RAPID COMMUNICATIONS

The Rapid Communications section is intended for the accelerated publication of important new results. Since manuscripts submitted to this section are given priority treatment both in the editorial office and in production, authors should explain in their submittal letter why the work justifies this special handling. A Rapid Communication should be no longer than 3½ printed pages and must be accompanied by an abstract. Page proofs are sent to authors.

Absolute ionization energy of the 2 ¹S level of helium

Craig J. Sansonetti and J. D. Gillaspy
National Institute of Standards and Technology, Gaithersburg, Maryland 20899
(Received 27 June 1991)

We have measured the absolute wave numbers of a series of transitions from the metastable 2 ¹S level of helium to the n ¹P ($n = 7$-74) excited states. From these data we determine the binding energy of the 2 ¹S level to 2.2 parts in 10^{10} by using a Ritz series formula. This high-precision determination of the 2 ¹S binding energy does not depend on theoretical calculation of the binding energy of any helium level. The result, 32033.228855(7) cm^{-1}, confirms our earlier finding of a relatively large discrepancy with the predicted two-electron Lamb shift for the 2 ¹S level.

PACS number(s): 35.10.Hn, 32.30.Jc, 12.20.Fv

In a recent paper [1], we reported an experimental determination of the binding energy of the first excited singlet state of helium (2 ¹S) that deviated significantly from the best available QED predictions. This result was subsequently confirmed to higher precision by Lichten, Shiner, and Zhou [2]. Although these two experiments observed transitions to different levels and used different metrological techniques, both relied upon the calculated energies of selected high-lying levels to tie experimentally measured transition energies to the ionization limit. It is assumed that these high-lying levels, for which QED corrections to the energy are small, can be calculated with very high accuracy. This approach to determining the 2 ¹S binding energy, while reasonable, remains somewhat unsatisfying since it is not entirely independent of theory. Clearly, any error that may exist in the calculated binding energy of the high-lying levels enters directly into the reported value for the 2 ¹S level.

In the present work we exploit the predictable behavior of an unperturbed Rydberg series of levels to determine the 2 ¹S ionization energy. We assume only the general form of the distribution of energy levels and do not require the results of any precise theoretical calculation. In this paper we will describe this approach, briefly discuss our experiment, and present our result for the 2 ¹S ionization energy. A more detailed report will be given in a subsequent publication.

It has long been known on an empirical basis that an unperturbed series of energy levels can be represented by the extended Ritz formula

$$E_n = E_\infty - R/(n - \delta_n)^2, \qquad (1)$$

where E_n is the energy of the level with principal quantum number n, E_∞ is the ionization energy, R is the finite-mass Rydberg constant for the species of interest, and δ_n is the quantum defect given by

$$\delta_n = A + B/(n - \delta_n)^2 + c/(n - \delta_n)^4 + \cdots .$$

This formula has been derived from general quantum-mechanical considerations by Hartree [3] and has been used extensively to derive ionization energies for atoms and ions. The robust nature of this method is demonstrated, for example, by the consistency previously obtained in deriving the Cs ionization limit from five different Rydberg series [4].

In the current work, we equate the transition energy for the He 2 ¹S-n ¹P transition to E_n. This is equivalent to measuring the energies of the n ¹P states with 2 ¹S as the reference level. We then fit the extended Ritz formula, appropriately modified as discussed below, to our entire measured series of transitions ($n = 7$-74) in order to determine the 2 ¹S ionization energy.

Our experimental apparatus for measuring the 2 ¹S-n ¹P transitions is an improved version of that described in our earlier work [1]. Briefly, a single-frequency cw laser

probes a highly collimated atomic beam of metastable helium atoms in the transverse direction in order to avoid Doppler broadening. For the current experiment we use an intracavity-doubled ring dye laser that produces 0.5–4.0 mW of tunable light near 312 nm with a bandwidth of about 1 MHz. The absolute wavelength of the undoubled laser output is measured in real time by interferometric comparison with an iodine stabilized helium-neon laser using a Fabry-Pérot wave meter developed in our laboratory [5]. For low-lying levels ($n < 25$), we monitor the depletion of the metastable flux at the end of the atomic beam line as the laser is tuned across an excitation resonance. For levels with $n > 22$ we observe the resonances with a better signal-to-noise ratio by field ionizing the highly excited atoms and counting the ions with a multichannel plate detector.

The laser beam intersects the metastable helium beam at the center of a graphite-coated spherical metal cavity which shields the excitation region from external electric fields. The laser and atomic beams pass through the 19-mm-diam cavity by way of four holes whose total area comprises less than 1% of the surface area of the sphere. The lifetime of the Rydberg-state atoms is sufficient to allow them to exit the excitation sphere and pass through a pair of field plates which ionizes them and accelerates them into the detector. The effect of field leakage from the ionization plates into the excitation sphere was unmeasurably small at $n = 74$, even when the ionizing field was artificially increased to 3 times its normal operating value.

A small constant residual electric field remained in the excitation region, presumably due to static charge or surface potentials on the inside of the spherical shield. The Stark shift due to this field was taken into account by adding an additional term to the Ritz formula which scaled as the seventh power of the principal quantum number, as is appropriate for the weak-field Stark shift [6]. The constancy of the residual field was carefully monitored by measuring the $2\,^1S$-$74\,^1P$ transition on each day of data taking.

TABLE I. Helium $2\,^1S$ ionization energy, quantum defect expansion coefficients, and Stark constant determined from the $2\,^1S$-$n\,^1P$ ($n = 7$-74) transitions. The uncertainties given in parentheses are the asymptotic standard deviations of the parameters determined by the nonlinear least-squares-fitting algorithm.

Parameter[a]	Value
E_∞	$32033.228855(5)$ cm^{-1}
A	$-0.012142046(345)$
B	$0.007328(50)$
C	$0.0175(17)$
S	$8.03(11)\times10^{-17}$ cm^{-1}

[a] Parameters derived by fitting the measured $2\,^1S$-$n\,^1P$ transition wave numbers to the formula $\sigma(2\,^1S - n\,^1P) = E_\infty - R_{\text{He}}/(n-\delta_n)^2 + Sn^7$, where $R_{\text{He}} = 109722.273515$ cm^{-1}, the quantum defect is given by $\delta_n = A + B/(n-\delta_n)^2 + C/(n-\delta_n)^4 + \cdots$, and the Sn^7 term models the small residual Stark shift in the data.
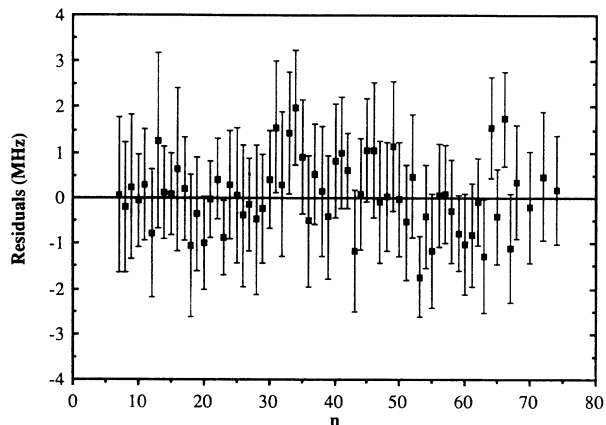

FIG. 1. Residuals (observed minus calculated level value) from the Ritz formula with added Stark term fit to the full set of $n\,^1P$ levels.

By fitting Eq. (1) with the added Stark term to the entire set of data ($n = 7$-74) we determine the $2\,^1S$ binding energy to be $32033.228855(5)$ cm^{-1}. The uncertainty indicated is the standard deviation of the ionization energy determined from the weighted nonlinear least-squares fit. The quantum defect expansion and Stark constants determined in the fit are given in Table I, and the residuals of the fit are shown in Fig. 1. The weights used for fitting were proportional to the inverse square of the individual uncertainties shown in the figure. These uncertainties were determined by measuring the absolute wave number of each transition approximately 10 times and adding the standard deviation in quadrature to an estimated systematic error of 0.000015 cm^{-1} due to the random error in realigning the laser beam perpendicular to the atomic beam for the measurements at each n.

Inclusion of the term with n^7 dependence in the Ritz formula accounts very well for the small Stark shifts due to the constant residual field inside the excitation sphere. Without this term no satisfactory fit to the full data set is possible. The fitted coefficient of the Stark term is well defined and corresponds to a shift of 29.3 MHz at $n = 74$. Evidence that the Stark shift was accurately described is provided by the fact that the ionization energy derived from fits restricted to data at low n, where the Stark shift becomes insignificant, are consistent with the results of the fit to the full data set (Fig. 2).

Two important systematic corrections to the data were made prior to fitting the Ritz formula. First, the raw data were reduced by 0.000014-0.000017 cm^{-1} to correct for the rather large photon recoil shift for this light atom. Second, the results were further reduced by 2 parts in 10^{10} (about 0.000006 cm^{-1}) to correct for variation in the iodine cell temperature [7] and intracavity power [8] of the reference laser from the recommended values. In the process of searching for other systematic effects we varied the laser polarization from circular to plane and observed no shift in the measured line centers, leading us to conclude that Zeeman shifts under the normal operating conditions of $> 98\%$ plane polarized laser light are entirely negligible. We believe that there are no uncorrected systematic errors large enough to bias the measurements at
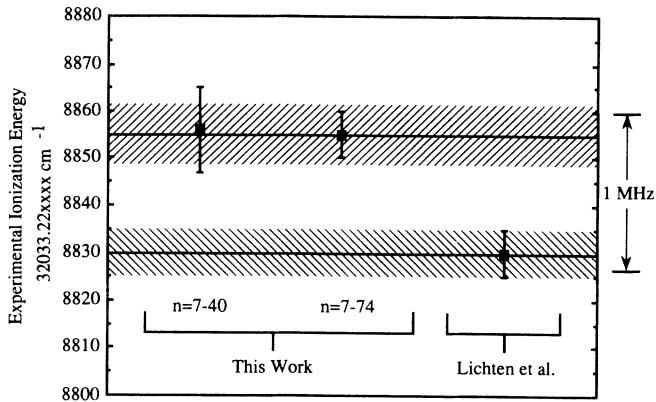
FIG. 2. Comparison of the 2 ^1S ionization energy determined in this work to that reported by Lichten, Shiner, and Zhou (Ref. [2]). Also shown is the good agreement between the ionization energies derived from our full data set ($n=7$–74) and that derived from levels with $n<40$ only for which residual Stark shifts are negligible. For results of the present work the error bars represent the statistical uncertainty of the Ritz fit while the shaded area represents the total one-standard-deviation uncertainty of the adopted ionization energy.

the final level of accuracy quoted.

The only significant systematic contribution to the uncertainty of our final result stems from the uncertainty in the frequency of the iodine stabilized reference laser. We use a laser constructed according to the same design as that calibrated in a direct frequency chain measurement by Jennings et al. [7] and adopt their result for the laser frequency. The uncertainty for this laser is 0.0000025 cm^{-1}. Additional uncertainties due to our corrections for deviations from the recommended operating parameters for the laser are insignificant. Combining this reference laser frequency uncertainty in quadrature with the statistical uncertainty of the fit we arrive at 32033.228855(7) cm^{-1} as the binding energy of the helium 2 ^1S level. This can be compared to the best available theoretical prediction which is 32033.231975 cm^{-1} (Ref. [9]). The results of the previous (semiexperimental) determinations were 32033.22868(18) cm^{-1} (Ref. [1]) and 32033.228830(5) cm^{-1} (Ref. [2]).

In principle, the full uncertainty in the frequency of the reference laser need not enter into the comparison of the experimental 2 ^1S binding energy with theory. The iodine stabilized helium-neon laser serves both as a realization of the Système International (SI) units in the visible region and as a convenient transfer standard that is common to

many laboratories. Its uncertainty as a realization of the meter enters only when an experimental result is reported in SI units; it places no intrinsic limitation on the accuracy with which the laser can be used as a transfer standard. If one converts the result of our experiment to atomic units (the natural units in which the theoretical values are calculated) by dividing by the Rydberg constant in SI units, the uncertainty due to the reference laser is partially eliminated since the Rydberg constant was measured with respect to the same standard. In these units only the lack of consistency in the frequency of reference lasers used in different laboratories makes a fundamental contribution to the experimental uncertainty; the degree to which the absolute frequency of the reference laser is uncertain makes no contribution. It is also advantageous from the point of view of theory to make the comparison in atomic units. Converting the theoretical results to SI units involves multiplication by the measured value of the Rydberg constant, a procedure that unnecessarily introduces an experimental uncertainty in the expression of the theoretical result. Unfortunately, in practice, the consistency (resetability) of the reference laser frequency is a major portion of its total uncertainty (47 kHz in a total uncertainty of 74 kHz) [7] and only a minor improvement in the comparison with theory can be realized by working in atomic units.

In conclusion, we have made a precise determination of the 2 ^1S ionization energy in helium that is fully independent of theoretical calculations. Our result confirms our earlier finding [1] of a substantial discrepancy between experiment and theoretical prediction [9] for the two-electron radiative corrections to the 2 ^1S level. Our determination falls within the error bars of our previous result, but is a factor of 25 more precise. Based on this result, the best reported calculation of the 2 ^1S binding energy [9] predicts a level which is too tightly bound by 0.003120 cm^{-1}, presumably because the calculated Lamb shift is too small.

Our value differs by only 8 parts in 10^{10} from that determined independently [2] in an experiment based on widely different assumptions and experimental methods. The 750-kHz disagreement, however, is 3 times larger than the reported one-standard-deviation uncertainties combined in quadrature. Although it is conceivable that undetected systematic errors in one or both experiments are responsible for the discrepancy, the possibility of a small error in the theoretical values of the 7–10 ^1D levels used by Lichten, Shiner, and Zhou [2] to determine their ionization energy cannot be ruled out.

[1] C. J. Sansonetti, J. D. Gillaspy, and C.L. Cromer, Phys. Rev. Lett. 65, 2539 (1990).

[2] W. Lichten, D. Shiner, and Zhi-Xiang Zhou, Phys. Rev. A 43, 1663 (1991).

[3] D. Hartree, Proc. Cambridge Philos. Soc. 24, 426 (1927).

[4] K.-H. Weber and C. J. Sansonetti, Phys. Rev. A 35, 4650 (1987).

[5] C. J. Sansonetti, in Advances in Laser Science—IV, edited by J. L. Gole, D. F. Heller, M. Lapp, and W. C. Stwalley, AIP Conf. Proc. No. 191 (AIP, New York, 1988), pp. 548–553.

[6] H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One- and Two-Electron Atoms (Plenum, New York, 1977).

[7] D. A. Jennings, C. R. Pollock, F. R. Petersen, R. E. Drullinger, K. M. Evenson, J. S. Wells, J. L. Hall, and H. P. Layer, Opt. Lett. 8, 136 (1983).

[8] H. P. Layer, W. R. C. Rowley, and B. R. Marx, Opt. Lett. 4, 188 (1981).

[9] G. W.F. Drake (private communication). Theoretical results are quoted in Ref. [1].