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## ATOMIC STANDARDS OF LENGTH AND TIME

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### INTRODUCTION

THE units of measurement used in science for evaluating the fundamental quantities length and time are the metre and the second. Although originally related to a particular dimension of the Earth, the metre has since 1889 been defined as the distance, under specified conditions, between two parallel lines engraved on a platinum-iridium bar (the International Prototype Metre) which is carefully preserved in a vault at the International Bureau of Weights and Measures at Sèvres in France. Two astronomical units of time are now recognised: one, the mean solar second, which is related to the diurnal period of rotation of the Earth, and the other, more precisely defined and designated in 1956 as the second,\* which is related to the period of revolution of the Earth about the Sun as represented by the duration of a particular tropical year. During the next few years there is every prospect that new definitions of the metre and the second will become adopted which will be expressed in terms of certain fundamental characteristics of the atom.

The history and prospects of the metric units outlined above most aptly illustrate, not only the means adopted to establish practical standards of measurement on bases which are currently considered to be the best available for the purpose, but also the careful surveillance maintained over these bases, extending to modification or even change if desirable, so that the emerging requirements of precision and accessibility of the standards for fine measurements are satisfied. Units of measurement remain abstract conceptions and cannot be used as the practical bases of measurement until they have been defined in one of two ways: either by reference

\* Also known as the second of ephemeris time.

to an arbitrary material standard, as in the case of the metre, or by reference to natural phenomena, including physical constants, physical "situations" and the properties of specified substances. A natural astronomical standard has always been favoured as the means of defining time because of the impressively regular recurrence of such phenomena as the day, the lunar month and the year.

Both the precision and superiority of the existing definitions of the metre and the second are now in question. The generally accepted estimate of the reproducibility of the metre embodied in the hierarchic system of platinum-iridium line-standards (including the International Prototype Metre, its "Témoins" and the family of national copies) is between 200 and 300 parts in 1000 millions ( $10^9$ ). An improvement of this figure of merit to 100 parts in  $10^9$ , and perhaps better, is probably attainable on the evidence of results obtained from certain copies which have been modified by providing them with high-quality defining lines suitable for observation in a comparator furnished with photoelectric microscopes. Nevertheless, material standards remain vulnerable to secular change, damage or destruction and must be shielded as securely as possible from these vicissitudes by setting up, for the maintenance and dissemination of the unit of measurement, a hierarchy of standards which renders the basic standard practically inaccessible, while the system as a whole may be subject to small but undisclosed secular variations of length. A natural basis for length measurements was the ideal in the minds of the creators of the metric system when they adopted a certain dimension on the Earth's surface as the foundation of measurement. For various reasons the original solution was abandoned in favour of one based on material standards, but the ideal has not diminished in its appeal to those responsible at the present time for enduing the metre with the desired qualities of precision, permanence, invariability and ubiquity. As will be seen later, a recommendation to define the metre in terms of wavelengths of light, with a prospective reproducibility of 1 part in  $10^9$ , as well as other desirable attributes, is likely to become adopted in 1960.

The astronomical standards of time possess the advantages of permanence and availability but they lack the precision required in some modern scientific and technological applications. To some extent this lack of precision is overcome by averaging the astronomical measurements over long periods by means of clocks, such as pendulum clocks or the more accurate quartz clocks. By averaging over a period of one hundred days, for example, the mean solar second can be determined with an accuracy of about 1 part in  $10^9$ . Quartz clocks always tend to lose or gain at a gradually changing

rate, but if the variation is regular the law can be obtained by measurements extending over long periods and they can then be used to provide a finely divided time scale which is adjusted from time to time so as to keep in step with the rotation of the Earth. A statistical analysis of the performance of quartz clocks has provided strong evidence that the rate of rotation of the Earth varies periodically throughout the year. A study of the periods of revolution of other bodies of the solar system during the past two centuries also indicates that the rotation of the Earth has changed in an irregular way by as much as 80 parts in  $10^9$ . It is because of these periodic and irregular variations in the mean solar second that the alternative unit based on the Earth's period of revolution about the Sun has been adopted. Unfortunately the astronomical measurements required to determine this unit are considerably less precise than those required to determine the mean solar second and they must be averaged over longer intervals to give the required accuracy. It has recently been determined with an accuracy estimated to be 2.2 parts in  $10^9$  from measurements extending over three years. Quartz clocks cannot be relied upon to give a uniform performance over such long periods and the value obtained for the second can therefore be regarded only as an average value, unless an atomic standard is used for interpolation. For most scientific purposes it is the value at a particular instant or over a short interval that is required and, as the accuracies needed even in current practical applications approach 1 part in  $10^9$ , it is clear that an astronomical standard is no longer adequate for all purposes.

#### THEORETICAL CONSIDERATIONS

It is well known that atoms can exist in a number of different states corresponding to different configurations of the electrons surrounding the nuclei, that each state is characterised by a particular energy value, and that changes from one state to another are accompanied by the quantised radiation or absorption of an electromagnetic wave having the frequency  $f$  where

$$f = (E_2 - E_1)/h \quad . \quad . \quad . \quad . \quad (1)$$

$h$  being Planck's constant and  $E_2$  and  $E_1$  the energies of the two states. As the wave travels, *in vacuo*, with the velocity  $c$ , the corresponding wavelength is

$$\lambda = ch/(E_2 - E_1) \quad . \quad . \quad . \quad . \quad (2)$$

As the quantities on the right-hand side of the equations are all fundamental constants, any atomic radiation is potentially a standard

of length or of frequency (or its inverse, time-interval). It is clear that the frequency of the radiation depends on the difference between the two energy states and that, as a great many states are normally possible, any particular group of the same kind of atoms can emit radiations at many different frequencies. Many of these fall in the optical region of the spectrum and can be seen by eye as lines in a spectroscope. Some of the lines are seen to be sharp but others are obviously wide and, by increasing the resolving power of the spectroscope, are often found to consist of a number of closely spaced lines. For a standard a single sharp line must be selected; but even single lines have a certain width, the main causes of which are listed in the table below. The width due to the Doppler effect is a simple function of the velocity of the atom (or molecule) but those due to pressure and saturation depend on the nature of the particle and the operating conditions. A compromise between line-width and intensity is often necessary and the widths given in the table simply show the order obtained in some typical experiments.

TABLE 1  
LINE WIDTHS

Cause.	Magnitude.	
	Optical Lines.	Microwave Lines.
	$f = 6 \times 10^{14}$ c/s $\lambda = 0.000\ 05$ cm	$f = 10^{10}$ c/s $\lambda = 3$ cm
Natural width . . .	$2 \times 10^{-7}$	$1 \times 10^{-15}$
Doppler width . . .	$1 \times 10^{-6}$	$1 \times 10^{-6}$
Pressure broadening .	$2 \times 10^{-7}$	$5 \times 10^{-6}$

The spectral lines used as wavelength standards cannot be used as frequency standards because there is no technique available for comparing such frequencies, of the order of  $10^{15}$  c/s, with that of the quartz clocks which are usually  $10^5$  or  $10^6$  c/s. There are, however, transitions between closely spaced levels, such as the hyperfine structure levels, which correspond to frequencies of the order of  $10^{10}$  c/s, and these can be compared easily and accurately with the quartz clock.

### THE LIGHT-WAVE STANDARD OF LENGTH

#### THE RED LINE OF CADMIUM

The idea of using the wavelength of light as a natural basis for measurements of length was first propounded by J. Babinet [1]

during the period in the nineteenth century when the wave theory of light was being developed and verified. But it was not until the last decade of the century that A. A. Michelson and J. R. Benoît, using the Michelson interferometer, made the first determination of the metre in wavelengths of the red line in the spectrum of cadmium. This practical achievement was important in demonstrating the feasibility of a metre specified solely by means of a number of light-waves. Then, during 1905-6, J. R. Benoît, C. Fabry and A. Perot repeated the determination, using the Fabry-Perot interferometer and an improved method for relating the optically measured length of their longest interferometer to the material standard of length. Their originally announced value of the cadmium wavelength  $\lambda_R$  in terms of the metre, namely  $6438.4696 \times 10^{-10}$  m in standard air,\* confirmed the Michelson-Benoît value within the accuracies of the two experiments, and the red line was adopted in 1907 [2] as the spectroscopy reference standard and as the means of defining the angstrom (Å). This was accomplished by assigning to  $\lambda_R$  the value  $6438.4696$  Å, and the angstrom has since served for all spectroscopic measurements of wavelength. The 7th General Conference of Weights and Measures, 1927 [3], although rejecting a proposal to adopt the cadmium standard as the ultimate reference for the metre, gave provisional sanction for measurements of length in terms of the accepted experimental value  $6438.4696 \times 10^{-10}$  m of this standard, which thus became a permitted alternative to direct reference to the Prototype Metre or its authorized copies. The nominal value of one angstrom is therefore  $1 \times 10^{-10}$  m.

In all, nine measurements of the metre in terms of  $\lambda_R$  have been announced, including the two already mentioned, and the results are collected together in Table 2.

The values in Table 2 are adjusted to standard atmospheric conditions and the results of seven of the measurements (including the Benoît-Fabry-Perot value) have been amended, since they were originally announced, to take account of subsequent conclusions about the values to be attributed to the metre standards employed in the various determinations. By a convenient fluke the mean of the nine values of  $\lambda_R$  is identical with the value adopted in 1907 for spectroscopy and in 1927 (provisionally) for metrology. Furthermore, there is no evidence of any trend of the values with time, thus indicating that the hierarchy of metre standards is secularly stable. The maximum deviation from the mean is equivalent to 260 parts

\* For spectroscopic purposes standard air is thus specified: dry air at 15° C under standard atmospheric pressure ( $1\ 013\ 250$  dyn/cm<sup>2</sup>) and containing 0.03 per cent. by volume of carbon dioxide.

TABLE 2

VALUES OF THE METRE IN WAVELENGTHS OF CADMIUM RED LIGHT AND THE DERIVED VALUES OF WAVELENGTH  $\lambda_R$  IN STANDARD AIR

Date.	Observers.	Laboratories.	Number of waves in 1 metre.	Wavelength $\lambda_R$ (unit = $1 \times 10^{-10}$ m).
1892-3	A. A. Michelson and J. R. Benoît	BIPM, Paris	1 553 164.25	6438.4691
1905-6	J. R. Benoît, C. Fabry and A. Perot	BIPM & CNAM, Paris	1 553 163.96	6438.4703
1927	N. Watanabe and M. Imaizumi	CBWM, Tokyo	1 553 164.46	6438.4682
1933	J. E. Sears and H. Barrell	NPL, Teddington	1 553 163.71	6438.4713
1933	W. Kösters and P. Lampe	PTR, Charlottenburg	1 553 164.29	6438.4689
1934-5	J. E. Sears and H. Barrell	NPL, Teddington	1 553 163.81	6438.4709
1934-5	W. Kösters and P. Lampe	PTR, Charlottenburg	1 553 164.27	6438.4690
1937	W. Kösters and P. Lampe	" "	1 553 164.03	6438.4700
1940	M. F. Romanova, G. V. Varlich, A. I. Kartashev and N. R. Batarchukova	IM, Leningrad	1 553 164.34	6438.4687
		Mean values	1 553 164.12	6438.4696
		<i>Standard deviation:</i> of single determination	0.26	0.0011
		of mean	0.09	0.0004

BIPM: Bureau International des Poids et Mesures  
 CNAM: Conservatoire National des Arts et Métiers  
 CBWM: Central Bureau of Weights and Measures  
 NPL: National Physical Laboratory  
 PTR: Physikalisches-Technische Reichsanstalt  
 IM: D.I. Mendeleev Institute of Metrology

in  $10^9$  (average deviation 140 parts in  $10^9$ ), which accords well with the accuracy associated with the measurement of length in terms of the material standards of the metre.

#### IMPROVED SOURCES OF MONOCHROMATIC LIGHT

Mention has already been made of one unsuccessful proposal for a wavelength definition of the metre and there have been others. Up to 1939 none was acceptable, mainly because no known optical radiation, emitted from a natural element, was considered to possess a significantly higher degree of reproducibility than the best material

standards. The highly favourable prospect which now exists for acceptance of a wavelength definition is due primarily to the introduction and development, during the past 20 years, of sources of monochromatic radiations using, instead of the natural elements, pure isotopes of even atomic mass and charge as the emitters of radiation. Most of the radiations formerly recognised for use as wavelength standards in spectroscopy and metrology are emitted by elements with a mixed isotope content, very often with appreciable abundances of isotopes having odd and even atomic masses, *e.g.* cadmium, krypton, mercury and iron. It is known that a single isotope with odd atomic mass produces spectral lines with hyperfine structure due to interactions between the nuclear and electronic spins. It is technically very much more difficult to isolate a single component of hyperfine structure from a radiation in the optical region than it is from one in the microwave region. A single isotope with even atomic mass and charge (zero nuclear spin) emits lines free from hyperfine structure and is the best source of an optical standard. Elements with two or more isotopes having different masses (whether odd or even) are also subject to isotope displacement effects—the corresponding radiations from isotopes of different masses are not coincident—and give rise to unwanted structure or to apparent broadening due to overlapping of hyperfine-structure components and isotope displacements. Although these effects apply to both emission and absorption of radiation by atoms, attention is confined in the following section to emission sources since these are of immediate practical importance for the definition and measurement of length.

#### LINE BROADENINGS AND DISPLACEMENTS

Besides the line-complexities characteristic of odd-mass isotopes and of isotope mixtures, there are other broadening effects on spectral lines which are now better understood and therefore capable of analysis. Three of the broadening effects, with their magnitudes in the optical and microwave regions, are listed in Table 1. Of these, the natural width is small but inevitable, for it is due to the finite widths of the two atomic energy states concerned in the emission of radiation. The Doppler width, due to random thermal motions of the emitting atoms, is dependent on the square root of the ratio of the absolute temperature to the atomic mass, and in most sources is several times larger than the natural width. Both these broadenings are symmetrical and do not produce any shift in the mean wavelength of the line. On the other hand pressure broadening effects are usually asymmetrical and introduce the

possibility of displacements of the line. They depend on the temperature, density and nature of the atomic neighbours of the emitters and can be classified as follows:

- (i) collision broadening, due to the presence of foreign atoms (impurities or carriers) with the emitters;
- (ii) resonance broadening, due to the presence of atoms of the same kind as the emitters;
- (iii) interatomic Stark broadening, due to the emitters being within the influence of ionic and electronic fields.

The broadening and complicating effects of external electric fields (Stark effects) and magnetic fields (Zeeman effects) can be rendered negligible if the best modern sources of high-quality monochromatic radiations are utilised under carefully controlled conditions of excitation.

Finally, there are the effects of self-absorption, or self-reversal, on emission lines. A radiation emitted by atoms in the central region of a discharge lamp may be partially absorbed by the surrounding cooler, less perturbed atoms and then exhibits self-reversal. The resonance lines, for which the lower energy level is the normal state, and lines for which the lower level is metastable are those most liable to self-reversal. This so modifies the intensity profile across the line that the radiation becomes effectively broadened or even assumes the appearance of a doublet when the central region of the line is completely absorbed. The broadening effect is often asymmetrical.

#### PRINCIPLES FOR A WAVELENGTH DEFINITION

It will now be evident that the closest approach to the ideal conception of monochromatic light could only be attained by a single, unperturbed atom, of even atomic mass and charge, in a state of rest relative to the observer, or perhaps by a group of such atoms each of the same mass which exert no influences upon one another. The radiation would be specified by means of the two spectral terms denoting the energy states of the atom between which the transition takes place giving rise to the emission of radiation, *cf.* equations (1) and (2). In order to eliminate the need to specify the refractive index of the medium in which the standard radiation is propagated it would be advisable to define the unit of length in terms of vacuum wavelengths. This is because the vacuum wavelength  $\lambda_0$  is a natural constant, whereas the wavelength  $\lambda_m$  in a medium of refractive index  $n_m$  varies inversely with  $n_m$ , and  $\lambda_0 = \lambda_m n_m$ .

Such indeed were the principles borne in mind by the Con-



sultative Committee for the Definition of the Metre, specially commissioned in 1952 by the International Committee of Weights and Measures for advisory purposes, when it made a number of recommendations at its first session held in 1953 [4]. This development was the result of a resolution, adopted by the 9th General Conference of Weights and Measures, 1948 [5], which drew attention to the advances made in the production of highly monochromatic light from pure even-mass isotopes and their importance for the practical definition and measurement of length.

The Consultative Committee also recommended that continuity with the existing definition of the metre would best be preserved through the intermediary of the wavelength  $\lambda_R$  of the red line of cadmium in standard air. For this purpose the mean value of  $\lambda_R = 6438.4696 \times 10^{-10}$  m, derived from the nine determinations of the metre (*see* Table 2), was accepted. Since any new reference wavelength standard selected to supersede the cadmium standard would be related to the vacuum condition, it was essential to specify a dispersion formula for standard air. The committee adopted that evaluated by B. Edlén [6] from the results of three independent experimental determinations. Edlén's dispersion formula had already been internationally accepted for purely spectroscopic reductions of wavelengths to the standard-air or the vacuum condition. These provisions ensured that, as far as possible, the numerical values of all optical wavelengths expressed in angstroms would remain unchanged and that 1 Å became, by definition, equal to  $1 \times 10^{-10}$  m exactly. All the above recommendations and provisions were accepted by the 10th General Conference of Weights and Measures, 1954 [7].

#### THE FINAL RECOMMENDATION

In 1957, the Consultative Committee met for its second session with a view to drafting, if possible, a final recommendation for the metre definition in harmony with the principles it had formerly established. It considered, in particular, a number of reports [8] from different laboratories describing investigations on the monochromatic radiations from discharge lamps containing the isotopes krypton-86 (or 84), mercury-198 and cadmium-114. For the radiations of krypton isotopes, the best source is the hot-cathode discharge lamp originally introduced by W. Kösters and E. Engelhard [9] and more recently improved by the latter. In this lamp the isotope is automatically maintained at a pressure of about 0.01 mmHg; resonance and inter-atomic Stark broadenings and shifts are quite small, the Stark effect of the exciting electric field is negligible and

the Doppler width is considerably reduced by running the lamp in a cryostat maintained at the triple point of nitrogen ( $63^{\circ}$  K). The effect of departures from the ideal conditions of excitation specified by the committee are calculable. The isotope of higher mass is the easier to isolate by the process of thermal diffusion [10] used to separate the constituents of natural krypton.

J. Terrien [11] reported the results obtained from some comparative studies of the Michelson visibility functions and the intensity profiles of suitable lines in the spectra of the krypton, mercury and cadmium isotopes, and established that two lines of krypton-86—the orange line ( $6056 \text{ \AA}$ ) and the yellow-green line ( $5650 \text{ \AA}$ )—are the sharpest ever known to have been produced from discharge lamps of the established kind.

For the mercury-198 isotope the best source is the simple form of electrodeless discharge lamp designed by W. F. Meggers and F. O. Westfall [12]. It consists of a tubular envelope of Vycor glass (96 per cent. silica), about 14 cm long and 5 mm internal diameter, containing one or two milligrams of the isotope\* with a small amount of argon as carrier gas, and is excited with high-frequency electrical oscillations at 150 Mc/s or higher. Cooling the lamp with a stream of water near  $0^{\circ}$  C eliminates the tendency to self-reversal exhibited at higher temperatures by the strong green line ( $5461 \text{ \AA}$ ), which has a metastable lower state. Electrodeless lamps, similarly excited, are used also for the krypton isotopes (without carrier gas and at  $63^{\circ}$  K) and the cadmium isotope† (with carrier gas and at about  $550^{\circ}$  K). Despite its simplicity of construction and operation, there are features of the electrodeless lamp which render it less satisfactory than the hot-cathode lamp used for the krypton isotopes. It is more difficult to specify the excitation conditions rigorously and the effects of departures from the ideally specified conditions are not calculable with any certainty. The need for a foreign carrier gas (usually argon) in mercury and cadmium lamps leads to some asymmetrical broadening of the lines and, consequently, to a slight red shift of their wavelengths. Such lamps are also subject to small secular changes in effective wavelength because the carrier gas slowly disappears by adsorption on the internal surfaces of the lamp under the influence of the discharge.

\* Mercury-198 is best produced by irradiating pure gold with neutrons in an atomic reactor; some of the gold is transformed by neutron absorption into a short-lived radioactive isotope of gold which, after emission of a  $\beta$ -ray, becomes the stable isotope of mercury with atomic mass 198, separable from the gold by distillation.

† Specially investigated at the Mendeleev Institute of Metrology, Leningrad.

Nevertheless, both lamps are used, the mercury-198 lamp very extensively, for many practical purposes of measuring length not requiring the highest attainable precision, *e.g.* for calibrating end-standards and slip gauges used as reference standards of length in precision engineering measurements.

Fig. 1 shows comparison interferograms of the Fabry-Perot fringes, at a path difference of 40 cm, for the orange line of krypton-86 and the green line of mercury-198. The fringes were photographed at the NPL from an Engelhard hot-cathode krypton-86 lamp at 63° K and a Meggers electrodeless mercury-198 lamp maintained near 0° C by water cooling. The superiority of the fringes in krypton light is clearly demonstrated.

Table 3 gives the values reported to the committee of the vacuum wavelengths of three lines of krypton-86 and three lines of mercury-198 as measured in five laboratories. The results were obtained by interferometric comparisons, at a path difference of 125 mm, with the red line of cadmium from the internationally specified Michelson lamp [13]. Since the accepted value of  $\lambda_R$  in standard air is  $6438.4696 \times 10^{-10}$  m and the refractive index of standard air for the red line calculated from Edlén's formula is 1.000 276 381, the derived vacuum wavelength of the cadmium red line used as the intermediary standard is  $6440.2490, \times 10^{-10}$  m.

TABLE 3  
VACUUM WAVELENGTHS OF KRYPTON-86 AND MERCURY-198 LINES  
DETERMINED IN FIVE LABORATORIES; UNIT =  $1 \times 10^{-10}$  m

Laboratory.	BIPM.	PTB.	NPL.	CIWM.	NRC.		Mean of five laboratories.
	Michelson and Fabry-Perot.	Michelson.	Fabry-Perot.	Fabry-Perot.	Fabry-Perot.	Michelson.	
Engelhard hot-cathode krypton-86 lamp	5651.1286	88	86	86	83	82	5651.1286; 6057.8021 6458.0720
	6057.8021	23	21	21	21	19	
	6458.0720	22	15	25	18	16	
Meggers electrodeless mercury-198 lamp	5462.2707	09	07	08	04	04	5462.2707 5771.1984; 5792.2685
	5771.1985	87	85	84	79	78	
	5792.2684	87	85	86	83	80	

BIPM: Bureau International des Poids et Mesures, Sèvres

PTB: Physikalisch-Technische Bundesanstalt, Braunschweig

NPL: National Physical Laboratory, Teddington

CIWM: Central Inspection Institute of Weights and Measures, Tokyo

NRC: National Research Council, Ottawa

The Consultative Committee decided to recommend the orange line of krypton-86, corresponding to the transition between the

energy levels of the krypton-86 atom denoted by the spectral terms  $2p_{10}$  and  $5d_5$ , as the means for future definition of the metre. On this basis the metre is equal to 1 650 763.73 vacuum wavelengths of the radiation, the corresponding value of the wavelength being  $6057.8021_1 \times 10^{-10}$  m. Recent work, particularly by E. Engelhard [14], indicates that the wavelength of the recommended line, when emitted from the hot-cathode krypton lamp at  $63^\circ$  K and corrected for the small departures from the ideally specified conditions of excitation, is reproducible to about 1 part in  $10^9$ . It would then be more precise by about two orders of magnitude than the material standard as the means of *defining* the unit of length. Practical calibrations of the highest quality of existing end-standards and line-standards of 1-metre length in terms of the krypton-86 wavelength standard would probably not attain a better precision than 10 parts in  $10^9$ , or about one order of magnitude better than can be obtained from comparisons with material standards of the metre.

In 1958, the International Committee of Weights and Measures [15] considered the final recommendation made by the Consultative Committee and unanimously agreed to submit a resolution embodying the recommendation for plenary discussion and decision at the 11th General Conference of Weights and Measures in 1960.

#### THE ATOMIC-BEAM SOURCE

An extremely interesting type of source, which would be expected to produce nearly ideal monochromatic radiation, is the atomic beam used either in emission or absorption. Preliminary studies of the atomic beam, using pure isotopes of suitable type, have been initiated in some laboratories with the object of assessing the practical possibilities of such a source for providing a standard superior even to the krypton-86 standard described above. Some discussion of these possibilities took place at the 1957 session of the Consultative Committee. It was especially considered whether or not it would be reasonable to defer the drafting of any recommendation to define the metre optically until there had been more opportunity to assess the chances of using a spectral line from an atomic-beam source as the basis. In the event the committee unanimously agreed to proceed with the krypton-86 standard on the grounds that a new definition of the metre is urgently needed. One of the two resolutions on this subject to be submitted by the International Committee to the next General Conference will instruct the Consultative Committee to continue the studies of improved wavelength standards, with special reference to atomic beam sources.

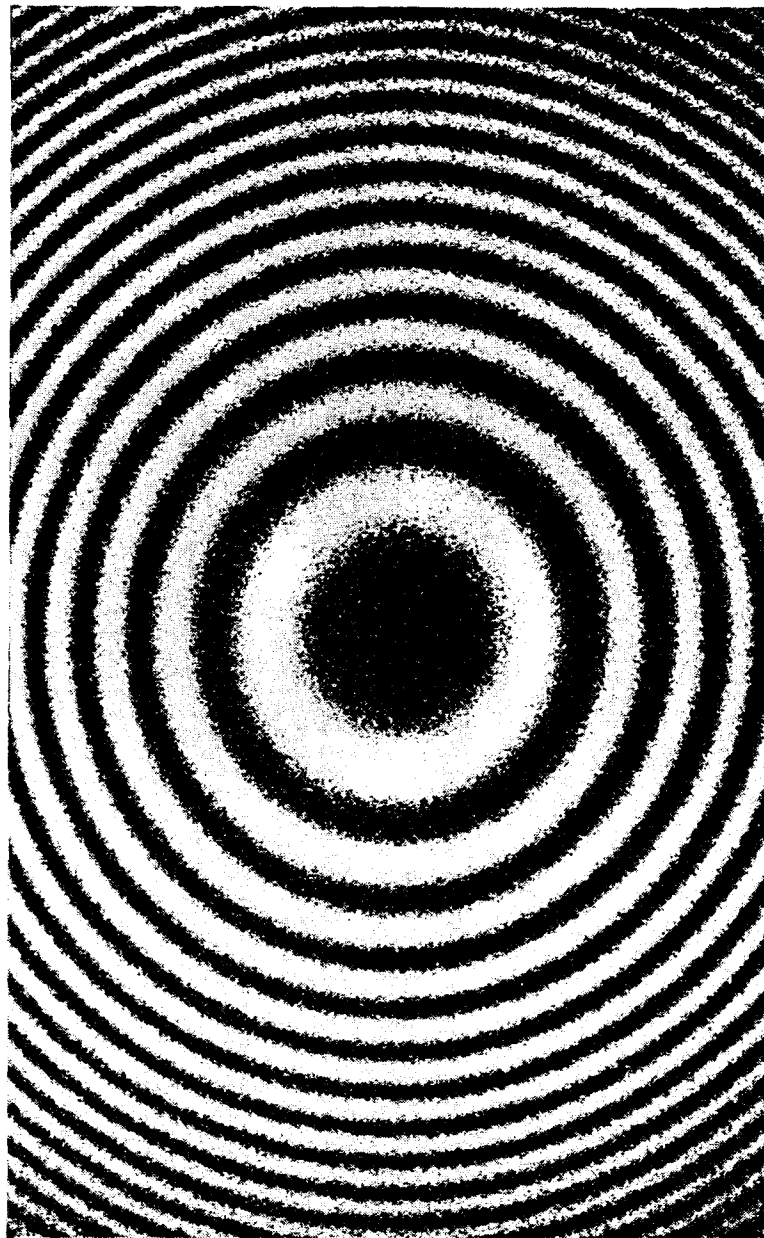
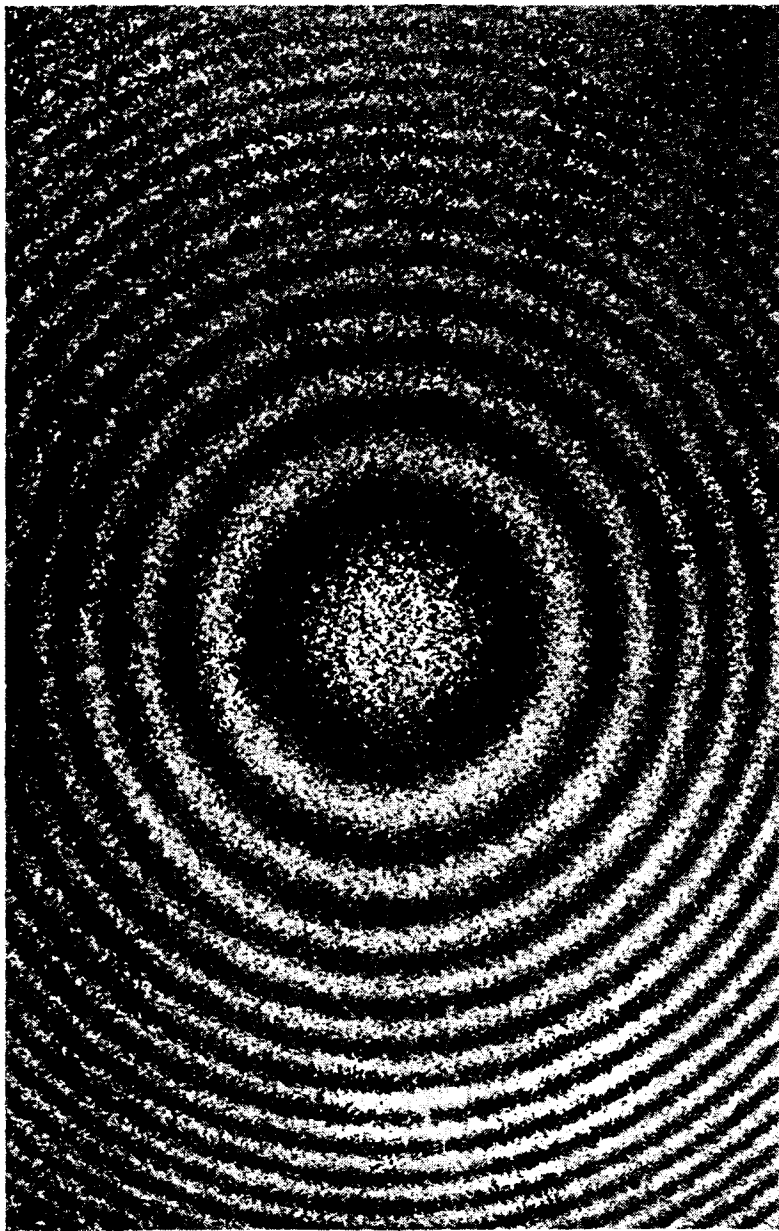
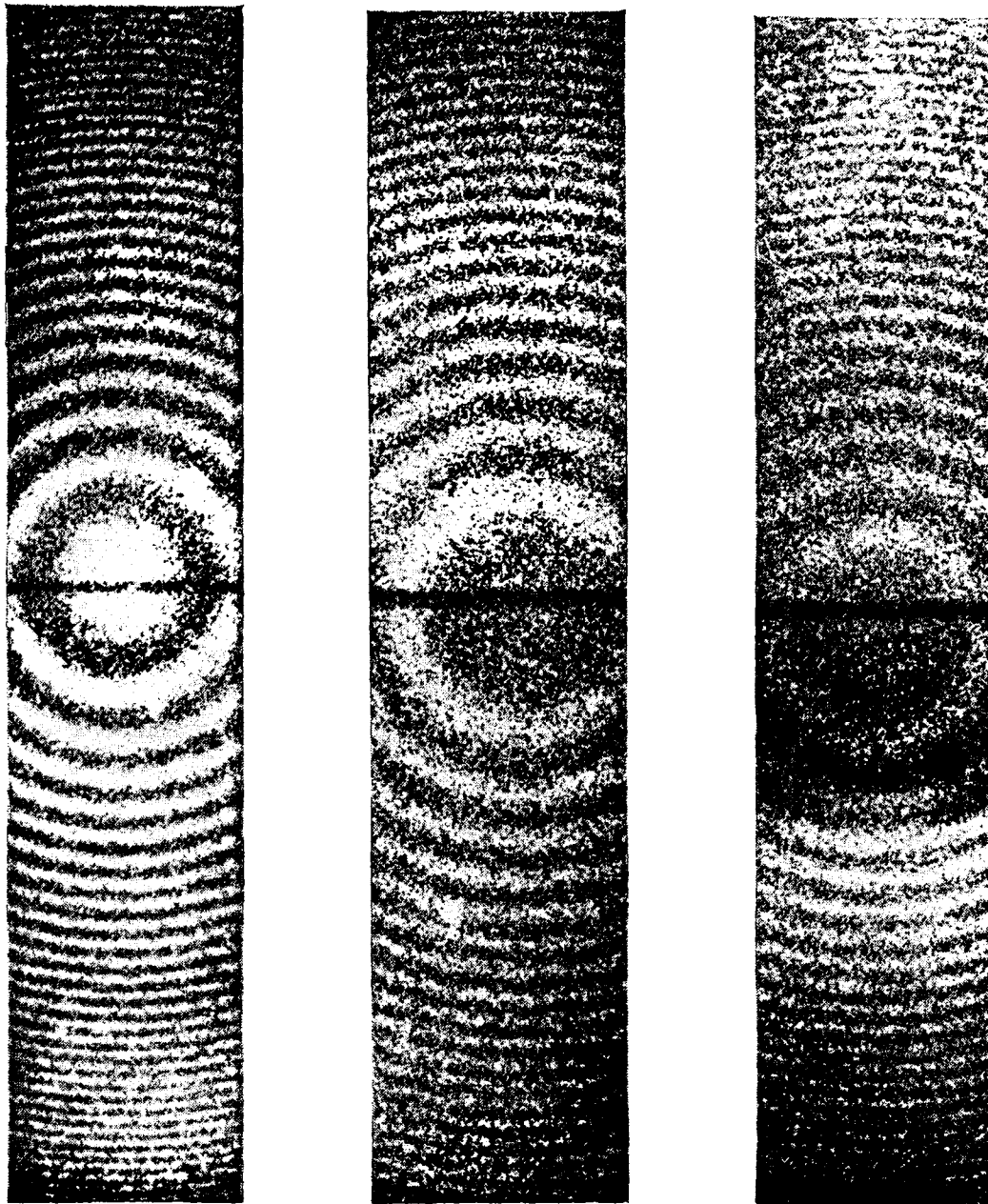


FIG. 1. — Fabry - Perot fringes in the green line of mercury-198 (*left*) and the orange line of krypton-86 at the same path of difference of 40 cm; approximate limits of interference are 50 cm for the mercury-198 line and 80 cm for the krypton-86 line.



78 cm

Retardation

94 cm

108 cm

FIG. 2.—Fabry-Perot fringes in the calcium-40 violet line from an atomic beam source, with collimation  $\approx 60$ , at various path differences up to 108 cm; the fringe visibility at greater retardations up to 130 cm suggests that the limit of interference is considerably higher. (Interferograms obtained by K. W. MEISSNER and V. KAUFMAN, Purdue University.)

Some indication is given in Fig. 2 of the extreme sharpness of a line emitted from an atomic beam. The Fabry-Perot interferograms were obtained, at path differences of 78, 94 and 108 cm, in the resonance line (4226 Å) of natural calcium (97 per cent. calcium-40) emitted from an atomic beam with a collimation factor of 60, by K. W. Meissner and V. Kaufman (Purdue University) [16], who have kindly given permission to reproduce their fringe pictures in this article. The fringes have also been photographed at longer path differences up to 130 cm, indicating that the line-width achieved is not far in excess of the natural width, and the experiments are continuing with the object of assessing the coherence (or limit of interference) of the radiation at path differences up to 200 cm.

### SPECTRAL-LINE FREQUENCY STANDARDS

#### THE AMMONIA STANDARD

The first radio-frequency lines to be found experimentally were those associated with the inversion of the ammonia molecule in which the nitrogen atom reverses its position relative to the plane of the hydrogen atoms. If energy from a klystron oscillator is passed down a waveguide containing ammonia at a pressure of 0.001 mmHg and the frequency of the oscillator is varied through the value of 23 870 Mc/s a detectable amount of the radiation is absorbed, the band width of the absorption line being about 0.1 Mc/s. This line (designated the Ammonia  $J = 3, K = 3$  line) is by far the strongest radio-frequency line and it has been intensively studied with the object of reducing the band width. Success was achieved by J. P. Gordon, C. H. Townes and H. J. Zeiger [17] at Columbia University and by N. G. Basov and A. M. Prokhorov [18] in the Soviet Union with the introduction of the "Maser" (Microwave amplification by the stimulated emission of radiation). In this form of standard a beam of molecules is directed through a system of electrodes (Fig. 3) which deflect molecules in the low energy state out of the beam, leaving those in the high energy state to enter the cavity resonator. In the resonator equilibrium between the states is established and sufficient radiation is liberated from the atoms to amplify a microwave signal at the line frequency or, under suitable conditions, to maintain oscillations at this frequency. It thus becomes a radio-frequency emission line and possesses the important advantage over optical emission lines excited in the usual way that the radiation is continuously coherent. The frequency however depends to a small extent on the electrical circuit and on the operating conditions, and it is therefore difficult to define it in terms of the

spectral line with a precision closer than  $\pm 10$  parts in  $10^9$ , although by careful control of the conditions and the adoption of a definite operational procedure J. Bonanomi, J. de Prins, J. Herrmann and

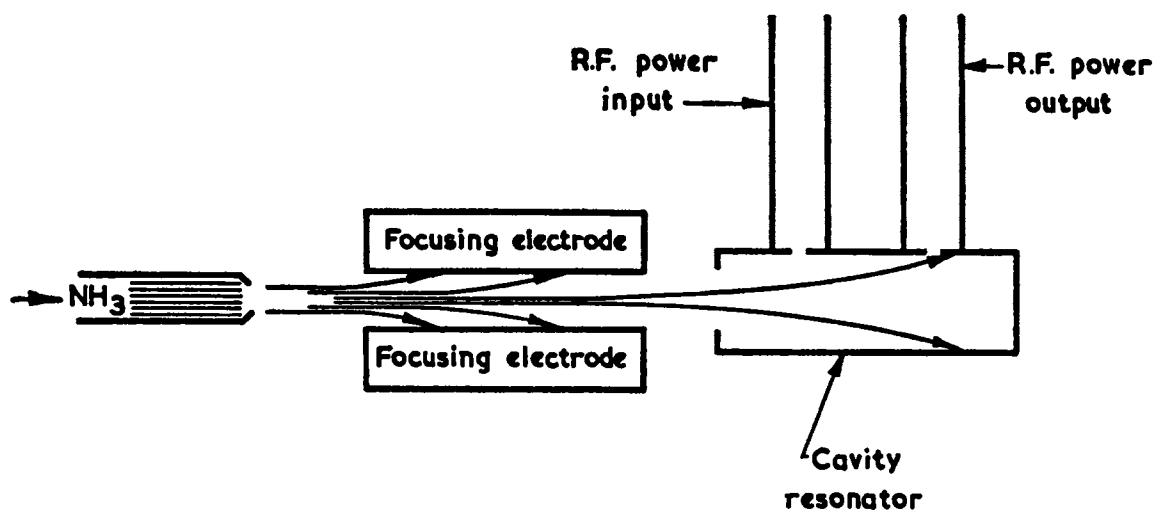


FIG. 3.—Representation of the maser.

P. Kartaschoff [19] obtained a repetition accuracy of  $\pm 0.2$  part in  $10^9$ . Their "masers" at Neuchâtel are used to give a calibration of the Observatory Time Signals in terms of an adopted value (23 870.129 235 Mc/s) of the ammonia spectral line.

#### THE CAESIUM STANDARD

A very different and so far a more successful method of using a spectral line as a frequency standard is based on a technique of radio-frequency spectroscopy developed at Columbia University by I. I. Rabi, P. Kusch, and J. R. Zacharias and described in detail by N. F. Ramsey [20]. Many atoms possess magnetic moments the strength and direction of which are functions of the energy states of the atom. An atom possessing a magnetic moment is deflected in a non-uniform magnetic field and it is thus possible to detect the occurrence of transitions between the different states by observing the deflection of the atoms in suitably disposed fields. The method possesses great sensitivity and spectral lines which would give a quite negligible absorption or emission of radiation can be detected with ease. A line in the hyperfine structure of the caesium atom promised to be particularly suitable as a standard because its frequency, near 9200 Mc/s (wavelength 3 cm), is very convenient for the electronic equipment, caesium atoms are the easiest of all to detect, and they can be produced in a simple manner by heating to about  $150^\circ \text{C}$  a mixture of caesium chloride and sodium.

The caesium atom can be regarded as consisting of a central



nucleus surrounded by a cloud of electrons and then by a single valency electron, which is spinning in either the same or the opposite direction to the nucleus. According to the direction of spin the atom possesses slightly different energy values, and also equal but opposite magnetic moments.

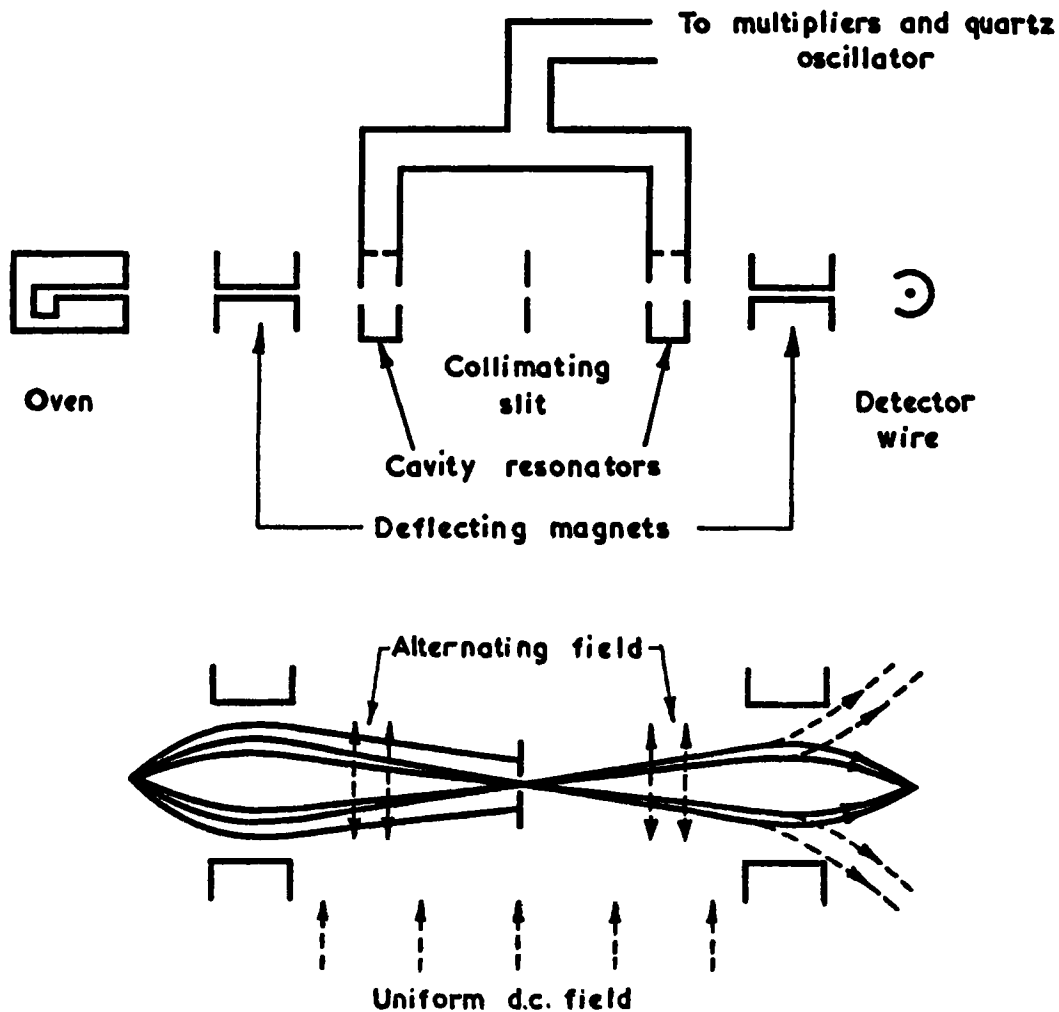


Fig. 4.—Component parts of the beam chamber and typical beam paths.

The method was the climax of a long development in the techniques of using atomic beams but the equipment required for a frequency standard [21] is remarkably simple as seen from Fig. 4. It consists of an oven, collimating slit and detector, with two magnets of a field strength and gradient of about 4000 oersted for producing the deflections and two cavity resonators for inducing transitions from one state to the other. All the components are housed in a highly evacuated enclosure, a vacuum of the order of  $1 \times 10^{-6}$  or  $1 \times 10^{-7}$  mmHg being required to enable the majority of the atoms to travel the length of the chamber without collisions.

Typical paths which the atoms are made to follow are shown in the lower part of the figure, the deflections being greatly exaggerated.

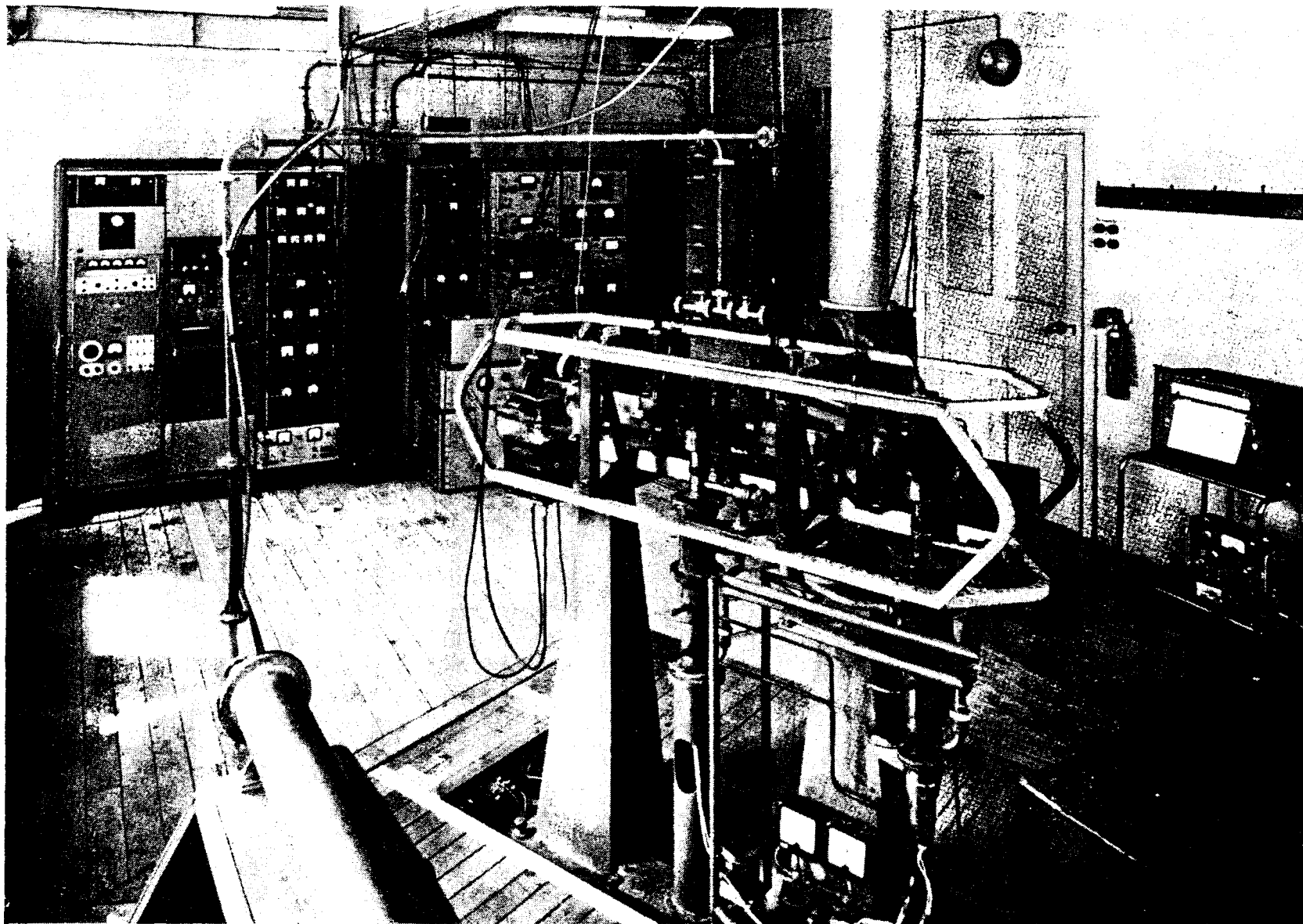


FIG. 5.—View of atomic caesium standards at the National Physical Laboratory.

The model in the centre surrounded by field coils has been in operation since 1955. In the foreground is a self-contained model and the vertical tube passing into the room above is a more accurate model under construction. An "Atomichron" can be seen in the background.

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path length. Widths as small as 120 c/s have been obtained and experience suggests that this could be reduced to 50 c/s. A long vertically mounted caesium standard nearing completion at the NPL for this purpose is shown in Figure 5.

The fact that such a high precision can be obtained in a short time enables another important property of any standard—that of permanence—to be tested by controlled experiments. The various parameters of the design can be altered and the effects on frequency measured by comparison with that of another atomic standard or, since stability over a short time only is required, with that of a quartz standard. Such an investigation has been carried out at the National Physical Laboratory. It was found that the frequency was independent of most of the parameters to an extraordinary degree and that the only two needing careful control are the value of the d.c. magnetic fields and the phasing of the alternating field in the two cavities. Both of these can be checked by observations made with the standard itself and it is thought that the effects of accidental variations should not exceed a few parts in  $10^{11}$ .

#### INTERCOMPARISON OF CAESIUM STANDARDS

Another important way of establishing the reliability of a new form of standard is to compare models of different and independent construction. A series of measurements [22] was made at the NPL between the NPL standard, two commercial "Atomichrons" and an experimental caesium standard made by the same firm (The National Company Inc.). There were large differences in the constructional details of the atomic beam tubes and in the methods of operation. The NPL caesium standard is used simply as a resonator to which an oscillator is set manually and measured in terms of the quartz standard, while the National Company standards are provided with a servo-loop which maintains the frequency of a quartz oscillator in a fixed relationship with that of the caesium frequency. For a month regular comparisons were made, the electronic controls of the "Atomichrons" being realigned for each measurement. The standard deviation of these results was 0.03 part in  $10^9$ . The average differences between the standards is given in Table 4.

TABLE 4

FREQUENCY DIFFERENCE BETWEEN THE NPL CAESIUM STANDARD, TWO ATOMICHRONS NOS. 111 AND 117 AND AN EXPERIMENTAL MODEL No. 857 X 1

111—NPL	$0.22 \pm 0.14 \times 10^{-9}$
117—NPL	$0.32 \pm 0.14 \times 10^{-9}$
857 X 1—NPL	$0.15 \pm 0.14 \times 10^{-9}$

The limits of error include the estimated systematic errors.

## INTERCOMPARISONS BY RADIO WAVES

Unlike the other standards of measurement a frequency standard can be made available at great distances by means of radio transmissions, and since 1955 the Rugby service of transmissions under the call sign MSF has been calibrated at the NPL in terms of the caesium line. The high frequency transmissions (frequencies of 2.5, 5 and 10 Mc/s) are received at a distance after reflection at the E and F reflecting layers of the ionosphere and variations in these layers give rise to a Doppler effect, so that the received frequency may differ by as much as 100 parts in  $10^9$  from the transmitted frequency. On the other hand, the propagation conditions for the 60 kc/s transmission and also for the 16 kc/s transmission (GBR) which is controlled by the same standard are remarkably stable. There is a small effect at sunrise and sunset but there is evidence that during the daytime the effects due to propagation are not greater than a few parts in  $10^{10}$  and perhaps on the average a few parts in  $10^{11}$ . The transmissions can be used for setting simple local standards to the caesium value and they can then be used for measurements of the highest precision. The Rugby transmissions, for example, have been used in both the U.S.A. and New Zealand.

The transmissions also enable distant standards to be compared and Table 5 gives the results of a series of comparisons [23] made between Harvard University, Boston and the NPL. At Harvard, J. A. Pierce measured both the MSF transmissions and also a transmission controlled by an "Atomichron" at Camden, New York, and thus obtained the relationship between them. Monthly averages are given but the standard deviation of individual daily measurements was only  $\pm 0.4$  part in  $10^9$ .

TABLE 5

COMPARISON BY RADIO BETWEEN THE NPL CAESIUM STANDARD AND THE ATOMICHRON AT CAMDEN, NEW YORK

Date.	Frequency difference NPL-Atomichron; unit $1 \times 10^{-9}$
November 1956 . . .	0.7
December . . . . .	0.2
January 1957 . . . . .	0.6
February . . . . .	0.8
March . . . . .	0.5
April . . . . .	0.0
May . . . . .	0.2

## THE UNIT OF FREQUENCY AND TIME-INTERVAL [24-27]

The first caesium standard was not in operation until the middle

of 1955 but the various checks and inter-comparisons that have been made give such convincing evidence of its reliability, reproducibility and ease of application that it is already generally accepted to be the most accurate of all the standards of measurement, including the astronomical standard for the unit of time interval. The caesium standard is ideally suited to measurements of frequency and to many physical measurements which involve relatively short intervals of time, because, when calibrated in terms of the caesium standard at regular intervals, quartz clocks serve to integrate the atomic unit quite reliably over intervals of days, weeks and even years. In this way the variations in the rate of rotation of the Earth have been measured [28]. Some measurements, particularly in the field of astronomy, extend over many years and these are more conveniently expressed in terms of an astronomical standard. The astronomical standard moreover serves the additional useful purpose of giving the time of day, which is essential for civil life.

There are thus strong reasons for using the atomic standard for precise measurements and retaining the astronomical standard for preserving continuity over long periods and for giving the time of day. On the other hand it would be undesirable and confusing to have two unrelated units of time and it should be possible to combine the useful features of both standards in the derivation of a single unit which will be adequate for all purposes.

Until this has been done it will be necessary to specify the unit employed in measurements having a precision better than a few parts in  $10^8$ . The unit of time made available by observatory time signals is the mean solar second or, more precisely, the second of Universal Time 2 (UT2). The internationally agreed fundamental unit, however, is the second of Ephemeris Time (ET) based on the revolution of the Earth about the sun and this is made available in retrospect by means of a correction to UT2. If a unit derived from an atomic standard is used it is necessary to state the adopted value of the frequency of the spectral line employed. For example, the provisional value adopted at the NPL for the caesium line was 9 192 631 830 c/s, which was based on the value in terms of UT2 when the measurement was first made (June 1955). A joint programme of work between the U.S. Naval Observatory and the NPL [29] has given a value based on a three-year average in terms of the second (of ET). This is

$$\begin{aligned} \text{Frequency of caesium } F_{m(4,0)} &\rightleftharpoons F_{m(3,0)} \text{ line at zero field} \\ &= 9\,192\,631\,770 \pm 20 \text{ c/s} \end{aligned}$$

and the value of 9 192 631 770, with limits of  $\pm 1$  c/s, is being used provisionally at the NPL and at the U.S. Naval Observatory to define a unit of atomic time.

### CONCLUDING REMARKS

There is little doubt that atomic standards will serve to define the fundamental units of length and time within the next decade. They are already in wide use and are proving themselves not only more precise but far more convenient and accessible than the standards which are the bases of the present definitions. A recommendation to establish a metre equal to 1 650 763.73 vacuum wavelengths of the orange radiation ( $2p_{10} - 5d_5$ ) of the krypton atom of mass 86 is to be submitted in 1960 for formal approval by the 11th General Conference of Weights and Measures. The reproducibility of the selected wavelength approaches 1 part in  $10^9$ . There is some prospect of establishing the second of time in terms of the frequency of an atomic (or molecular) radiation, with a reproducibility of 1 part in  $10^{10}$ , when the General Conference meets again in 1966. Appropriate means will, however, need to be devised to correlate and harmonise, as and when found necessary, the units of time furnished by the astronomical and atomic standards. Both these must continue to be used, the former for the long-term purposes of astronomy and the latter to meet the immediate needs of physics and radio-engineering.

After many years have elapsed it should be possible to ascertain whether there is any difference between atomic and astronomical scales of time—a matter of some cosmological interest to which attention has been directed by E. C. Bullard [30]. The atomic beam is at present the most favoured source of the frequency standard and a beam of suitable atoms may well, in due course, provide a wavelength standard superior even to the krypton-86 line emitted by the hot-cathode discharge lamp at  $63^\circ$  K.

In the present state of knowledge there seems little hope of defining the fundamental unit of mass in terms of a natural standard, *e.g.* an elementary particle, atom or molecule, with the precision of 1 part in  $10^8$  ascribed to the existing material standard. R. D. Huntoon and U. Fano [31] have drawn attention, however, to the possible use of the gyromagnetic ratio of the proton as the means of defining a third basic unit (probably the gauss). This with the units of length and time, also depending on atomic characteristics, might provide in the future the stable foundations of a comprehensive system of measurement for all other physical quantities.

## REFERENCES

1. J. BABINET (1829), *Ann. chim. phys.*, **40**, 177.
2. *Trans. int. Un. Sol. Res.* (1907), **20**, 28.
3. *Trav. Bur. int. Poids Mes.* (1930), **18** (C.R. 7<sup>e</sup> Conf. gén. 1927), 52.
4. *P.V. Com. int. Poids Mes.* (1954), **24**, M43.
5. *C.R. 9<sup>e</sup> Conf. gén. Poids Mes.* (1948), **44** (Paris: Gauthier-Villars, 1949).
6. B. EDLÉN (1953), *J. opt. Soc. Amer.*, **43**, 339.
7. *C.R. 10<sup>e</sup> Conf. gén. Poids Mes.* (1954), **46** (Paris: Gauthier-Villars, 1955).
8. *P.V. Com. int. Poids Mes.* (1958), **26-B**, M7.
9. W. KÖSTERS and E. ENGELHARD (1950). *P.V. Com. int. Poids Mes.*, **22**, 137.
10. E. ENGELHARD (1958), *P.V. Com. int. Poids Mes.*, **26-B**, M51.
11. J. TERRIEN (1958), *P.V. Com. int. Poids Mes.*, **26-B**, M135.
12. W. F. MEGGERS and F. O. WESTFALL (1950), *J. Res. Nat. Bur. Stand.*, **44**, 447.
13. *P.V. Com. int. Poids Mes.* (1935), **17**, 91.
14. E. ENGELHARD—private communication, results to be published.
15. *P.V. Com. int. Poids Mes.* (1958), **26-A** (to be published).
16. K. W. MEISSNER and V. KAUFMAN (1959), *J. opt. Soc. Amer.* (to be published).
17. J. P. GORDON, C. H. TOWNES and H. J. ZEIGER (1955), *Phys. Rev.*, **99**, 1264.
18. N. G. BASOV and A. M. PROKHOROV (1955), *Bull. Acad. Sci. USSR*, **101**, 1.
19. J. BONANOMI, J. DE PRINS, J. HERRMANN and P. KARTASCHOFF (1957), *Helv. Phys. Acta*, **30**, 288 and 492.
20. N. F. RAMSAY (1956), *Molecular Beams* (Oxford: Clarendon Press).
21. L. ESSEN and J. V. L. PARRY (1957), *Phil. Trans. A*, **250**, 45.
22. L. ESSEN, J. V. L. PARRY, J. H. HOLLOWAY, W. A. MAINBERGER, F. H. REDER and G. M. R. WINKLER (1958), *Nature (Lond.)*, **182**, 41.
23. L. ESSEN, J. V. L. PARRY and J. A. PIERCE (1957), *Nature (Lond.)*, **180**, 526.
24. L. ESSEN (1956), *Nature (Lond.)*, **178**, 34.
25. L. ESSEN and J. V. L. PARRY (1956), *Nature (Lond.)*, **177**, 744.
26. L. ESSEN (1957), *Nature (Lond.)*, **180**, 137.
27. L. ESSEN (1957), *Research*, **10**, 217.
28. L. ESSEN, J. V. L. PARRY, W. MARKOWITZ and R. G. HALL (1958), *Nature (Lond.)*, **181**, 1054.
29. W. MARKOWITZ, R. G. HALL, L. ESSEN and J. V. L. PARRY (1958), *Phys. Rev. Letters*, **1**, 105.
30. E. C. BULLARD (1955), *Nature (Lond.)*, **176**, 282.
31. R. D. HUNTOON and U. FANO (1950), *Nature (Lond.)*, **166**, 167.