presented in Table II. There is good agreement with the theoretical predictions, which are given with uncertainties from Ref. [9].

In conclusion, we have measured the frequency of 2^3S_1-3^3D_1 transition in ^{4}He with an uncertainty of 7.1×10^{-11}. The deduced 2^3S_1 Lamb shift is the most precise determination at the present time. The comparison between different precise measurements gives access to accurate values of other Lamb shifts. To complete these

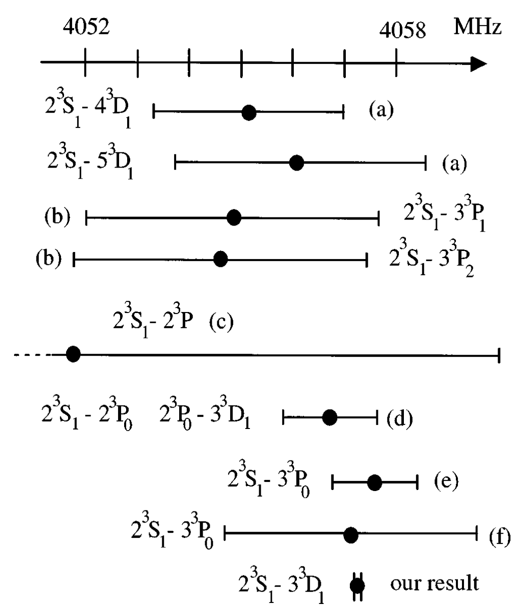

FIG. 3. Comparison of 2^3S_1 Lamb shift deduced from our result and recent high precision measurements in ^{4}He with the theoretical calculations from Ref. [9]: (a) Ref. [17], (b) Ref. [11], (c) Ref. [12], (d) Refs. [12,17], (f) Ref. [13]. (e) corresponds to Ref. [13] using the theoretical values from Ref. [7].

TABLE II. Comparison with other transitions.

2^3S_1-3^3D_1 combined with	Deduced Lamb shift	Theoretical predictions [e]
2^3S_1-3^3P_1 [a]	$L(3^3P_1) = -357.3(1.5)$ MHz	$L(3^3P_1) = -359.7(2.4)$ MHz
2^3S_1-3^3P_2 [a]	$L(3^3P_2) = -356.6(1.5)$ MHz	$L(3^3P_2) = -359.7(2.4)$ MHz
2^3S_1-3^3P_0 [b]	$L(3^3P_0) = -359.51(20)$ MHz	$L(3^3P_0) = -359.7(2.4)$ MHz
2^3S_1-4^3D_1 [c]	$L(4^3D_1) = -10.1(1.8)$ MHz	$L(4^3D_1) = -7.887(10)$ MHz
2^3S_1-5^3D_1 [c]	$L(5^3D_1) = -5.4(2.4)$ MHz	$L(5^3D_1) = -4.224(6)$ MHz
2^3S_1-2^3P [d]	$L(2^3P) = -1253.9(1)$ MHz	$L(2^3P) = -1259.5(8.0)$ MHz

[a]Ref. [11].
[b]Ref. [13].
[c]Ref. [17].
[d]Ref. [12].
[e]Ref. [9].

data, we plan to make a pure frequency measurement of the 2^3S_1-4^3D_1 two photon transition in ^{4}He thanks to the coincidence between the excitation laser frequency and that of the HeNe/I_2 standard (the frequency difference is only $\Delta\nu = 112$ GHz). The same technique frequency scheme could also be applied to measure the 2^1S_0-16^1D transition in ^{4}He ($\Delta\nu \approx 130$ GHz).

The authors thank M. Inguscio for stimulating discussions, P. Indelicato for much theoretical advice, and M. D. Plimmer for a critical reading of the manuscript. This work is partially supported by the Bureau National de Métrologie.

*Present address: Laboratoire d'Optique Appliquée, CNRS URA 1406, ENSTA-Ecole Polytechnique, 91120 Palaiseau, France.

[1] K. Pachucki *et al.*, J. Phys. B **29**, 177 (1996).
[2] S. G. Karshenboim, J. Phys. B **29**, L31 (1996).
[3] B. de Beauvoir *et al.*, Phys. Rev. Lett. **78**, 440 (1997).
[4] M. Weitz *et al.*, Phys. Rev. A **52**, 2664 (1995).
[5] D. J. Berkeland, E. A. Hinds, and M. B. Boshier, Phys. Rev. Lett. **75**, 2470 (1995).
[6] S. Bourzeix *et al.*, Phys. Rev. Lett. **76**, 384 (1996).
[7] G. W. F. Drake, *Long Range Casimir Forces: Theory and Recent Experiments on Atomic Systems*, edited by F. S. Levin and D. A. Micha (Plenum, New York, 1993).
[8] J. D. Baker *et al.*, Bull. Am. Phys. Soc. **38**, 1127 (1993).
[9] G. W. F. Drake in *Atomic, Molecular & Optical Physics Handbook*, edited by G. W. F. Drake (AIP, New York, 1996). In this reference, the theoretical uncertainties are more conservative than in Ref. [7].
[10] K. S. E. Eikema, W. Ubachs, W. Vassen, and W. Hogervorst, Phys. Rev. Lett. **76**, 1216 (1996).
[11] C. S. Adams, E. Riis, A. I. Ferguson, and W. R. C. Rowley, Phys. Rev. A **45**, 2667 (1992).
[12] D. Shiner, R. Dixson, and P. Zhao, Phys. Rev. Lett. **72**, 1802 (1994).
[13] F. S. Pavone, F. Marin, P. De Natale, M. Inguscio, and F. Biraben, Phys. Rev. Lett. **73**, 42 (1994). This value is deduced by using the theoretical calculations from Ref. [7]. With the new calculations from Ref. [9], the uncertainty increases and this result becomes 4057.1(2.4) MHz. These two values are shown in Fig. 3.
[14] D. Shiner, R. Dixson, and V. Vedantham, Phys. Rev. Lett. **74**, 3553 (1995).
[15] F. Marin *et al.*, Z. Phys. D **32**, 285 (1995).
[16] E. Giacobino and F. Biraben, J. Phys. B **15**, L385 (1982).
[17] L. Hlousek, S. A. Lee, and W. M. Fairbank, Phys. Rev. Lett. **50**, 328 (1983).
[18] W. Litchen, D. Shiner, and Zhi-Xiang Zhou, Phys. Rev. A **43**, 1663 (1991).
[19] S. Bourzeix, M. D. Plimmer, F. Nez, L. Julien, and F. Biraben, Opt. Commun. **99**, 89 (1993).
[20] J. C. Garreau, M. Allegrini, L. Julien, and F. Biraben, J. Phys. (Paris) **51**, 2263 (1990); **51**, 2275 (1990); **51**, 2293 (1990).
[21] D. Touahri *et al.*, Opt. Commun. **133**, 471 (1997).
[22] F. Nez, F. Biraben, R. Felder, and Y. Millerioux, Opt. Commun. **102**, 432 (1993).
[23] C. Bordé, C. R. Hebd. Séan. Acad. Sci. **282**, B341 (1976); F. Biraben, M. Bassini, and B. Cagnac, J. Phys. (Paris) **40**, 445 (1979).