

## Speed of Light from Direct Frequency and Wavelength Measurements of the Methane-Stabilized Laser

K. M. Evenson, J. S. Wells, F. R. Petersen, B. L. Danielson, and G. W. Day  
*Quantum Electronics Division, National Bureau of Standards, Boulder, Colorado 80302*

and

R. L. Barger\* and J. L. Hall†  
*National Bureau of Standards, Boulder, Colorado 80302*  
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The frequency and wavelength of the methane-stabilized laser at  $3.39\ \mu\text{m}$  were directly measured against the respective primary standards. With infrared frequency synthesis techniques, we obtain  $\nu = 88.376\ 181\ 627(50)$  THz. With frequency-controlled interferometry, we find  $\lambda = 3.392\ 231\ 376(12)$   $\mu\text{m}$ . Multiplication yields the speed of light  $c = 299\ 792\ 456.2(1.1)$  m/sec, in agreement with and 100 times less uncertain than the previously accepted value. The main limitation is asymmetry in the krypton  $6057\text{-}\text{\AA}$  line defining the meter.

The speed of light is one of the most interesting and important of the fundamental (dimensioned) constants of nature.<sup>1</sup> It enters naturally into ranging experiments, such as geophysical distance measurements which use modulated electromagnetic radiation, and astronomical measurements such as microwave planetary radar and laser lunar ranging. Basically, very high-accuracy measured delay times for electromagnetic waves are dimensionally converted to distance using the light propagation speed. Recent experiments have set very restrictive limits on any possible speed dependence on direction<sup>2</sup> or frequency.<sup>3</sup> Another interesting class of applications involves the speed of propagating waves in a less obvious manner. For example, the conversion between electrostatic and electromagnetic units involves the constant  $c$ , as does the relativistic relationship between the atomic mass scale and particle energies.

With the perfection of highly reproducible and stable lasers, their wavelength-frequency duality becomes of wider interest. We begin to think of these lasers as *frequency* references for certain kinds of problems such as optical heterodyne spectroscopy.<sup>4</sup> At the same time, we use the wavelength aspect of the radiation, for example, in precision long-path interferometry.<sup>5</sup>

It has been clear since the early days of lasers that this wavelength-frequency duality could form the basis of a powerful method to measure the speed of light. However, the laser's optical frequency was much too high for conventional frequency measurement methods. This fact led to the invention of a variety of modulation or differ-

ential schemes, basically conceived to preserve the small interferometric errors associated with the short optical wavelength, while utilizing microwave frequencies which were still readily manipulated and measured. These microwave frequencies were to be modulated onto the laser output or realized as a difference frequency<sup>6</sup> between two separate laser transitions. Indeed, a proposed major long-path interferometric experiment<sup>7</sup> based on the latter idea has been made obsolete by the high-precision direct frequency measurement<sup>8</sup> summarized in this Letter. An ingenious modulation scheme, generally applicable to any laser transition, has recently successfully produced an improved value for the speed of light.<sup>9</sup> While this method can undoubtedly be perfected further, its differential nature leads to limitations which are not operative in the present, direct method.

The product of the frequency and wavelength of an electromagnetic wave is the speed of propagation of that wave. For an accurate determination of both of these quantities, the source should be stable and monochromatic and should be at as short a wavelength as possible. At shorter optical wavelengths the accuracy of the wavelength measurement increases. A suitable source of such radiation is the methane-stabilized He-Ne laser<sup>10</sup> at  $3.39\ \mu\text{m}$  (88 THz). Direct frequency measurements were recently extended to this frequency<sup>11</sup> and subsequently refined<sup>8</sup> to the present accuracy of 6 parts in  $10^{10}$ . The wavelength of this stabilized laser has been compared<sup>12</sup> with the krypton-86 length standard to the limit of the usefulness of the length standard (approximately

3 parts in  $10^9$ ). The product of the measured frequency and the wavelength yields a new, definitive value for the speed of light,  $c$ . The previously accepted value<sup>13</sup> of  $c$  was similarly determined by measuring the frequency and wavelength of a stable electromagnetic oscillator; however, it oscillated at 72 GHz (more than 1000 times lower in frequency than in the case of the present measurements). The 100-fold improvement in the presently reported measurement comes mainly from the increased accuracy possible in the measurement<sup>12</sup> of the shorter wavelength.

With suitable point-contact mixer diodes, a chain of stabilized lasers and klystron frequency sources has allowed direct harmonic generation and frequency mixing from the National Bureau of Standards frequency standard upward to the CO<sub>2</sub> laser at 29 THz (10.3  $\mu\text{m}$ ) and thence upward to the methane-stabilized laser at 88 THz (3.39  $\mu\text{m}$ ). Five different types of laser and five klystrons were used in the three-step measurement process. An interpolating counter referenced to a cesium clock counted the X-band frequency at the base of the chain. The X-band klystron was phase locked to the 74-GHz klystron which was phase locked to the free-running HCN laser. A times-12 multiplication brought a harmonic of the HCN laser to within 29 GHz of the free-running H<sub>2</sub>O laser frequency (at 28  $\mu\text{m}$ ), and the tens of megahertz beat note produced in this diode was measured on a spectrum analyzer and counter. The H<sub>2</sub>O laser's third harmonic fell 19 GHz above the CO<sub>2</sub> R(10) laser at 9.3  $\mu\text{m}$ . The CO<sub>2</sub> laser was frequency stabilized to the central tuning dip (Lamb dip) of the saturated fluorescence in a low-pressure CO<sub>2</sub> absorption cell.<sup>14,15</sup> All of the above described beat notes were measured simultaneously in this first step of the experiment, yielding the frequency 32.134 266 891(24) THz for the R(10) line. The interval from this R(10) frequency to the R(30) line of CO<sub>2</sub> at 10.3  $\mu\text{m}$  was measured as the HCN laser's third harmonic +19.5 GHz. The resulting CO<sub>2</sub> R(30) frequency was 29.442 483 315(25) THz. The third harmonic of this laser's frequency falls 49 GHz short of the He-Ne laser (3.39  $\mu\text{m}$ ) stabilized to the saturated absorption peak in methane. The final methane-stabilized frequency [ $F_1^{(2)}$  component of  $P(7)$ ] was found to be 88.376 181 627(50) THz. The fractional uncertainties in the molecular frequencies of CO<sub>2</sub> are somewhat larger than that of methane due to larger possible offsets from true line centers of the CO<sub>2</sub> absorptions.

These offsets would not affect either the value or the uncertainty of the measured methane frequency. The (1 standard deviation) error estimates result from careful analysis<sup>8</sup> of both random and possible systematic effects. The dramatic accuracy improvement over previous infrared frequency measurements stems from the use of better microwave and laser frequency control and measurement electronics, improved mixer signal-to-noise ratios, and, most importantly, the use of molecular saturated-absorption stabilization of the measured infrared frequencies.

In a coordinated effort, the wavelength of the 3.39- $\mu\text{m}$  line of methane has been measured with respect to the Kr<sup>86</sup> 6057- $\text{\AA}$  primary standard of length. Using a frequency-controlled Fabry-Perot interferometer with a pointing precision of about  $2 \times 10^{-5}$  orders, we have made a detailed search for systematic offsets inherent in the experiment, including effects due to the asymmetry of the Kr standard line. Offsets due to various experimental effects (such as beam misalignments, mirror curvatures and phase shifts, phase shift over the exit aperture, diffraction, etc.) were carefully measured and then removed from the data with an uncertainty of about 2 parts in  $10^9$ . This reproducibility for a single wavelength measurement illustrates the high precision which is available using the frequency-controlled interferometer.

Unfortunately, after the Kr<sup>86</sup> transition at 6057  $\text{\AA}$  was adopted as the primary standard of length it was discovered that this line is slightly asymmetric,<sup>16</sup> resulting in a small shift of effective wavelength with the order of interference. For example, in our experiment<sup>12</sup> the apparent measured wavelength showed a fractional systematic dependence of  $\pm 1.1 \times 10^{-8}$  upon the mirror spacing, in basic agreement with other work.<sup>16,17</sup> Following Rowley and Hamon,<sup>16</sup> a two-component model of the krypton asymmetry was used to analyze our data. This model reduced the standard deviation of the twenty wavelength measurements from 6.4 to 2.7 parts in  $10^9$ , and shifted the point of fringe maximum intensity by 4.1 parts in  $10^9$ . The deviations were improved somewhat further and the average wavelength red-shifted by 1.2 parts in  $10^9$  when we also assumed a radial dependence<sup>18</sup> of the Doppler shift<sup>19</sup> across the capillary bore of the krypton standard lamp.

In view of the (small) intrinsic asymmetry of the Kr standard line, it is necessary to specify the point on the line profile to which the defined

wavelength (6057.802 105 Å) is applied. At present there is no universal convention for this choice. Thus if the defined value is applied to the maximum-intensity point of the Kr line, we find  $\lambda = 33\,922.314\,04$  Å; if the defined value is applied to the center of gravity of the Kr line,  $\lambda = 33\,922.313\,76$  Å. Detailed consideration<sup>20</sup> of random and known systematic effects, along with uncertainties in the krypton asymmetry model, leads to an estimated 68% confidence interval of  $\delta\lambda = \pm 1.2 \times 10^{-4}$  Å or  $\delta\lambda/\lambda = \pm 3.5 \times 10^{-9}$  for both of these results.

The methane wavelength has also been measured by Giacomo.<sup>21</sup> Although he does not state his reference-point convention, his quoted result (33 922.313 76 Å) is identical to ours for our case where the defined Kr wavelength is applied to the line center of gravity.

In the absence of an international agreement on this question of reference point for the krypton length definition, we feel it simplifies presentation of our numerical result if we adopt the arbitrary convention that the defined wavelength is to be applied to the center of gravity of the krypton line. With this choice, the values of the wavelength and frequency of the methane-stabilized He-Ne laser are

$$\lambda = 3.392\,231\,376(12) \mu\text{m} \quad (\delta\lambda/\lambda = \pm 3.5 \times 10^{-9})$$

and

$$\nu = 88.376\,181\,627(50) \text{ THz} \quad (\delta\nu/\nu = \pm 6 \times 10^{-10}).$$

Therefore,

$$c = 299\,792\,456.2(1.1) \text{ m/sec}$$

$$(\delta c/c = \pm 3.5 \times 10^{-9}).$$

The uncertainties quoted are 1-standard-deviation (68% reliance) estimates and include both random and residual systematic uncertainties. This result is in agreement with the previously accepted value of  $c = 299\,792\,500(100)$  m/sec and is about 100 times more accurate. As mentioned above, a recent differential measurement of the speed of light has been made by Bay, Luther, and White<sup>9</sup>; their value is  $299\,792\,462(18)$  m/sec, which is in agreement with the presently determined value. If the maximum-intensity point of the Kr line is chosen,<sup>22</sup> the methane wavelength, and hence the value of  $c$ , is increased by 8.3 parts in  $10^9$  [ $c_{\text{max}I} = 299\,792\,458.7(1.1)$  m/sec].

The fractional uncertainty in our value for the speed of light,  $\pm 3.5 \times 10^{-9}$ , essentially arises from the interferometric measurements with the

incoherent krypton radiation which operationally defines the international meter. This limitation is indicative of the remarkable growth in optical physics in recent years: The present krypton-based length definition was adopted only in 1960!

One view of this situation is that with lasers (and *great* care) it is possible to measure optical lengths more precisely than they may be operationally expressed in meters. Thus, one is easily led to consider choosing a suitable stabilized laser as a new basic standard of length. Both the methane-stabilized<sup>10,23</sup> He-Ne laser at  $3.39 \mu\text{m}$  (88 THz) and the  $I_2$ -stabilized<sup>24</sup> He-Ne laser at  $0.633 \mu\text{m}$  appear to be suitable candidates for the basic standard of length. They also can serve as secondary standards of frequency in the near-infrared and visible regions. The methane-stabilized He-Ne laser frequency is already known to 6 parts in  $10^{10}$ , and further measurements are expected to increase the accuracy to a few parts in  $10^{11}$  in the next year or two. A new value of the speed of light with this accuracy should thus be achievable if the standard of length were redefined.

Alternately, one can consider defining the meter as a specified fraction of the distance light travels in one second in vacuum (that is, one can define the speed of light). With this definition, the wavelength of stabilized lasers would be known to the same accuracy with which their frequencies can be measured. Stabilized lasers would thus provide accurate secondary standards of both frequency and length. It should be noted that an adopted nominal value for the speed of light is already in use for high-accuracy astronomical measurements.

Independent of which type of definition is chosen we believe that research on simplified frequency synthesis chains bridging the microwave-optical gap will be of great interest, as will refined experiments directed toward an understanding of the factors that limit laser optical frequency reproducibility. No matter how such research may turn out, it is clear that ultraprecise physical measurements made in the interim can be preserved through wavelength or frequency comparison with a suitable stabilized laser such as the  $3.39\text{-}\mu\text{m}$  methane device.

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\*Quantum Electronics Division.

†Laboratory Astrophysics Division, and Joint Institute for Laboratory Astrophysics (operated jointly by the National Bureau of Standards and University of Colorado).

<sup>1</sup>The interested reader will find a useful, critical discussion of the speed of light in D. D. Froome and L. Essen, *The Velocity of Light and Radio Waves* (Academic, New York, 1969).

<sup>2</sup>T. S. Jaseja, A. Javan, J. Murray, and C. H. Townes, *Phys. Rev.* **133**, A 1221 (1964), using infrared masers; D. C. Champeney, G. R. Isaak, and A. M. Khan, *Phys. Lett.* **7**, 241 (1963), using Mössbauer effect.

<sup>3</sup>B. Warner and R. E. Nather, *Nature (London)* **222**, 157 (1969), from dispersion in the light flash from pulsar NP 0532, obtain  $\Delta c/c \leq 5 \times 10^{-18}$  over the range  $\lambda = 0.25$  to  $0.55 \mu\text{m}$ .

<sup>4</sup>E. E. Uzgiris, J. L. Hall, and R. L. Barger, *Phys. Rev. Lett.* **26**, 289 (1971).

<sup>5</sup>J. Levine and J. L. Hall, *J. Geophys. Res.* **77**, 2595 (1972).

<sup>6</sup>J. L. Hall and W. W. Morey, *Appl. Phys. Lett.* **10**, 152 (1967).

<sup>7</sup>J. Hall, R. L. Barger, P. L. Bender, H. S. Boyne, J. E. Faller, and J. Ward, *Electron Technol.* **2**, 53 (1969).

<sup>8</sup>K. M. Evenson, J. S. Wells, F. R. Petersen, B. L. Danielson, and G. W. Day, to be published.

<sup>9</sup>Z. Bay, G. G. Luther, and J. A. White, *Phys. Rev. Lett.* **29**, 189 (1972).

<sup>10</sup>R. L. Barger and J. L. Hall, *Phys. Rev. Lett.* **22**, 4 (1969).

<sup>11</sup>K. M. Evenson, G. W. Day, J. S. Wells, and L. O. Mullen, *Appl. Phys. Lett.* **20**, 133 (1972).

<sup>12</sup>R. L. Barger and J. L. Hall, to be published.

<sup>13</sup>K. D. Froome, *Proc. Roy. Soc., Ser. A* **247**, 109 (1958).

<sup>14</sup>C. Freed and A. Javan, *Appl. Phys. Lett.* **17**, 53 (1970).

<sup>15</sup>F. R. Petersen and B. L. Danielson, to be published.

<sup>16</sup>W. R. C. Rowley and J. Hamon, *Rev. Opt., Theor. Instrum.* **42**, 519 (1963).

<sup>17</sup>This <sup>86</sup>Kr reproducibility limit is just larger than the  $1 \times 10^{-8}$  stated in *Comité Consultatif pour la Définition du Mètre, Rapport, 1970* (Bureau International des Poids et Mesures, Sèvres, France, 1972).

<sup>18</sup>A radial variation of the Doppler shift was postulated in 1963 by F. Bayer-Helms of the Physikalische-Technische Bundesanstalt (private communication to R.L.B. and J.L.H.).

<sup>19</sup>K. M. Baird and D. S. Smith, *Can. J. Phys.* **37**, 832 (1957). See also Ref. 14.

<sup>20</sup>For a more detailed discussion of asymmetry corrections, see R. L. Barger and J. L. Hall, to be published.

<sup>21</sup>P. Giacomo, in *Proceedings of Fourth International Conference on Atomic Masses and Fundamental Constants, Teddington, England, September 1971* (Plenum, New York, 1972).

<sup>22</sup>In the absence of an international agreement on this question, we slightly prefer the center-of-gravity definition since it probably would be less affected by lamp operating conditions (influence on Doppler width).

<sup>23</sup>J. L. Hall and R. L. Barger, in *Proceedings of the Symposium on Basic and Applied Laser Physics, Esfahan, 1971* (Wiley, New York, to be published).

<sup>24</sup>G. R. Haines and C. E. Dahlstrom, *Appl. Phys. Lett.* **14**, 362 (1969); G. R. Haines and K. M. Baird, *Metrologia* **5**, 32 (1969).