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

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Wavelength of the 3.39-μm laser-saturated absorption line of methane

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The wavelength of the 3.39-μm line of methane has been measured with respect to the Kr^{86} 6057-Å standard by using a frequency-controlled Fabry-Perot interferometer. We have exhaustively studied systematic offsets inherent in the experiment, including effects due to asymmetry of the Kr standard line. Lacking a convention relating the defined Kr wavelength 6057.802 105 Å to observables of the krypton line (e.g., center of gravity or fringe maximum intensity point), we report two methane wavelengths: $\lambda_{\max I} = 33\,922.314\,04$ Å and $\lambda_{cg} = 33\,922.313\,76$ Å. Both results have an uncertainty of $\delta\lambda = \pm 1.2\times 10^{-4}$ Å or $\delta\lambda/\lambda = \pm 3.5\times 10^{-9}$. Multiplication by the frequency measurement of the preceding letter gives the speed of light, $c_{cg} = 299\,792\,456.2(1.1)$ m/sec.

The laser-saturated absorption line of methane at 3.39 μm [the $P(7)$ line of the ν_3 band] has been shown to have remarkable characteristics desired of radiation which might be used as a standard of length (or frequency).[1] The Zeeman coefficient is small[2] ($+0.31\ \mu$N), and the transition is free from first-order Stark shift.[3] Linewidths as narrow as 25 kHz have been achieved. This, together with the signal-to-noise ratio of several thousand attainable with laser radiation, results in an achieved[4] frequency stability of a few parts in 10^{14}. Line center has been shown[1] reproducible to better than 1×10^{-11}. The selective-collision process in saturated absorption results in a pressure shift to broadening ratio of less than about 1:200 rather than the approximately 1:4 produced in van der Waals interactions. With a half-width half-maximum (HWHM) of 25 kHz, the pressure broadening (of methane by methane) is 10 kHz/mTorr, and the shift is ≤ 50 Hz/mTorr.[4]

These properties of the molecule and the saturation process, together with the laser's high intensity and spatial coherence, result in the 3.39-μm radiation having the most accurately measurable wavelength obtained to date. Thus it is of interest to measure the methane wavelength as accurately as possible in terms of the international standard of length, the meter, which is defined by the Kr^{86} orange line at 6057.802 105 Å. Long before a consensus emerges on the optimum technical route for redefinition of the meter, we believe the precision measurement community will find the methane transition an attractive and useful wavelength reference. One example of the new precision measurement possibilities is the extension of the range of direct frequency measurements to the 3.39-μm line.[5] This frequency value, taken with the presently reported wavelength determination, results in a definitive value for the speed of light as reported here and discussed elsewhere.[6]

The methane-stabilized laser used in this measurement had an intracavity methane absorption cell. The 10-mTorr pressure of methane in the cell together with the laser mode radius of 0.75 mm resulted in a saturation peak with an intensity of 3% of the laser power and a half-width half-maximum of 300 kHz (1×10^{-5} cm^{-1}, line Q of 1.4×10^{8}). Thus, the desired measurement accuracy of a few parts in 10^9 for $\Delta\nu/\nu$, which is the practical limit for the krypton standard, corresponds to a measurement of methane line center to only within the line half-width.

The Kr^{86} standard lamp was the type described by Englehard and Terrien.[7] The tube contained Kr^{86} with an isotopic purity of 99.8% quoted by the manufacturer. The discharge was run with a current density of 0.3 A/cm^2 with the tube immersed in a bath of liquid nitrogen held at the triple point. The observed light travels from cathode toward anode. The 6057-Å line emitted from a Kr lamp run under these conditions has a defined accuracy of one part in 10^8, but it is possible to measure a point on the line, and to hold the systematic errors associated with this point, to a few parts in 10^9 as has been done in our measurements. As a precaution against an unknown systematic error being associated with our lamp, we have compared the 6057-Å line from our lamp with that from a lamp loaned to us by another laboratory.[8] The two wavelengths were offset by 1 ± 2.5 parts in 10^9, well within the accuracy limit for these lamps. To make such precise measurements, it is essential to deal effectively with the small asymmetry of the krypton emission-line profile.

We have developed and used for the measurement reported here a new interferometry technique[9] wherein the interferometer length is servo controlled to be precisely a whole number of wavelengths of 3.39-μm laser radiation coming from a local-oscillator laser, which in turn is frequency-offset locked[1] from the methane-stabilized laser. The frequency of this radiation (and thus the interferometer length) is scanned in the desired manner with a stability nearly equal to that of the methane-stabilized laser. With this technique we have obtained reproducible fringe-pointing precisions of 2×10^{-5} order for the methane line, making it possible to accurately measure the various systematic effects inherent in interferometry.

The measurement system is indicated in Fig. 1. The interferometer was of the plane-parallel Fabry-Perot configuration. The length could be varied from about 1 mm, to give the large free spectral range necessitated by the initially poorly known methane wavelength ($\Delta\lambda/\lambda \approx 10^{-6}$), to a maximum of 25 cm to span the length

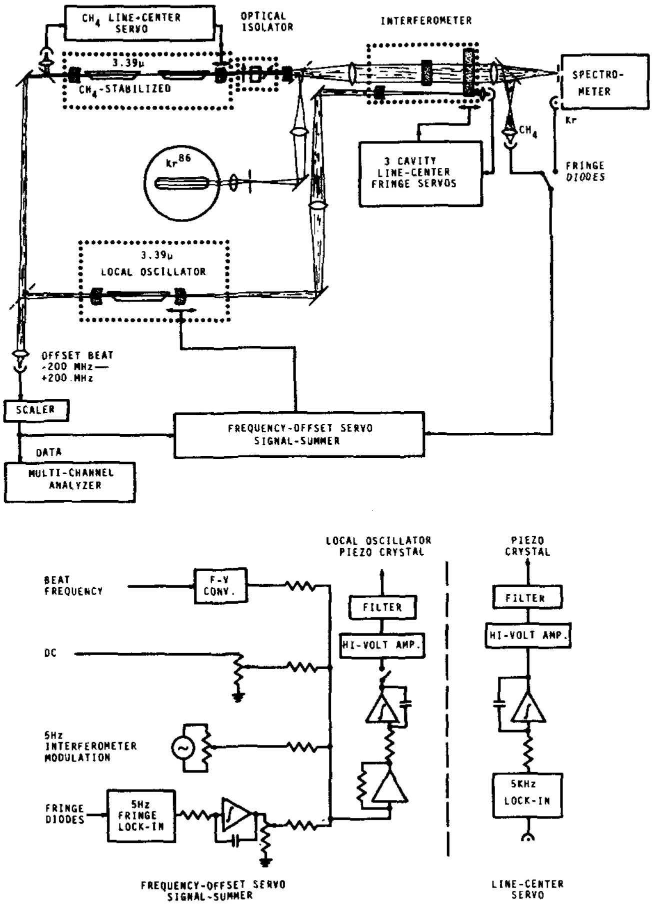

FIG. 1. Frequency-controlled interferometer measurement system.

(15–20 cm) giving optimum performance with the Kr line. For short cavities, the local-oscillator frequency could not be scanned over a full spectral range of the cavity. Therefore, the motion of the 2-in.-diam scanning flat was controlled with three 30-cm-long servo cavities formed between three spots near the edge of the flat and three small spherical mirrors spaced at 120° around the circumference of the Invar frame. The measurement cavity was formed between the 2-in.-diam flat and a $1\frac{1}{2}$-in.-diam flat which could be translated to the desired interferometer length and adjusted for parallelism inside the vacuum chamber. With the ±250-MHz tuning range of our local oscillator, this allowed the interferometer to be scanned over about one order for methane radiation and nearly six orders for visible radiation. The interferometer length was modulated at 5 Hz, and first-derivative detection was used for fringe pointing. Servo techniques used are indicated in Fig. 1.

Wavelengths have been calculated using the method of fractional fringes. Dispersion of the phase change on reflection was eliminated by subtracting data for two values of interferometer length to obtain data for a virtual spacer. We have used spacers of lengths 4.67, 11.20, 17.71, and 19.56 cm to give the five virtual spacers for which wavelengths have been calculated.

Various systematic offsets were contributed by some of the experimental techniques and by the asymmetry inherent in the Kr line. Effects related to techniques included those due to alignment of light beams not perfectly coaxial and normal to the interferometer, finite size of the pinhole at the Kr fringe pattern center, small plate figure effects, signal-to-noise limitations of Kr, and residual optical feedback effects in the laser case. By using the precision measurement capabilities of the frequency-controlled interferometer, these systematic effects have been carefully measured and corresponding corrections have been made to the wavelength measurement. The total fractional-wavelength uncertainty resulting from these was about 1.8×10^{-9}, with the largest contribution (about 1.5×10^{-9}) coming from misalignment of the Kr light beam (any misalignment gives a unidirectional wavelength shift, but the uncertainty is small enough that no correction has been applied).

Diffraction effects were measured for the 3.39-μm radiation by measuring the effective change ΔT of the interferometer length T as a function of T and interferometer aperture diameter D. Data were least-squares fit to the equation $\Delta T = c(\lambda/D)^2 T$ to determine the diffraction constant $c = 0.056$. For 3.39 μm and for a 1-cm aperture this corresponds to a wavelength correction $\Delta\lambda/\lambda$ of about 6×10^{-9} with an uncertainty of 1×10^{-9}. For comparison, ideal Gaussian modes apertured at both mirrors would give a theoretical value $c = 0.070$.[10]

With the considerations outlined above, we have removed from the data all known inherent systematic effects associated with the interferometric technique to an uncertainty of about 2×10^{-9}. However, the apparent 3.39-μm wavelength varied by $\pm1.1\times10^{-8}$ as a function of the spacer length. This spacer dependence of the effective Kr wavelength is a manifestation of a small intrinsic asymmetry in the Kr emission-line profile and thus is characteristic of all interferometric measurements referenced to the Kr standard. Our observations of the magnitude and sign of this effect are in basic agreement with previous work.[11,12]

Using the precision capability of the frequency-controlled interferometer, it has been possible to carefully measure a radial dependence of the Doppler shift across the Kr discharge capillary,[13] as well as the spacer-dependent change of the effective wavelength due to asymmetry. Indeed, in our experiment the Doppler-shift radial variation was more apparent than is usual,[14] since our technique of lamp observation accepted only light which emerged parallel to the capillary axis and mapped this into the interferometer so that it was parallel to the interferometer axis; this preserved a radial variation of the Doppler shift at the interferometer. In the usual method of observation a light ray at any radius of the interferometer contains light originating at nearly all radii of the discharge; thus all light rays have approximately the same Doppler shift[14] (1.31×10^{-4} cm^{-1}), which is the weighted average over the full discharge bore.

In calculating the necessary Kr wavelength corrections we have assumed that (a) the krypton line asymmetry may be modelled by a doublet[11] and (b) the radially dependent Doppler shift has a smooth form with the intensity-weighted average given by previous studies.[14] The line shape[11] assumed was

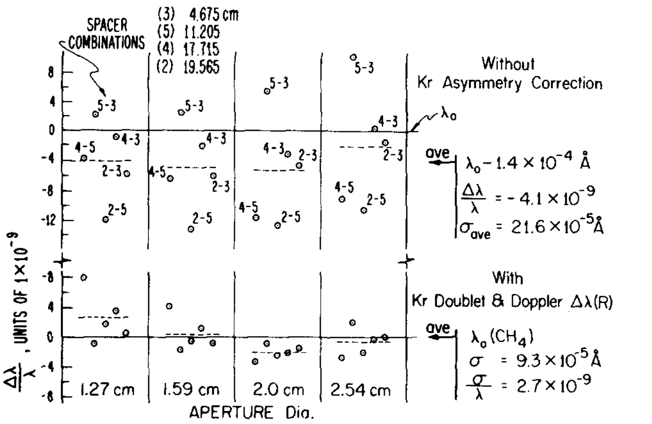

FIG. 2. Wavelength results. Top row corrected only for offsets due to diffraction and finite exit aperture. Bottom row with additional corrections for asymmetry of Kr standard line.

$$I/I_0 = \exp[-\ln 2(\Delta\sigma/\Delta\sigma_{1/2})^2] + 0.06\exp\{-\ln 2[(\Delta\sigma+\delta)/\Delta\sigma_{1/2}]^2\},$$

where

$$\Delta\sigma = (\sigma - \sigma_0) - \Delta\sigma(r)_{\text{Doppler}}$$

and

$$\Delta\sigma_{1/2} = \text{HWHM} = 0.006\,34\ \text{cm}^{-1}.$$

After a few iterations, the parameters which approximately fit our data were

$$\delta = 0.008\ \text{cm}^{-1}$$

and, for the discharge capillary of 2.1-mm diameter,

$$\Delta\sigma(r)_{\text{Doppler}} = \Delta\sigma_0 = 2.4\times10^{-4}\ \text{cm}^{-1}, \quad r = 0-0.28\ \text{mm},$$
$$= \Delta\sigma_0 \exp\{-2[(r-0.28\ \text{mm})/0.21\ \text{mm}]^2\},$$
$$r = 0.28-1.05\ \text{mm}.$$

Fringe shifts were calculated by folding the asymmetric Kr line shape with the appropriate interferometer fringe-intensity function to give the asymmetric fringe intensity $I\text{asym}(T)$ as a function of interferometer length T. Another fringe intensity $I\text{sym}(T)$ was calculated for a symmetric Kr line located at some reference point on the asymmetric Kr line (for instance, the maximum intensity).[15] The shift in T of the maximum intensity points of the simulated fringes we denote by ϵ, which typically was a few times $10^{-3}\lambda_{\text{Kr}}/2$.

From the wavelengths uncorrected for asymmetry, a set of ideal ϵ's for the virtual spacers was determined which would entirely remove the apparent CH_4 wavelength variation with interferometer length and aperture. The ϵ's calculated with the above asymmetry model were within about $\frac{1}{2}\times10^{-3}\lambda_{\text{Kr}}/2$ of these ideal ϵ's for all apertures and spacers. Our CH_4 wavelength variations were reduced by about an order of magnitude by this Kr asymmetry correction.

Wavelengths for all virtual spacers and apertures are shown in Fig. 2, both uncorrected for asymmetry and corrected. Each point represents the wavelength obtained with the indicated virtual spacer (numbers 4–5, etc., next to points in the top row). The two sets in each vertical column are for the indicated aperture. Wavelengths corrected for experimental systematic effects, but not for asymmetry, are shown in the first row (average of all points is indicated by the "ave" arrow at the right). Results for additional corrections due to Kr asymmetry are given in the second row, with the total average λ_0. Averages for each aperture are shown by the dashed lines.

Without asymmetry corrections, the aperture averages deviate from the total average by only about 2×10^{-9}, and the total average is offset from λ_0 by only -4.1×10^{-9}. Despite these small offsets of the wavelength averages, the largest deviations[12] of the λ's is about 1.1×10^{-8} and the small offset of the average λ from λ_0 is only due to the choice of spacers used. This is indicated also by the large standard deviation σ of 21.6×10^{-5} Å ($\sigma/\lambda = 6.4\times10^{-9}$).

With the inclusion of asymmetry corrections, the largest single deviation is reduced to less than 10^{-8} and the standard deviation to $\sigma = 9.3\times10^{-5}$ Å ($\sigma/\lambda = 2.7\times10^{-9}$). Use of the Doppler $\Delta\lambda(r)$ correction has improved the aperture average λ variation somewhat, but a better hypothetical distribution of velocities in the discharge could probably be found to further reduce this variation.

Our final value λ_0 for the CH_4 wavelength, obtained using the asymmetry discussed above, depends upon how the defined Kr wavelength (6057.802 105 Å) is applied to the line. Since the intrinsic asymmetry of the Kr emission line was discovered only after the meter definition was adopted in 1960, it is necessary to adopt a further convention relating the defined wavelength to the observables of the line. We shall consider two possibilities: (i) The defined wavelength applies to the asymmetric-line maximum-intensity point, or (ii) it applies to the center of gravity point.[16] If the definition corresponds to the maximum intensity, we obtain $\lambda_0 = 33\,922.314\,04$ Å, and, if to the center of gravity, $\lambda_0 = 33\,922.313\,76$ Å.

The uncertainty in the two individual values should probably be larger than the 2.7×10^{-9} standard deviation of the wavelength set due to the uncertainty in the Kr asymmetry model. For instance, if the Rowley-Hamon[11] model is used without the radial Doppler dependence, the wavelength is decreased by 1.2×10^{-9}. Thus, for our result we estimate a 68% confidence interval of

$$\delta\lambda = \pm 1.2\times10^{-4}\ \text{Å},$$

$$\delta\lambda/\lambda = \pm 3.5\times10^{-9}$$

by combining quadratically the 2.7×10^{-9} standard deviation with two times the 1.2×10^{-9} model dependence shift.

This methane wavelength has also been measured by Giacomo.[17] Wavelengths inferred from his measurement for the two cases discussed here are somewhat lower than ours, although 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.

Using the measured methane-stabilized laser frequency[5] of $\nu = 88.376\,181\,627(50)$ THz ($\delta\nu/\nu = \pm 5.6\times10^{-10}$) and the presently reported wavelength of $\lambda = 3.392\,231\,376\,(12)$

μm ($\delta\lambda/\lambda = \pm 3.5\times 10^{-9}$), we calculate the speed of light to be $c = 299\,792\,456.2\,(1.1)$ m/sec ($\delta c/c = \pm 3.6\times 10^{-9}$). This value is based on the arbitrary convention that the krypton defined wavelength is to be applied to the center of gravity of the krypton line; if the maximum intensity point were chosen instead,[16] the methane wavelength—and hence the value of c—would be increased by 8.3 parts in 10^9 [$c_{max\,I} = 299\,792\,458.7\,(1.1)$ m/sec]. These values are in agreement with the previously accepted value[18] of 299 792 500 (100) m/sec and are about 100 times more accurate. They are also in agreement with the recent measurement by Bay, Luther, and White,[19] who report $c = 299\,792\,462\,(18)$ m/sec.

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