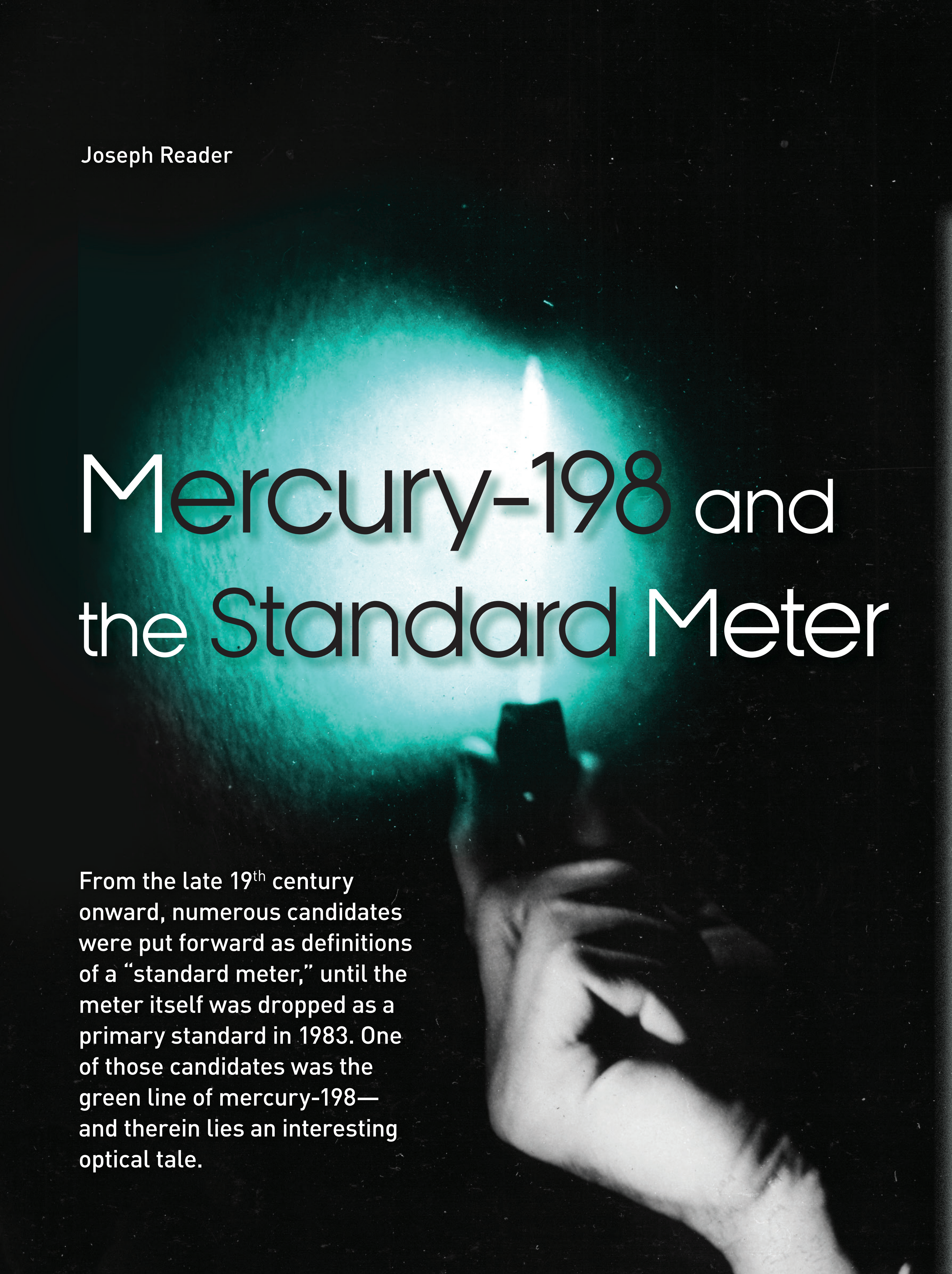
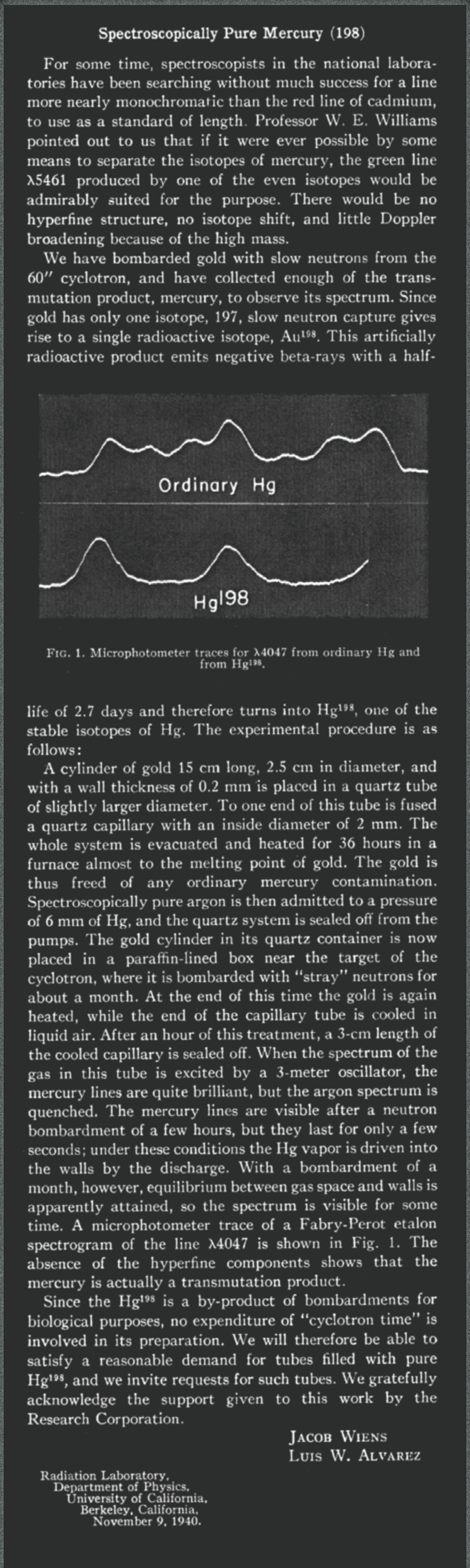


Joseph Reader

Mercury-198 and the Standard Meter

From the late 19th century onward, numerous candidates were put forward as definitions of a "standard meter," until the meter itself was dropped as a primary standard in 1983. One of those candidates was the green line of mercury-198—and therein lies an interesting optical tale.



Spectroscopically Pure Mercury (198)

For some time, spectroscopists in the national laboratories have been searching without much success for a line more nearly monochromatic than the red line of cadmium, to use as a standard of length. Professor W. E. Williams pointed out to us that if it were ever possible by some means to separate the isotopes of mercury, the green line $\lambda 5461$ produced by one of the even isotopes would be admirably suited for the purpose. There would be no hyperfine structure, no isotope shift, and little Doppler broadening because of the high mass.

We have bombarded gold with slow neutrons from the 60″ cyclotron, and have collected enough of the transmutation product, mercury, to observe its spectrum. Since gold has only one isotope, 197, slow neutron capture gives rise to a single radioactive isotope, Au^{198}. This artificially radioactive product emits negative beta-rays with a half-life of 2.7 days and therefore turns into Hg^{198}, one of the stable isotopes of Hg. The experimental procedure is as follows:

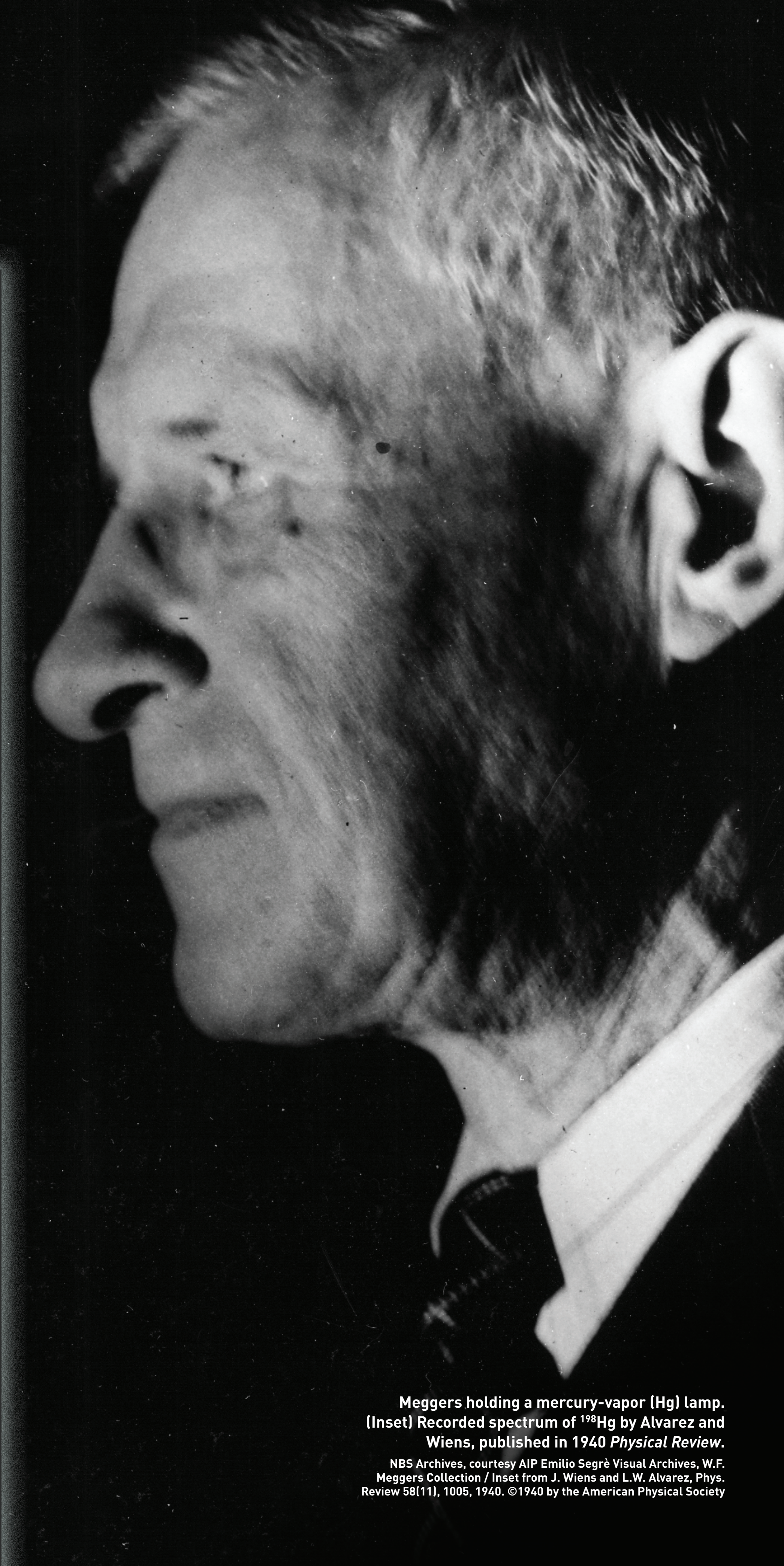
Fig. 1. Microphotometer traces for $\lambda 4047$ from ordinary Hg and from Hg^{198}.

A cylinder of gold 15 cm long, 2.5 cm in diameter, and with a wall thickness of 0.2 mm is placed in a quartz tube of slightly larger diameter. To one end of this tube is fused a quartz capillary with an inside diameter of 2 mm. The whole system is evacuated and heated for 36 hours in a furnace almost to the melting point of gold. The gold is thus freed of any ordinary mercury contamination. Spectroscopically pure argon is then admitted to a pressure of 6 mm of Hg, and the quartz system is sealed off from the pumps. The gold cylinder in its quartz container is now placed in a paraffin-lined box near the target of the cyclotron, where it is bombarded with "stray" neutrons for about a month. At the end of this time the gold is again heated, while the end of the capillary tube is cooled in liquid air. After an hour of this treatment, a 3-cm length of the cooled capillary is sealed off. When the spectrum of the gas in this tube is excited by a 3-meter oscillator, the mercury lines are quite brilliant, but the argon spectrum is quenched. The mercury lines are visible after a neutron bombardment of a few hours, but they last for only a few seconds; under these conditions the Hg vapor is driven into the walls by the discharge. With a bombardment of a month, however, equilibrium between gas space and walls is apparently attained, so the spectrum is visible for some time. A microphotometer trace of a Fabry-Perot etalon spectrogram of the line $\lambda 4047$ is shown in Fig. 1. The absence of the hyperfine components shows that the mercury is actually a transmutation product.

Since the Hg^{198} is a by-product of bombardments for biological purposes, no expenditure of "cyclotron time" is involved in its preparation. We will therefore be able to satisfy a reasonable demand for tubes filled with pure Hg^{198}, and we invite requests for such tubes. We gratefully acknowledge the support given to this work by the Research Corporation.

Jacob Wiens
Luis W. Alvarez

Radiation Laboratory,
Department of Physics,
University of California,
Berkeley, California,
November 9, 1940.



Meggers holding a mercury-vapor (Hg) lamp. (Inset) Recorded spectrum of ^{198}Hg by Alvarez and Wiens, published in 1940 *Physical Review*.

NBS Archives, courtesy AIP Emilio Segrè Visual Archives, W.F. Meggers Collection / Inset from J. Wiens and L.W. Alvarez, Phys. Review 58(11), 1005, 1940. ©1940 by the American Physical Society

Before the 18th century, units measuring distances and other lengths varied from place to place—and the measured size of an object could vary depending on where the measurement took place. An English "foot," for example, was the average length of the feet of sixteen random men coming out of a church. That changed beginning in France in the 1790s, with the development of the *Système international d'unités* (SI) or metric system, eventually adopted by most countries in the world and, in particular, by the scientific community. The basic SI unit of length is the meter—originally defined as one ten-millionth of the length of an arc from the Earth's equator to its north pole, passing through Paris. After several expeditions to determine this length, a "standard meter" was created—a platinum/iridium bar stored in Paris, with two scratches to show the precise distance marked out by a meter. Yet scientists soon realized that some sort of natural quantity would constitute a better standard of length than a single metal bar stored in Paris.

Among the leading candidates for such a natural length standard was the wavelength of a prominent spectral line. The line favored by Albert Michelson, the first American to win a Nobel Prize in Physics, was the green line of mercury, at 546.1 nm, but Michelson eventually concluded that this line was too complex to work as the standard. (Much later, it became known that the complexity Michelson observed was the result of the several isotopes present in natural mercury, and of magnetic hyperfine structure in two of the isotopes.) He eventually settled on the red line of cadmium, at 643.8 nm, as a better candidate—and, in 1892, used his interferometer to measure the distance between the scratches on the meter bar in Paris in terms of the wavelength of this line.

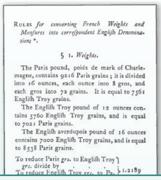
As a result of Michelson's experiments, the red line of cadmium became the standard for wavelength measurements. But it did not replace the meter bar in Paris as the standard of length, and the search for a better standard meter continued. Eventually, attention returned to the green line of mercury, which Michelson had rejected. Just why ^{198}Hg came back into the limelight is a story of work by legendary scientists at the physics department of the University of California, Berkeley, and the National Bureau of Standards (NBS)—the precursor of the National Institute of Standards and Technology (NIST)—in Washington, D.C. (There's even a measure of controversy in the story, which played out in the national news media of the day.)

Since I received my Ph.D. from U.C. Berkeley and have spent nearly my entire career at NIST, I became familiar with the both sides of the story. What follows is a personal view of the rise and fall of ^{198}Hg's green line as a standard-meter candidate.

The "Meggers lamp"

I originally became acquainted with the green line of mercury while working on a Master's thesis in spectroscopy at Purdue University, Ind., USA, in the 1950s. I was making wavelength measurements calibrated by the green line of mercury, as produced by an electrodeless lamp containing monoisotopic ^{198}Hg. (An electrodeless lamp consists of a sealed glass tube

THE STANDARD-METER QUEST



Rules *for converting French Weights and Meaſures into correſpondent Engliſh Denominations* *.

§ 1. *Weights.*

The Paris pound, poids de mark of Charlemagne, contains 9216 Paris grains; it is divided into 16 ounces, each ounce into 8 gros, and each gros into 72 grains. It is equal to 7561 Engliſh Troy grains.

The Engliſh Troy pound of 12 ounces contains 5760 Engliſh Troy grains, and is equal to 7021 Paris grains.

The Engliſh averdupois pound of 16 ounces contains 7000 Engliſh Troy grains, and is equal to 8538 Paris grains.

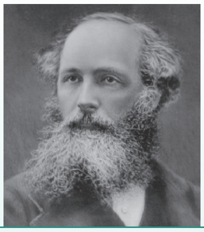
To reduce Paris *grs.* to Engliſh Troy *grs.* divide by . . . } 1.2189
To reduce Engliſh Troy *grs.* to Pa.

7 OCTOBER 1790
In Revolutionary France, a report of the Paris Academy of Sciences recommends a new decimal system of weights and measures.

30 MARCH 1791
The meter is defined as a ten-millionth of the distance between the north pole and the equator, in a meridian through Paris.

1793
An expedition measuring a length of meridian between Dunkirk and Barcelona results in a provisional standard meter bar in brass.

23 JUNE 1799
A "final" standard meter bar is created in platinum. It will remain the standard meter for much of the 19th century.



1859
James Clerk Maxwell suggests the wavelength of the yellow line of sodium as a natural standard.

Sources: Wikipedia / NIST

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Scientists realized that a natural quantity would constitute a better standard of length than a metal bar stored in Paris.

containing the material of interest. There are no electrodes; the discharge is powered by radio waves from an external source.)

For the work I was doing, this electrodeless mercury lamp had a key advantage: it provided a sharp, well-defined line that made for an excellent calibration standard. Because mercury is a heavy element, its spectral lines have low Doppler broadening; further, ^{198}Hg has an even number of protons and an even number of neutrons, so its lines do not suffer from magnetic hyperfine structure, which can complicate a spectral line's appearance. And only one isotope is present in the lamp, so there's no complication due to the presence of lines from several isotopes.

The specific lamp I was using came from the NBS laboratory of William F. Meggers—the "dean of American spectroscopists," and indeed one of the most famous spectroscopists in the world. Meggers received the Frederic Ives Medal of The Optical Society (OSA) in 1947; was the society's president in 1949-50; and has an OSA award, an award of the Society for Applied Spectroscopy, and even a crater on the moon named after him.

Meggers used Fabry-Pérot interferometers to measure the wavelength of the green line of ^{198}Hg relative to the red line of cadmium—Michelson's wavelength standard from 1892. Once those measurements were made, he distributed the lamps—each one accompanied by a certificate stating the wavelength of the green line as emitted by that lamp—to labs around the world.

Berkeley: Alvarez and Wiens

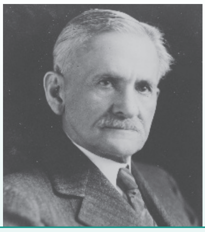
The electrodeless ^{198}Hg lamps, which played a key role in mercury-198's brief heyday as a candidate for the standard meter, came to be known to most spectroscopists as "Meggers lamps." Yet it turns out that William Meggers was not the one who invented them—as I found out after completing my Master's degree at Purdue, and moving to U.C. Berkeley in 1957 to start work on a Ph.D.

At Berkeley I became a student of another well-known spectroscopist, Francis Jenkins (the coauthor, with Harvey White, of the textbook *Fundamentals of Optics*). While looking for some equipment one day, I ran across a small box labeled "original Hg-198 lamp," inside of which was an electrodeless lamp. When I next saw Jenkins, I asked: "Francis, what is the story on this lamp? Everybody knows that these lamps were invented by William Meggers at NBS in Washington." Jenkins replied "Oh no; the mercury-198 lamp was invented by Luie with a graduate student in my spectroscopy lab."

"Luie" was the famous Luis Alvarez—one of Jenkins' buddies on the Berkeley faculty, a high-energy nuclear physicist and a sidekick of Ernest Lawrence at the University of California Radiation Laboratory, or Rad Lab (now the Lawrence Berkeley Laboratory). Alvarez—already famous for his work on the Manhattan Project

1889

The 1st General Conference on Weights and Measures (GCWM) defines the meter as the distance between 2 lines on a platinum-iridium bar.

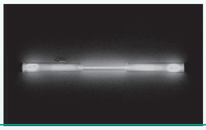


1892

A.A. Michelson measures the platinum-iridium meter in terms of the wavelength of the Cd red line, which he suggests as a standard.

1940-1960

Various groups argue for spectral lines of cadmium-114, mercury-198 and krypton-86 as possible bases for a standard meter.



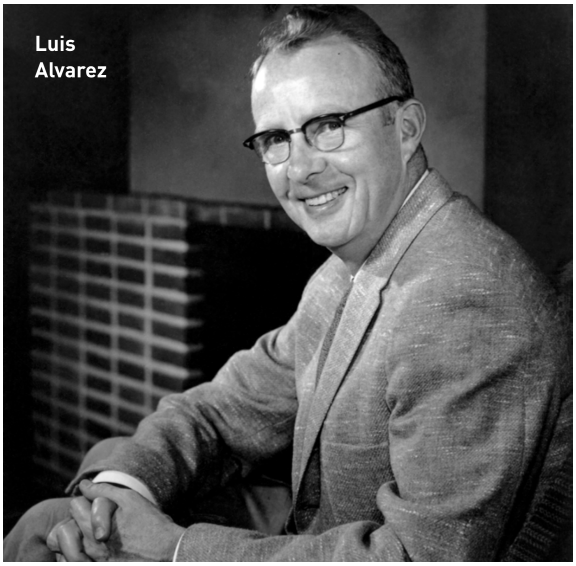
14 OCTOBER 1960

The 11th GCWM defines the meter as 1,650,763.73 vacuum wavelengths of light from a specific energy-level transition in krypton-86.

21 OCTOBER 1983

The 17th CGPM defines the meter as the length traveled by light in vacuum during an interval of 1/299,792,458 seconds.

> Jacob Wiens, who was looking for a project for his Ph.D. thesis, thought that he could design a superior oscillator for the ^{198}Hg lamp. Luis Alvarez, it turned out, had an idea of his own.



Lawrence Berkeley National Laboratory

and on the development of radar during World War II—would go on to receive the Nobel Prize in Physics in 1968 for his elementary-particle discoveries with the hydrogen bubble chamber. Still later, he would work with his son Walter, a geologist at Berkeley, to show that the impact of an asteroid at the end of the Cretaceous Period had likely led to the extinction of the dinosaurs. As students in Berkeley in the late 1950s, we would see Lawrence and Alvarez swoop down from the Rad Lab with their entourage on Wednesday afternoons for the weekly department seminar. It was always a thrill to see this group and mix with them at the pre-talk tea.

Alvarez had begun his career in Berkeley in 1936. A couple of years thereafter, W. Ewart Williams from Kings College, London, spoke at a seminar at Berkeley about using high-frequency radio waves from an oscillator to excite spectra of minute quantities of various samples. He mentioned that if a sample of one of the even-numbered isotopes of mercury could be obtained, it might be possible to build a lamp that would replace cadmium as the standard of wavelength. That would be a desirable thing, because the red line of cadmium, while known to be very sharp, presumably had an underlying structure due to the existence of eight stable isotopes, some of which would have their own magnetic hyperfine structure.

Jacob H. Wiens, a graduate student present at the talk, recounts what happened next in his contribution to a 1987 volume of tributes to Alvarez by his students and colleagues. As Wiens tells it, in discussions before the talk, Alvarez had mentioned to Williams that monoisotopic ^{198}Hg could be produced by irradiating ordinary gold (monoisotopic ^{197}Au) with neutrons, in the reaction $^{197}_{79}\text{Au} + \text{n} \rightarrow {}^{198}_{79}\text{Au} \rightarrow {}^{198}_{80}\text{Hg} + \beta^-$. That marked out an easy way to obtain a highly desirable isotope: absorption of neutrons by natural gold produces radioactive ^{198}Au (with a half-life 2.7 days), which decays promptly to stable ^{198}Hg.

Creating the lamp

Wiens, who was in his second year at Berkeley and was looking for a project for his Ph.D. thesis, was gifted at electronics, and thought that he could design a superior oscillator for the ^{198}Hg lamp. Alvarez, it turned out, had an idea of his own—while Wiens was discussing the possibility with a fellow student, Alvarez interrupted, saying that he could obtain ^{198}Hg using stray neutrons from a cyclotron at the Rad Lab. He offered to collaborate with Wiens.

The first step, of course, was to obtain the mercury isotope. Alvarez placed a gold foil close to the 60-inch cyclotron and waited. After four days, the gold had enough radioactivity to indicate the presence of some ^{198}Hg. Wiens placed the gold in a glass tube and attached it to a vacuum system containing a lamp blank. The gold was heated to drive some of the mercury that had formed into the lamp blank, which was then sealed off from the system.

When excited with a 100-MHz oscillator, the lamp produced a spectrum of pure ^{198}Hg—but only for a few seconds. The mercury vapor had been driven into the walls by the discharge. Wiens and Alvarez then irradiated the gold for a month and got a larger sample, from which they recorded a spectrum of ^{198}Hg with a Fabry-Pérot interferometer. They published their results in 1940 in *Physical Review.*

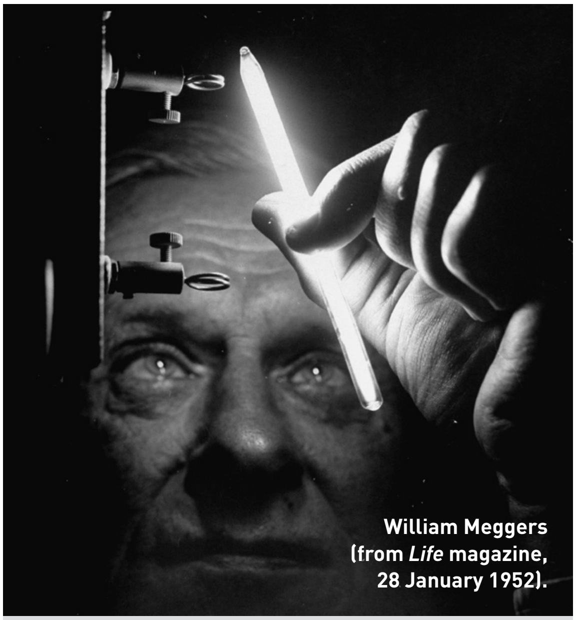
Wiens continued by exposing gold to neutrons for 10 months, and got a lamp that lasted much longer. He later wrote, "The ease of excitation, low operating temperature, high visual intensity, and inherently sharper line produced by the mercury lamp should be adequate reasons for seriously considering the green line of ^{198}Hg as a new primary standard of wavelength." Wiens received his Ph.D. from Berkeley in 1944 and went on to become a professor at the College of San Mateo in California.

From "sick fireflies" to a 50-hour discharge

Even as Wiens was wrapping up his Ph.D., the Meggers group at NBS had also started to work on the ^{198}Hg problem—and scientists from Meggers' lab placed a gold foil near a cyclotron in Berkeley to produce their own supply of the isotope. Meggers wrote that "several tiny discharge tubes were prepared in 1943, but in size, brightness, and life they were reminiscent of sick fireflies and had no practical value."

But then the project took a dramatic turn. "The prospects were very discouraging," Meggers further wrote, "when, near the end of the war, there were rumors of a secret source of neutrons thousands of times more effective than the largest cyclotron." (Clearly, these were rumblings of nuclear reactors being developed in the Manhattan Project.) "One day a dirty gold bar with a secret history was delivered to us carefully wrapped in oily paper. The gold was said to contain ^{198}Hg." From this gold they managed to make a lamp that lasted 50 hours. In 1950, Meggers published measurements for several lines of ^{198}Hg. And, though Meggers cited Wiens and Alvarez for the origin of the method, *Life* Magazine, one of the era's most popular periodicals, mistakenly credited Meggers with its discovery—a mistake that led to some long-term bad feelings (see sidebar "*Life* takes a wrong turn").

(An interesting side note: in 1952, Meggers attended a meeting of the International Astronomical Union in Rome, carrying with him one of his electrodeless lamps. While there, he met with Pope Pius XII—and, upon his return, related to his colleagues that his lamp had been blessed by the Pope. This lamp is now in the collection of the NIST Museum. In a celebrated photograph that shows Meggers with the circular rings from a Fabry-Pérot interferometer projected on a wall, the glowing ^{198}Hg lamp sits in a holder next to a lens.)



William Meggers (from *Life* magazine, 28 January 1952).

Andreas Feininger/The LIFE Picture Collection/Getty Images

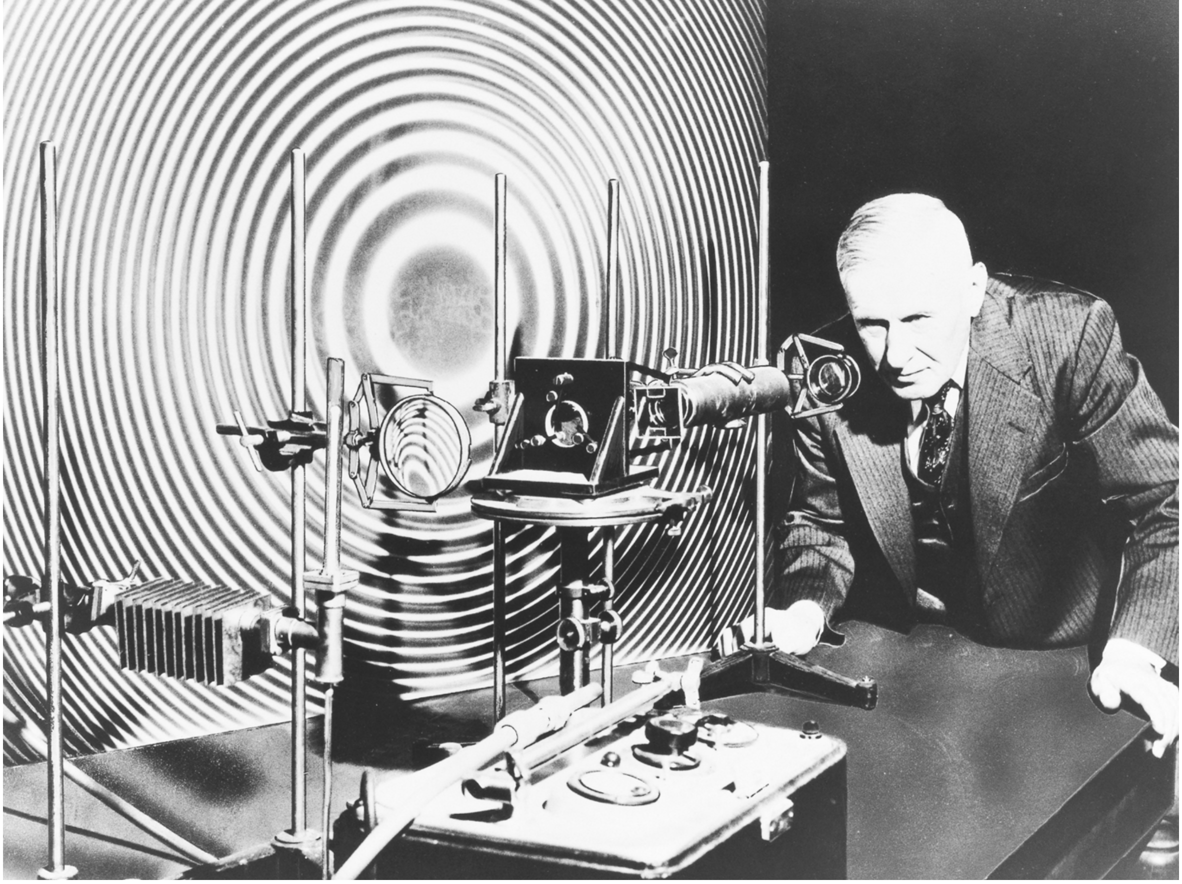
Life takes a wrong turn

In the scholarly literature, William Meggers clearly credits Jacob Wiens and Luis Alvarez for the origin of the mercury lamp. But the popular press intervened—and created a very different impression.

On 28 January 1952, *Life* Magazine, one of the era's most popular publications, carried a story about the ^{198}Hg lamp titled "Measuring with Mercury: The rays of its pale light form the world's most precise yardstick." Above the title was a picture of Meggers, with the caption "Mercury lamp's inventor, Dr. William Meggers of the Bureau of Standards, makes it glow by holding it near two high-frequency antennae."

Wiens took exception to this, and wrote a letter to *Life*, claiming that he had developed the mercury-198 lamp as part of his work in Berkeley. *Life* published Wiens' letter on 18 February 1952—but added the following note: "Dr. Meggers made the first practical mercury lamp and, with it, the first measurements.—ED."

To Wiens, that was "like crediting Lockheed with inventing the airplane because they made the first practical device that would carry 400 people from San Francisco to London." At the end of his contribution in a book of tributes to Alvarez, Wiens bemoaned his fate. "I was in Alvarez's presence less than 10 hours during my life," he wrote, "but from our interaction came my most important piece of physics, as well as pain that I carry even now." Wiens died in 1986, shortly before the book was published.



William F. Meggers with Fabry-Perot interference rings. The ^{198}Hg lamp is the white vertical tube within a holder in the photo's left half, between the lens and the mounting bracket. NIST

Toward a natural wavelength standard

With the publication of his initial successful lamp results, Meggers wrote: "Now that the isotopic curse has been lifted from mercury and ^{198}Hg has become available in abundance … in all probability the green line will be used as the ultimate standard of length." Meggers further found that if he replaced the RF oscillator with a magnetron operating at 2.45 GHz, the lamps would have almost unlimited life. He became a strong advocate of adopting the green line as the primary standard of length.

Several labs around the world studied the possibility. They found, however, that the "Meggers lamp" could not be made with sufficient reproducibility to serve as a primary standard (presumably because of the difficulty of maintaining the lamp's temperature and the pressure of its argon buffer gas). NBS pressed on by proposing to use a different ^{198}Hg line—the 254-nm line, in an atomic beam—but this had the problem of complicated construction, as well as the possibility of Doppler shifts from non-orthogonality of the beam to the optic axis.

The search for a lamp that could serve as a length standard continued. After long deliberations, the International Bureau of Weights and Measures decided in 1960 to adopt the wavelength of the orange line of isotopic krypton-86 (605.6 nm)—produced by operating a discharge of ^{86}Kr in a liquid-nitrogen bath—as the primary standard of length. The standard meter was defined as 1,650,763.73 wavelengths of the orange line.

The fall of the standard meter

The reign of ^{86}Kr did not last long, however. Construction and operation of the ^{86}Kr lamp itself was challenging, and only a few actually ever went into operation. The lamp finally met its end as a standard with the advent of lasers that could provide spectral lines much sharper and more stable than any line from a gas discharge.

In 1972, a team from NBS undertook to determine the speed of light, *c*, by making separate measurements of the frequency and wavelength of an infrared transition of the methane molecule (3390 nm), and then calculating *c* as the product of the measured frequency and the wavelength. First, the researchers used a chain of interlocked lasers to measure the frequency of the line against the primary frequency standard, the cesium atomic clock. Then, they measured the wavelength by comparison to the primary standard of length, the orange line of ^{86}Kr with a Fabry-Pérot interferometer.

The value for the speed of light that came out of this work was more accurate than any previous value. But its accuracy was still limited by the accuracy with which the researchers could measure the wavelength, which was much worse than that of the frequency

Ironically, if history could be rewritten, the 198 isotope would not be the best choice for the green line of mercury.

measurement. Because of an asymmetry in the shape of the krypton line, the wavelength could only be determined to a precision of 30 parts in 10^{10}, compared with 6 parts in 10^{10} for the frequency.

The stage was set for a radical change in philosophy. Since it was generally agreed that the speed of light in vacuum was a fundamental constant that would never change, the NIST team proposed that the speed of light be declared to have a fixed, exact value—and that the meter be defined as the distance that light travels during a specific period. The international community accepted the proposal, and in 1983 the speed of light was officially declared to be 299,792,458 m/s. The meter was taken to be the distance that light travels in 1/299,792,458 s. The era of the meter as a primary standard—whether defined by a fraction of the distance between the pole and the equator, a platinum-iridium bar in Paris, or the emission wavelength of ^{198}Hg, ^{86}Kr or anything else—had come to an end.

The practical meter

It is, of course, virtually impossible to use the speed-of-light-based definition of the meter in ordinary lab studies. Today, a meter can be realized using a helium/neon laser that is stabilized by a hyperfine transition of molecular iodine with wavelength in vacuum λ_{HeNe}(f component of R_{127} of I_2) = 632.99121258 nm. Six other lines of iodine, a line of ^{40}Ca, and the 3390-nm line of methane are also available for this purpose. The wavelengths of these lines are known to a few parts in 10^{11}. The green line of ^{198}Hg and the red line of ^{114}Cd still serve as secondary wavelength standards.

Ironically, if history could be rewritten, the 198 isotope would not be the best choice for the green line of mercury. Nowadays, mercury isotopes are obtained from natural mercury by magnetic separation in calutrons (isotope-separating mass spectrometers). Since ^{202}Hg has a much higher natural abundance (29.8 percent) than ^{198}Hg (10.0 percent), it is much less expensive, and thus a more logical choice as a standard. All of the wavelengths that have been adopted as secondary standards are for ^{198}Hg, however—and so mercury-198 remains the relevant isotope, and a part of the long story of the quest for the standard meter. OPN

Luis Alvarez, pictured here with personally built electronics and BF-3 ionization chamber, went on to win the Nobel Prize in Physics in 1968. Lawrence Berkeley National Laboratory

I thank Kristen Frederick-Frost, NIST Historian at the time, for very helpful discussions and suggestions.

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