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

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(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 1s2s2p2 3P0,1,2 ground-term levels to the 1s2s2p3 3S1 excited level in neutral carbon at an absolute accuracy of 4 × 10−9. 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 C12/C13 transition isotope shift was also obtained yielding 0.5107(13) 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 α=e2/4πε0hc 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: ω=ω0+qαx where x=α2/α02−1. Here ω0 and α0 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 3P0,1,2 ground state with the 3S1 level of the 2s2p3 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 12C and 13C 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...
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 3×10$^9$ s$^{-1}$. 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 “$r$” 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 nm results. This corresponds to a relative accuracy of $6\times10^{-8}$.

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 $A=6.1\times10^9$ s$^{-1}$ [20] and a significantly smaller value of $A=3.41\times10^9$ s$^{-1}$ [21]. If the XUV bandwidth (=300 MHz) and some residual Doppler broadening are subtracted from the observed width.
TABLE I. Resulting wavelengths (in nm) for the $^{3}_1$ spectral lines in both $^{12}$C and $^{13}$C. A comparison is made with the values from Johansson [22].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\lambda_{\text{measured}}$</th>
<th>Ref. [22]</th>
<th>$\lambda_{\text{analyzed}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{12}$C</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{3}_1$</td>
<td>94.518 751 (6)</td>
<td>94.519 1</td>
<td>94.518 752 (4)</td>
</tr>
<tr>
<td>$^{3}_{0}$</td>
<td>94.518 751 (6)</td>
<td>94.519 1</td>
<td>94.518 752 (4)</td>
</tr>
<tr>
<td>$^{3}_{1}$</td>
<td>94.518 751 (6)</td>
<td>94.519 1</td>
<td>94.518 752 (4)</td>
</tr>
<tr>
<td>$^{13}$C</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{3}_1$</td>
<td>94.518 293 (8)</td>
<td>94.518 291 (6)</td>
<td>94.518 291 (6)</td>
</tr>
<tr>
<td>$^{3}_{0}$</td>
<td>94.518 293 (8)</td>
<td>94.518 291 (6)</td>
<td>94.518 291 (6)</td>
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<tr>
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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.
[19] The program for calculating absolute frequencies of hyperfine components in the $I_2$ saturation spectrum was kindly provided by Dr. Knöckel of the University of Hannover; see also, B. Bodermann, H. Knöckel, and E. Tiemann, Eur. Phys. J. D 19, 31 (2002).