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New Measurement of the Rydberg Constant Using Polarization Spectroscopy of H_α

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We have determined a new Rydberg value, $R_\infty = 109\,737.314\,76(32)$ cm^{-1}, from an absolute wavelength measurement of the $2S_{1/2}$-$3P_{1/2}$ component of the hydrogen Balmer-α line. Polarization spectroscopy of a mild helium-hydrogen discharge with a cw dye laser provided a threefold higher accuracy than the only previous Doppler-free measurement. Another twofold improvement can be obtained by a more precise wavelength measurement of a near-coincident $^{127}I_2$ reference line.

The Rydberg constant provides the scale for the energy levels of one-electron atoms via Bohr's formula, corrected for finite nuclear mass, Dirac fine structure, and Lamb shifts. We have determined a new value of the Rydberg constant, $R_\infty = 109\,737.314\,76(32)$ cm^{-1}, by measuring the absolute wavelength of the D_1 ($2S_{1/2}$-$3P_{1/2}$) fine-structure component of the hydrogen Balmer-α line in a mild helium-hydrogen dc discharge, using sensitive polarization spectroscopy[1] with a cw dye laser. Our result is three times more accurate than the previous value, $R_\infty = 109\,737.3143(10)$ cm^{-1}, which was obtained several years ago by measuring the wavelength of the broader $2P_{3/2}$-$3D_{5/2}$ fine-structure component of the Balmer-α line in a Wood's discharge, using saturated-absorption spectroscopy with a pulsed dye laser.[2]

To obtain the wavelength of the H_α D_1 component, the absolute wavelength of a near-coincident $^{127}I_2$ reference line has been determined by comparing it with a previously measured $^{127}I_2$ line[3] at 632.8 nm. Next the separation between the D_1 component and the iodine reference line has been measured. Finally, systematic effects due to discharge conditions have been carefully investigated and corrected for. A more precise wavelength measurement of the reference line H_α could immediately provide at least another twofold improvement in the accuracy of the Rydberg constant.

The excited $n = 2$ hydrogen atoms are produced in a 5.5-cm-diam hot-cathode dc discharge tube, filled with a mixture of 15% H_2 in He. Even though the Pyrex walls are left uncoated, the hydrogen near the middle of the tube is completely dissociated at currents above 50 mA. The tube can operate down to lower pressures than the previously used Wood's tube and needs about three times smaller current densities for the same polarization signal. The axial electric field in the positive column, as measured with two probe electrodes 14 cm apart, is almost an order of magnitude smaller than in the Wood's tube, ranging from 0.9 V/cm at 0.015 Torr to 4.7 V/cm at 1.5 Torr. Radial fields remain below 0.3 V/cm within a radius of 1 cm from the tube axis. The gas pressure is measured with a McLeod gauge, and the temperature is monitored with a thermocouple. We find almost no interdependence between pressure and current in the range 50–500 mA, and thus line shifts and broadening due to the buffer-gas density and induced by the microscopic and macroscopic electric fields in the discharge can be measured separately.

The optical scheme of the polarization spectrometer used to observe the H_α line is similar to that described by Weber and Goldsmith.[4] A single-mode cw dye laser (Coherent Inc. Model 599, Rhodamine 101 dye) pumped by the 568.2-nm line of a krypton ion laser (Spectra Physics Model 171) provides up to 50 mW near 656 nm with a bandwidth of about 1 MHz. Part of the laser output is divided into a probe beam (0.5 mW) and a saturating beam (15 mW) which are sent in nearly opposite directions through a 55-cm-long section of the positive column of the discharge. The beams cross at an angle of 1–5 mrad, with a typical absorption near resonance of 40%. Telescopes expand the beams to 7 mm diam in order to avoid light shifts.

The probe beam monitors the absorption region placed between two linear polarizers, rotated from the perfectly crossed position by about 2 deg. Less than 0.05% of the probe light reaches the photomultiplier in this way, and noise due to laser intensity fluctuations is greatly reduced. The circularly polarized saturating beam produces a small gyrotropic birefringence in the gas sample which can rotate the polarization of the probe. The light flux reaching the detector is a very sensitive indicator for anu such change in polarization. A dispersion-shaped resonance signal is observed at the center of a Doppler-broadened line, where both beams are interacting with the same atoms. A small asymmetry is produced by an additional Lorentzian signal due to light-induced dichroism. This asymmetry can easily be accounted for by comparing spectra with the analyzing polarizer uncrossed by an equal amount in opposite directions. The derivative of the dispersion-shaped resonances is recorded by frequency modulating the dye laser at 2.3 kHz with a modulation depth of about ± 8 MHz and using lockin detection. Bell-shaped resonances of typical width 35–40 MHz are obtained in this manner, with the linewidth reduced by a factor of 2 compared to the dispersion-shaped resonances.

The setup for observing the iodine lines used as wavelength references is very similar, but without the beam expanding telescopes, and with a smaller frequency modulation applied to the dye laser (typical modulation depth ± 2 MHz). An evacuated and temperature-stabilized 1-m confocal interferometer provides frequency markers for the calibration of the laser scan. Aluminum-coated laser-quality mirrors (83% reflecting, 2% transmitting) yield a measured finesse of 8 with negligible dispersion in their reflective phase shift. The frequency markers shift less than 1 MHz/h as a result of residual thermal drift.

A typical polarization spectrum of H_α is shown in Fig. 1(a). The dye laser was scanned at a rate of 15 MHz/sec, with the integrating time constant of the lockin amplifier set at 0.1 sec. The previous Doppler-free measurement of the Rydberg constant used the strong $2P_{3/2}$-$3D_{5/2}$ fine-structure component on the left-hand side of Fig. 1(a). Our improvement in signal-to-noise ratio makes it practical to use the D_1 ($2S_{1/2}$-$3P_{1/2}$) component instead, which has a natural linewidth (29 MHz) more than a factor of 3 smaller than the P-D component, and a less complicated hyperfine structure. Since the $3P_{1/2}$ hyperfine splitting of 17.55 MHz is not resolved, the minimum D_1 linewidth is about 50 MHz. An expanded spectrum of the D_1 component, recorded with a scan speed of about 5 MHz/sec and a 0.1-sec time constant, is shown in Fig. 1(b). The use of a circularly polarized saturating beam suppresses transitions from the $F=0$ hyperfine level of the $2S_{1/2}$ state, since atoms with zero angular momentum cannot be oriented. The small signals observed from the $2S_{1/2}(F=0)$-$3P_{1/2}$ transition, and its crossover with the $2S_{1/2}(F=1)$-$3P_{1/2}$ component, are due to a slight optical orientation of the short-lived $3P_{1/2}$ state.

The ith hyperfine component of the ${}^{127}I_2$ B-X $R(73)$ 5-5 transition[5] is chosen to serve as a

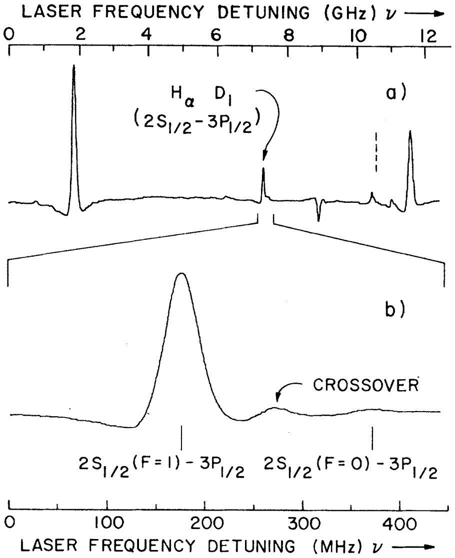

FIG. 1. (a) Polarization spectrum of H_α. The dashed line indicates the position of the iodine reference line. (b) Polarization spectrum of the H_α D_1 ($2S_{1/2}$-$3P_{1/2}$) fine-structure component.

wavelength reference near H_α. Its position relative to the H_α spectrum is indicated by the dashed line near the right-hand side of Fig. 1(a). The wavelength of this line is determined relative to the *i*th hyperfine component of the $^{127}I_2$ B-X $R(127)$ 11-5 transition at 632.8 nm. The value 15 798.002 948(22) cm^{-1} is adopted for the absolute wave number of this basic reference line,[3] measured relative to the ^{86}Kr wavelength standard. The *i*th hyperfine component of the $^{127}I_2$ B-X $R(73)$ 5-5 transition at 656.3 nm lies 564.635 558(35) cm^{-1} to smaller wave number than the 632.8-nm basic reference line, yielding an absolute wave number of 15 233.367 390(42) cm^{-1} for the former.[6]

The separation between the 656.3-nm reference line and the H_α D_1 line is measured using the second polarization spectrometer, sharing a common dye laser, with the laser scan extended to cover both lines. Operating the discharge at the conditions given in Table I, and averaging over sixteen spectra taken on two days, we find a frequency separation of 3368.04(30) MHz, with the iodine line at higher frequency. The uncertainty includes the measured standard deviation of the mean of 0.16 MHz, conservatively increased to 0.30 MHz to allow for possible uncompensated asymmetries in the observed line shape.

The systematic effects due to pressure and discharge conditions have been investigated very carefully by evaluating some 500 individual spectra, and will be discussed elsewhere in detail.[7] The systematic corrections applied to the measured frequency separation between the D_1 line and the iodine reference line are summarized in Table I. The large error in the current correction takes into account an uncertainty in the low-current threshold behavior of the discharge. The $2S_{1/2}$ hyperfine correction is applied because the ground state of the observed D_1 transition is the $2S_{1/2}(F=1)$ hyperfine state, rather than the center of gravity of the $2S_{1/2}$ state. No correction is necessary for the $3P_{1/2}$ state because of collisional decoupling of the hyperfine structure. No other experimental parameters (e.g., laser power, frequency-modulation depth, polarizer uncrossing angle) are found to have measurable ($\gtrsim 0.1$ MHz) effects. When we apply the corrections, the frequency separation between the center of gravity of the H_α D_1 ($2S_{1/2}$-$3P_{1/2}$) line and the iodine reference line becomes 3318.80(50) MHz.

Combined with our value for the absolute wavelength of the iodine reference line, the above frequency separation yields the value 15 233.256 686(45) cm^{-1} for the absolute wave number of the center of gravity of the H_α D_1 component. The Rydberg constant can now be determined by fitting this measured wave number with a calculated wave number, which is proportional to an assumed Rydberg value. Using recent calculations by Erickson,[8] we arrive at the value $R_\infty = 109\,737.314\,76(32)$ cm^{-1} for the Rydberg constant, which is consistent with and three times more accurate than the only previous Doppler-free Rydberg measurement. Since the ^{86}Kr wave-

TABLE I. Measured wave numbers (frequencies) and systematic corrections used in determining the Rydberg constant.

656.3-nm iodine reference line[a]	15 233.367 390(42) cm^{-1} [456 684 866.6(1.0) MHz]
Measured H_α D_1–iodine separation	– 3368.04(30) MHz
Systematic corrections:	
Discharge current (180 ± 20 mA)	+2.92(33) MHz
Pressure [He-H_2(15%), 0.130 Torr]	+1.26(20) MHz
Electric field (2.51 V/cm)	+0.65(10) MHz
$2S_{1/2}$ hyperfine splitting	+44.41(00) MHz
Total	+49.23(40) MHz
Corrected H_α D_1–iodine separation	– 3318.81(50) MHz
H_α D_1 wavelength (frequency)	15 233.256 686(45) cm^{-1} [456 681 547.8(1.1) MHz]
Calculated Rydberg constant	109 737.314 76(32) cm^{-1} [3 289 841 941.8(7.9) MHz]

[a] The $^{127}I_2$ B-X $R(73)$ 5-5 line, *i*th hyperfine component, measured relative to the ^{127}I B-X $R(127)$ 11-5 line, *i*th hyperfine component, $\bar{\nu} = 15\,798.002\,948(22)$ cm^{-1} (Ref. 3), $\nu = 473\,612\,214.8(0.3)$ MHz (Ref. 9).

length standard is inherently less accurate than the precision achievable for the reference lines used, these and the values for H_α D_1 and for R_∞ are also given in frequency units (Table I), determined relative to the frequency of the 632.8-nm reference line,[9] $\nu(^{127}I_2\ B\text{-}X\ R(127)$ 11-5, ith hyperfine component$) = 473.612\,214\,8(3)$ THz. This frequency has been measured relative to the Cs frequency standard. The frequency data in Table I can easily be readjusted for any redefinition in the standard system, in particular, for a unified system based on a defined velocity of light. The quoted error in R_∞ is due almost entirely to uncertainties in the wavelength measurement of the iodine reference line at 656.3 nm, and not introduced by corrections for the discharge conditions. Using our value for the frequency separation of the H_α D_1 line and the iodine line at 656.3 nm, a still more precise value for the Rydberg constant could be obtained immediately from a more precise measurement of the absolute wavelength (frequency) of the iodine reference line. Such a measurement could yield a value of the Rydberg constant accurate to 1.1 parts in 10^9, making the Rydberg the most precisely measured of all fundamental constants.

For future measurements of hydrogen wavelengths at the level of accuracy of one part in 10^9, it would be desirable to replace the discharge tube with a beam of metastable $2S$ hydrogen atoms, so that systematic corrections due to discharge conditions would not be necessary. The resolution obtainable with polarization or saturation spectroscopy, however, would still be limited by the short lifetime of the P state involved. One alternative would be to use the method of double-quantum saturation spectroscopy; we observed linewidths (without derivative narrowing) of less than 20 MHz for the $2S_{1/2}$-$3P_{1/2}$-$3S_{1/2}$ transition in the helium-hydrogen discharge.[4] Observed in a beam, the $2S_{1/2}$-$3S_{1/2}$ transition could be 30 times narrower than the intermediate $2S_{1/2}$-$3P_{1/2}$ transition. Still higher resolution could in principle be obtained by Doppler-free two-photon spectroscopy of the hydrogen $1S$-$2S$ transition, where the ultimate linewidth is limited only by the $\frac{1}{7}$-sec natural lifetime[10] of the $2S$ state.

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[1] C. Wieman and T. W. Hänsch, Phys. Rev. Lett. 36, 1170 (1976).

[2] T. W. Hänsch, M. H. Nayfeh, S. A. Lee, S. M. Curry, and I. S. Shahin, Phys. Rev. Lett. 32, 1336 (1974).

[3] W. G. Schweitzer, E. G. Kessler, R. D. Deslattes, H. P. Layer, and J. R. Whetstone, Appl. Opt. 12, 2927 (1973).

[4] E. W. Weber and J. E. M. Goldsmith, Phys. Rev. Lett. 41, 940 (1978).

[5] We follow the notation of G. R. Hanes and C. E. Dahlstrom, Appl. Phys. Lett. 14, 362 (1969). The iodine hyperfine components are labeled alphabetically in order of decreasing frequency.

[6] J. E. M. Goldsmith, E. W. Weber, F. V. Kowalski, and A. L. Schawlow, to be published.

[7] E. W. Weber and J. E. M. Goldsmith, to be published. The collisional decoupling and narrowing effect is analogous to that observed for nuclear magnetic resonance of molecules in gases, discussed by A. Abragam, *The Principle of Nuclear Magnetism* (Clarendon, Oxford, 1961), Chap. X.

[8] G. W. Erickson, J. Phys. Chem. Ref. Data 6, 831 (1977).

[9] H. P. Layer, R. D. Deslattes, and W. G. Schweitzer, Jr., Appl. Opt. 15, 734 (1976), combined with the frequency difference $\nu[^{129}I_2(k)] - \nu[^{127}I_2(i)] = 96.0399(98)$ MHz from Ref. 3.

[10] See, for example, H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One and Two Electron Atoms* (Springer-Verlag, New York, 1957), p. 285.