

Wavelength calibration of the C I line at 94.5 nm for comparison with quasar data

I. Labazan,* E. Reinhold, and W. Ubachs†

Laser Centre Vrije Universiteit, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands

V. V. Flambaum

School of Physics, The University of New South Wales, UNSW Sydney NSW 2052, Australia

(Received 26 January 2005; published 13 April 2005)

With the use of an ultra-narrow-band extreme ultraviolet laser source, tunable near 94 nm, transition wavelengths are determined for lines connecting the $1s^2 2s^2 2p^2 \ ^3P_{0,1,2}$ ground-term levels to the $1s^2 2s 2p^3 \ ^3S_1$ excited level in neutral carbon at an absolute accuracy of 4×10^{-8} . With the determination of the zero-velocity rest-frame wavelengths these lines can be included in an analysis of a possible temporal variation of the fine-structure constant α from a comparison with quasar data. A value for the C^{12}/C^{13} transition isotope shift was also obtained yielding $0.5107(13) \text{ cm}^{-1}$, in average over the three fine-structure lines. The latter measurement will allow to study isotopic evolution in the universe and test models of nuclear processes in stars.

DOI: 10.1103/PhysRevA.71.040501

PACS number(s): 32.10.-f, 33.20.Ni, 98.54.Aj

Theories unifying gravity with other interactions suggest a possibility of temporal and spatial variation of major constants of physics. A review of these theories as well as measurement results can be found in [1]. A very sensitive many-multiplet (MM) method to search for the variation of the fine-structure constant $\alpha = e^2/4\pi\epsilon_0\hbar c$ by comparison of quasar absorption spectra with laboratory spectra has been suggested in Ref. [2]. Webb *et al.* [3–6] used the MM method and found statistical evidence of α variation, while other groups [7,8] have used the same method [2] but found no evidence of α variation (note, however, that the authors of Refs. [3–6] used data from the Keck telescope located in the northern hemisphere while the authors of Refs. [7,8] used data from the southern hemisphere).

The MM method requires first-principles atomic-structure calculations of relativistic corrections to level energies, which allows one to find a dependence of atomic transition frequencies on α : $\omega = \omega_0 + qx$ where $x = \alpha^2/\alpha_0^2 - 1$. Here ω_0 and α_0 are the laboratory values, while ω and α refer to the rest values of transition frequency and fine-structure constant for an atom or ion in a remote cloud located at a distance up to 12 billion light years from us. Coefficients q are small in light atoms (anchor lines which are not sensitive to a variation of α), large positive (positive shifters), or large negative (negative shifters). To detect a variation of α and control systematic effects (which do not “know” about sign and magnitude of q) one should have representatives of all three classes (anchors, positive shifters, and negative shifters) in each absorption system. An example of an anchor line is Si II 152.671 nm, a positive shifter Zn II 206.614 nm, and a negative shifter Cr II 206.224 nm. The q value for the C I line at 94.5 nm has been calculated at 130 (60) [9] and therewith falls in the class of anchor lines.

Calculations of q for many atoms and for a large number of transitions have been performed in Refs. [9–11]. However, only 23 transitions have been used up to now (about 6–9 lines of each class). There exists a much larger number of observed spectral lines in absorption clouds. However, they cannot be used because of the absence of accurate laboratory measurements [12]. An increase of the number of useful lines is important since it allows to extend measurements of α variation to new absorption clouds located in different positions in space-time, to significantly increase statistics, and to provide efficient control of systematic errors (especially when a wide variety of q values are covered in each absorption cloud).

It is also important to measure the isotopic shifts in the spectral lines. The isotopic abundance ratios in the distant gas clouds may not match those on earth. If the isotopic abundances are very different this may generate spectral line shifts, which could mimic variations of α . To estimate this systematic effect one has to measure the isotopic shifts. Knowledge of isotopic shifts also allows to study another important problem: isotopic evolution in the universe. This provides a very sensitive test of models of nuclear processes in stars (see, e.g., Refs. [6,10,13,14]).

Here we report on a highly accurate measurement in neutral atomic carbon of the transition wavelengths of all three fine-structure components connecting the $^3P_{0,1,2}$ ground state with the 3S_1 level of the $2s2p^3$ configuration. For the measurements, use is made of a laser-based narrow band and tunable source of extreme ultraviolet (XUV) radiation developed in the Amsterdam Laser Centre [15]. The ground-state fine-structure splittings for both ^{12}C and ^{13}C are extremely accurately known from far-infrared spectroscopy [16,17], such that measurement of all three components in fact produces redundant information, therewith providing a consistency check on the wavelength calibrations.

The experimental setup of the laser-based XUV source, its application to high-resolution atomic and molecular spectroscopic studies, and the frequency calibration techniques, have been described in detail before [15]. A collimated XUV beam, generated via third-harmonic generation of the output

*Permanent address: Institute of Physics, PO Box 304, HR-10000 Zagreb, Croatia.

†Electronic address: wimu@nat.vu.nl (URL: <http://www.nat.vu.nl/~atom>)

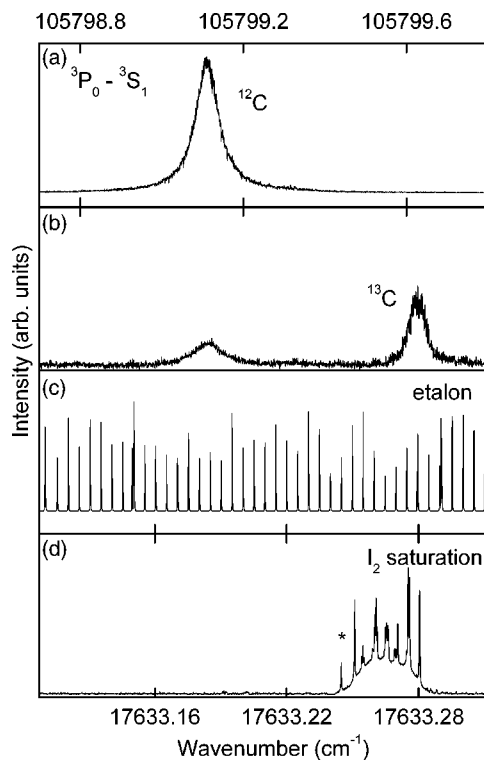


FIG. 1. Recording of the 3P_0 - 3S_1 spectral line of C I. (a) ${}^{12}\text{C}$; (b) ${}^{13}\text{C}$; (c) Etalon trace; (d) I_2 saturation spectrum with the “ t ”-hyperfine component of the B - $X(18,0)$ $R(82)$ line marked with (*); this component has a frequency $17\,633.2446\text{ cm}^{-1}$ in the visible range. Transition frequencies are displayed along the upper axis (XUV range) and the lower axis (visible frequency before conversion).

of a Fourier-transform-limited and frequency doubled pulsed laser system is crossed perpendicularly with a skimmed atomic beam. A feature is the use of a pulsed nozzle beam source, based on a Jordan Valve adapted with a pulsed discharge section, similar to the design of van Beek and ter Meulen [18]. Atomic carbon is produced in the beam by pulsing and discharging a 1% $\text{C}_2\text{H}_2/\text{He}$ mixture. The resulting beam is thereafter skimmed before entering the interaction zone. Signal is recorded by inducing $1\text{XUV}+1\text{UV}$ photoionization and subsequent detection of ions, where the UV is obtained from the UV laser beam, at $3\lambda_{\text{XUV}}$, that is used for harmonic conversion.

In Fig. 1 an example of a recording of the 3P_0 - 3S_1 spectral line is shown. The two upper spectra are recordings of incoming ions after a time-of-flight mass separation zone, with two boxcar gates set to record masses 12 and 13. The ${}^{13}\text{C}$ trace is hence obtained from the naturally abundant ${}^{13}\text{C}$ isotope (1%) in the beam. The spectral peak occurring at the ${}^{12}\text{C}$ resonance in the ${}^{13}\text{C}$ trace is an artefact, due to leaking of part of the strong signal into the time window set for mass 13.

Spectral calibration of the wavelength is derived from the simultaneously recorded transmission fringes from an actively stabilized etalon (free spectral range (FSR) 148.96 MHz) and a saturation spectrum of iodine. In the latter spectrum the “ t ”-hyperfine component, which is used for absolute

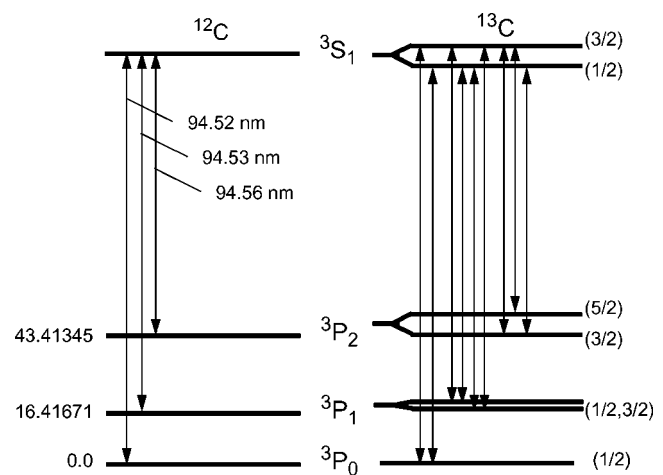


FIG. 2. Level scheme of the 94.5-nm spectral line in neutral carbon. The fine-structure splittings in the 3P ground state are accurately known from far-infrared studies [16,17]. The numbers on the left-hand side give energies of 3P levels in cm^{-1} . In ${}^{13}\text{C}$ the 3P_2 and 3P_1 levels are split twofold by the $I=1/2$ nuclear spin with splittings 372.4 and 4.1 MHz, respectively. The electronically excited 3S_1 level in ${}^{13}\text{C}$ is also split by the hyperfine interaction for which the value is unknown. For ${}^{13}\text{C}$ the spectral hyperfine components are drawn, which are, however, unresolved in the present work.

frequency calibration, is marked in the lower trace of Fig. 1. Note that these spectra were recorded with the continuous-wave output of a seed-laser running at $6\lambda_{\text{XUV}}$ [15]. The reference frequencies of the I_2 lines were obtained from the analysis by Knöckel and co-workers at an accuracy better than 5 MHz for all “ t ” components in the entire visible and near-infrared range [19]. Analysis of the uncertainty budget reveals that two systematic effects give decisive contributions to the error budget. The first is a possible Doppler shift due to a nonperfect perpendicular alignment of the atomic beam with respect to the laser beam. The shift is addressed by measuring the C I lines from both $\text{C}_2\text{H}_2/\text{He}$ and $\text{C}_2\text{H}_2/\text{Xe}$, thereby changing the velocity of the beam by more than a factor of 2 for the same angle. Analysis yields an upper limit to a Doppler shift of 0.003 cm^{-1} . A second major contribution to the uncertainty is the so-called frequency chirp induced in the dye amplifiers; based on previous investigations [15] we conservatively estimate this to produce an uncertainty in the XUV frequency of smaller than 0.003 cm^{-1} . Combined with some smaller contributions to the uncertainty (FSR etalon, accuracy of I_2 -reference standard, statistical errors from the fitting of line profiles) a total uncertainty of 0.006 cm^{-1} or $0.000\,006\text{ nm}$ results. This corresponds to a relative accuracy of 6×10^{-8} .

The widths of the observed lines are on average 1450 MHz, which is predominantly associated with natural lifetime broadening. Although the 3S_1 level is above the ionization potential, selection rules forbid autoionization, at least within LS coupling. Two studies have focused on *ab initio* calculations of the radiative decay yielding $A=6.1 \times 10^9\text{ s}^{-1}$ [20] and a significantly smaller value of $A=3.41 \times 10^9\text{ s}^{-1}$ [21]. If the XUV bandwidth ($\approx 300\text{ MHz}$) and some residual Doppler broadening are subtracted from the observed width

TABLE I. Resulting wavelengths (in nm) for the $2s^22p^2$ ${}^3P_{0,1,2}-2s2p^3$ 3S_1 spectral lines in both ${}^{12}\text{C}$ and ${}^{13}\text{C}$. A comparison is made with the values from Johansson [22].

	$\lambda_{\text{measured}}$	Ref. [22]	$\lambda_{\text{analyzed}}$
${}^{12}\text{C}$			
${}^3P_{0-3}S_1$	94.518 751 (6)	94.519 1	94.518 752 (4)
${}^3P_{1-3}S_1$	94.533 423 (6)	94.533 8	94.533 421 (4)
${}^3P_{2-3}S_1$	94.557 551 (6)	94.557 9	94.557 552 (4)
${}^{13}\text{C}$			
${}^3P_{0-3}S_1$	94.518 293 (8)		94.518 291 (6)
${}^3P_{1-3}S_1$	94.532 955 (8)		94.532 959 (6)
${}^3P_{2-3}S_1$	94.557 093 (8)		94.557 091 (6)

a natural line broadening of ≈ 1 GHz results, which would be in accordance with the larger value from Ref. [20].

For ${}^{13}\text{C}$ the same linewidths are observed. In Fig. 2, a level scheme for the ${}^{13}\text{C}$ isotope is displayed, including the effect of hyperfine structure. Each fine-structure-resolved spectral line consists of two (3P_0), three (3P_2), or four (3P_1) hyperfine components, but no evidence of additional broadening or of significantly asymmetric line shapes is observed. From this fact it is concluded that the hyperfine splitting in the 3S_1 upper level does not exceed 400 MHz. The hyperfine structure is ignored in the frequency analysis, although for ${}^{13}\text{C}$ the uncertainty is somewhat increased to account for possible hyperfine effects.

In Table I the results on the wavelength calibrations for all three fine-structure lines for both isotopes are listed. A comparison is made with the findings from classical spectroscopy from Johansson [22]. A systematic discrepancy is found with the previous values being higher in wavelength by 0.0004 nm. In Ref. [22] no explicit uncertainty is specified, but we assume that a deviation of four units in the last digit displayed is reasonable. In order to avoid confusion in the use of the data we state explicitly that in our work, frequencies were measured that were subsequently converted into vacuum wavelengths.

All three spectral lines connect each of the three 3P levels to the single 3S_1 level, and hence the wavelength calibrations provide redundant information on its level energy. Since some contributions to the total error budget in determining the level energy of the 3S_1 level are statistical, and even the systematic effects may vary with wavelength, some additional averaging is appropriate. With the information from the infrared studies [16,17] the level energy for 3S_1 and its uncertainty is $105\,799.109(4)$ cm^{-1} for ${}^{12}\text{C}$. The values for the separations between fine-structure levels, as given in Fig. 2, are the same for ${}^{13}\text{C}$, when averaging over hyperfine levels

TABLE II. Resulting ${}^{12}\text{C}-{}^{13}\text{C}$ isotope shifts (IS) on the 94.5-nm C I transition.

	Isotope shift (MHz)	Isotope shift (cm^{-1})
${}^3P_{0-3}S_1$	15 435 (120)	0.514 4 (40)
${}^3P_{1-3}S_1$	15 480 (210)	0.516 0 (70)
${}^3P_{2-3}S_1$	15 295 (50)	0.509 8 (16)

and within an uncertainty of 0.001 cm^{-1} . The same procedure leads to a level energy of $105\,799.628(6)$ cm^{-1} for the 3S_1 level in ${}^{13}\text{C}$. These level energies, obtained via averaging over the three independent measurements, can be used to recalculate the transition wavelengths. The results are listed in the last column of Table I. This procedure yields an estimation of transition wavelengths with a relative uncertainty of 4×10^{-8} .

As for the isotope shifts continuous scans can be recorded covering the span of both ${}^{12}\text{C}$ and ${}^{13}\text{C}$ components. For these relative measurements the major systematic contributions to the uncertainties cancel out, resulting in improved values for the isotope shifts as displayed in Table II. Due to experimental constraints on the tunability of the laser system this procedure did not work well for the ${}^3P_{1-3}S_1$ line; hence its isotope shift was derived from the excited-state level energies. A statistical average over the three transition isotope shifts then yields a value of $0.5107(13)$ cm^{-1} for the multiplet. *Ab initio* calculations of isotope shifts have been performed in the past for electronic transitions involving the 3P ground state of C I [23,24], however not for the transition to the 3S_1 level.

In conclusion, transition wavelengths combining the three levels of the $1s^22s^22p^2$ ${}^3P_{0,1,2}$ ground term to the $1s^22s2p^3$ 3S_1 excited state have been determined with an accuracy of 4×10^{-8} . This accuracy can be considered exact in comparisons between laboratory data from the present epoch with data obtained from quasars at high redshifts. The C I line at 94.5 nm is therewith a spectral line that may be included for searches of temporal variation of the fine-structure constant. We also measured the isotopic shift between ${}^{12}\text{C}$ and ${}^{13}\text{C}$ lines. Knowledge of this isotopic shift allows one to measure the isotopic ratio ${}^{12}\text{C}/{}^{13}\text{C}$ in distant clouds and perform an important test of models of nuclear processes in stars [13,14].

The authors wish to thank E. Salumbides for his assistance during the measurements. The Space Research Organisation Netherlands (SRON) and the Netherlands Foundation for Fundamental Research on Matter (FOM) are gratefully acknowledged for financial support.

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