PHYSICAL REVIEW VOLUME 70, NUMBERS 11 AND 12 DECEMBER 1 AND 15, 1946

Production of Hg^{198} as a Possible Source of an Improved Wave-Length Standard*

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(Received September 26, 1946)

The green line 5461A from any of the even isotopes of mercury is superior, in many respects, to the red line 6438A of cadmium for a primary standard of wave-length. The mercury isotope of mass 198 was produced by utilizing the nuclear transformation

$$_{79}Au^{197}+{}_0n^1\rightarrow{}_{80}Hg^{198}+{}_{-1}\beta^0.$$

One ounce of pure gold was sealed in a quartz tube and a section of 5-mm inside diameter quartz tube was sealed on the quartz-gold tube. The system was outgassed and 4-mm Hg pressure of spectroscopically pure argon was admitted and the tube was sealed. The gold was exposed to stray neutrons near the sixty-inch cyclotron for ten months. The gold was then heated and the mercury was condensed in the 5-mm quartz tube. The mercury vapor in the presence of argon gas was excited by means of a 100-megacycle oscillator and the spectrum was observed. The lines produced by the discharge were mercury lines, and the position of the lines of a Fabry-Perot etalon spectrogram agree with the position assigned by Schüler and Jones to the mercury isotope of mass 198. Larger quantities of gold exposed to known superior sources of neutrons will produce an adequate supply of the isotope for scientific purposes.

INTRODUCTION

FOR several decades the red cadmium line has been used as the primary standard of wave-length. A specially constructed cadmium lamp is used in conjunction with a Fabry-Pérot etalon to accurately compare the wave-lengths of the secondary standards with that of the red cadmium line. The lamp is also conveniently applied to measurement of standard scales of length, and to the testing of high quality optical parts and instruments, using various forms of the Michelson interferometer.

The accuracy with which these measurements and tests can be made depends upon the sharpness of the spectral line used for the standard. The ease with which some of the tests can be made depends upon the visual intensity of the standard. In recent years it has been recognized that while the cadmium lines are sufficiently sharp to produce adequate interference fringes for some purposes, a source giving rise to sharper lines is an urgent necessity for work of the highest precision.

Michelson and Benoit, in 1893, first selected the red cadmium line for the standard of wave-length, and it was adopted as such by the International Union for Cooperation in Solar Research in a resolution passed in 1907.[1] The present accepted value of 6438.4693A is the mean of seven intercomparisons of the wave-length with the meter made during the period from 1893 to 1936.[2]

Meggers,[3] in a study of the characteristics of a lamp suitable for use as a primary standard of wave-length, specified that the lamp should avoid or minimize the Doppler-Fizeau effect, hyperfine structure, self-reversal, and pressure effect. The Michelson H-type lamp as specified[4] has a number of disadvantages,[5] the most serious of which concern Meggers' first two specifications. Since the Doppler width is proportional to $\lambda(T/M)^{\frac{1}{2}}$, where λ is the wave-length, T the absolute temperature, and M the atomic weight, it is clear that the lamp should operate at as low a temperature as possible and should have as large an atomic weight as possible.

The sharpness of a spectral line may be specified by the maximum order of interference obtainable with it, which should theoretically be[6]

$$N=1.22(10)^6(M/T)^{\frac{1}{2}}.$$

For the standard cadmium lamp, with the gas in the lamp at 320°C, the computed value of N for 6438A is 530,000. For a mercury lamp, operating at a temperature of 40°C, the value for 5461A is 965,000.

* Submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Physics in the Graduate Division of the University of California.

[1] Trans. I.U.C.S.R. **2**, 20 (1907).

[2] J. E. Sears, Sci. Prog. **31**, 209 (1936).

[3] W. F. Meggers, Rev. Mod. Phys. **14**, 2–3, 59 (1942).

[4] Procès-Verbaux Comité Int. Poids et Mes. **2**, 17, 91 (1935).

[5] C.-W. Hsueh, J. Op. Soc. Am. **36**, 160 (1946).

[6] H. Buisson and Ch. Fabry, J. de physique **5**, 2, 442 (1912).

The ideal lamp should be capable of operating with very low currents and ion densities in order to prevent broadening due to the Stark effect. The pressure in the lamp should remain reasonably constant during operation so as not to cause a shift in the line due to the pressure effect. The influence of pressure on the wave-length can be further minimized by the correct choice of the line used as the standard. In general, the wave-lengths due to the triplet $P-S$ transitions are less susceptible to pressure than the $P-D$ transitions. The pressure effect, furthermore, always increases for lines where one of the levels involved is near the ionization limit. Self-reversal is most apt to occur where the ground state or low metastable states are concerned. The choice for a line, therefore, resolves itself into selecting a transition in the middle of the atomic energy diagram. Such a compromise is represented by the $^1P_1-{}^1D_2$, 6438A of cadmium and $^3P_2-{}^3S_1$, 5461A of mercury. Thus the most suitable cadmium line is in the red region, which results in low visual intensity. In order to increase the intensity for spectrographic work, the lamp must be viewed end-on and this reduces the field of view. Lastly, the red cadmium line possesses a hyperfine structure which, although it has never been resolved[7-9] increases the width of the fringes obtained with it.

Mercury has most of the properties that would make it a good choice for the standard. The green line has a very high visual intensity, is easily excited, and satisfies the conditions as to self-reversal and pressure effect as well as does the red cadmium line. The only difficulty is that the lines of mercury all show hyperfine structure as well as isotope shift.[10] Seven isotopes are present but since the isotope of mass 202 is the most plentiful, one might be inclined to try to use the mass spectrograph for separating this isotope from ordinary mercury. This and all other methods for separating the mercury isotopes in quantities large enough to produce a strong light source have until now been unsuccessful.

In 1939, W. E. Williams[11] suggested the utilization of the known nuclear reaction[12]

$$_{79}\mathrm{Au}^{197}+{}_0n^1\rightarrow{}_{79}\mathrm{Au}^{198}$$
$$_{79}\mathrm{Au}^{198}\rightarrow{}_{80}\mathrm{Hg}^{198}+{}_{-1}\beta^0$$

for the production of the mercury isotope of mass 198, using neutrons from the cyclotron. Schüler and Keyston[10] had shown that the even isotopes of mercury have no hyperfine structure and hence that any of the even isotopes of mercury would be suitable for use in a lamp designed to furnish the primary standard of wave-length.

Fermi and his associates had used as their source of neutrons a mixture of beryllium powder and radon, a source which produced approximately 800,000 neutrons per second. When the present experiment was started in 1939, a much more copious source of neutrons was available in the cyclotron, neutrons being the by-product of almost every nuclear reaction produced by it. The manufacture of the mercury isotope of mass 198 therefore was accomplished by utilizing the stray neutrons existing near the cyclotron as a result of other bombardments.

OBJECTIVES

The purposes of this experiment were:

1. To verify, by spectrographic methods, the nuclear transformation $_{79}\mathrm{Au}^{197}+{}_0n^1\rightarrow{}_{80}\mathrm{Hg}^{198}+{}_{-1}\beta^0$. This was to be done by observing the spectrum of the product of the reaction.

2. To show that the resulting mercury isotope is the isotope of mass 198. Inasmuch as the odd isotopes are responsible for the hyperfine structure and the even isotopes yield simple spectral lines, this had to be accomplished by observing the lines with a Fabry-Pérot etalon. The lines should be simple lines and should coincide with the lines in ordinary mercury which had previously been assigned[13] to the isotope of mass 198.

3. To produce a sufficient amount of the single isotope of mercury to show that it is feasible to develop a lamp for use as a primary standard of wave-length.

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[8] H. Westmeyer, Zeits. f. Physik **94**, 590 (1935).
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[10] H. Schüler and J. E. Keyston, Zeits. f. Physik **72**, 430 (1931).
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[13] H. C. Burger and P. H. Van Cittert, Physica **5**, 177 (1938).

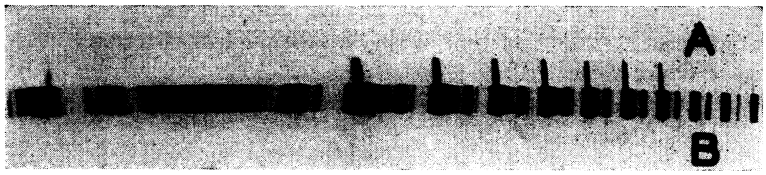

FIG. 1. Fabry-Pérot etalon spectrogram of 5461A. *A*, mercury 198. *B*, ordinary mercury. Etalon separation, 9.015 mm.

PRELIMINARY CALCULATIONS AND EXPERIMENTS

In order to determine the feasibility of the nuclear transformation of gold into the mercury isotope of mass 198, one ounce of gold was exposed to neutrons in the vicinity of the 37-inch cyclotron. After four days the gold was found to have an activity of 2.4 millicuries. From this and the mean life of radioactive gold, a simple calculation indicated that the sample of gold would produce 3.45×10^{13} atoms of mercury in equilibrium, and that this amount of mercury would yield a pressure of 3.16×10^{-3} mm Hg in a tube having a volume of 0.3 cm^3.

Williams[11] reported that in his experiments he had obtained an electrodeless discharge in tubes containing mercury vapor at a pressure of 10^{-4} to 10^{-5} mm Hg, provided that an oscillator having a frequency of approximately 50 megacycles was used. Extensive experiments showed that, while this was true, a much higher vapor pressure is necessary if the tube is to be sealed off, due to the adsorption of mercury on the walls of the tube. An initial vapor pressure of the order of 10^{-2} was found satisfactory for a discharge tube of the size and type used in this experiment.

When the vapor pressure of the mercury is between 10^{-2} and 10^{-5} mm, the addition of a small quantity of argon increases the intensity of the discharge by a factor of at least 10. The first excitation potential of mercury is lower than the metastable state of argon. The argon atoms yield the energy of their metastable states to the mercury atoms in inelastic collisions, thus facilitating the excitation of mercury. An argon pressure of the order of 4 mm was found to be satisfactory, and very little difference was noted between the effects with 2 mm and 6 mm.

In order to determine if it would be feasible to separate completely any mercury produced from the parent gold, and to find whether it would be possible to completely purify the gold from any mercury contamination that might occur during processing, a drop of mercury was placed on a piece of gold foil. The gold was then sealed in a quartz tube attached to a vacuum system and the gold was heated in an electric furnace to about 900°C, very near to its melting point. Enough mercury vapor was found in the glass tubing between the gold and the first liquid air trap to be excited by means of a leak-tester. Heating the glass walls of the tube with a gas-oxygen torch did not satisfactorily remove the mercury.

An oscillator having a frequency of 100 megacycles was then attached to the glass tubing by wrapping the radiofrequency output leads around the tubing at points about a foot apart. A discharge formed along the length of the tubing, and enough high frequency energy was supplied to heat the glass close to its softening point. The heating effect was greatly increased by the addition of pure argon at a pressure of about 5 mm. The outgassing cycle consisted of admitting argon, operating the oscillator for five minutes, and then pumping out the gases while the oscillator was still operating. Fifteen to twenty such cycles were sufficient to outgas the system so completely that no trace of mercury could be found. Heating the glass walls by means of a gas-oxygen torch did not release any further mercury or other foreign gases.

EXPERIMENTAL PROCEDURE

A one-ounce sheet of pure gold foil 0.1 mm thick was chemically treated to remove surface contamination, sealed into a glass tube, and attached to the vacuum system. The gold was then placed in an electric furnace and heated to approximately 200°C to remove all traces of water and other easily volatile substances. The temperature of the furnace was then raised to about 450°C, or to a point where the glass showed a dull red in total darkness, and during the pumping process the outgassing oscillator was

operated so that a discharge appeared in the entire region between the gold and the first liquid air trap. Twelve to twenty hours of pumping were required to outgas the system so completely that the pressure remained at 10^{-5} mm of Hg for an hour after the system was isolated from the pumps.

The gold was then transferred to a quartz tube in order to heat it to just below its melting point. The outgassing oscillator and argon were used as before until the pressure remained at 10^{-6} mm of Hg for an hour after the system was shut off. At this time a 15-cm section of 4-mm i.d. quartz tubing was sealed to the far end of the large quartz tube containing the gold. Since the mercury produced by the reaction is to be distilled into this tube, sealing it on at this time prevents impurities from distilling into the section during the outgassing period.

After the complete system had been outgassed, a pressure of 4 mm of spectroscopically pure argon was admitted and the unit was sealed off. The complete assembly was placed in a suitable box and embedded in paraffin before being placed near the 60-inch Crocker cyclotron. The paraffin served to slow down the fast neutrons as well as to cushion the quartz tube during handling.

When the run was completed, the quartz tube was removed and the gold was again heated to just below the melting point while the far end of the 4-mm i.d. quartz tube was cooled to liquid air temperature by means of a specially constructed Dewar flask. After two to three hours, a 6-cm section was sealed off and this contained, besides argon, most of the mercury that had been produced. The mercury in this tube was then excited by means of the 100-megacycle oscillator, with the power adjusted so that a satisfactory intensity was obtained.

EXPERIMENTAL RESULTS

The first successful experiment was the result of exposing the one-ounce sample of gold to neutrons in the vicinity of the 60-inch cyclotron for one week. The discharge in the tube was examined with a constant deviation spectrometer and mercury lines were found. After about one minute the discharge decreased in intensity and soon disappeared.

The next tube was the result of one month's exposure to the same neutron source. Enough mercury was obtained from this bombardment to make it possible to photograph a Fabry-Pérot etalon spectrogram.[14] The mercury lines were found to be simple lines, and the absence of hyperfine components showed that the mercury is actually a transmutation product. This source operated successfully for about five minutes.

The gold was then left near the cyclotron for ten months, during which time the cyclotron was operated almost continuously. The mercury produced by this neutron bombardment was sufficient so that the discharge tube could be run for thirty minutes with no loss in intensity.[15] During this thirty minutes of operation, the spectrograms in Figs. 1–3, as well as some ten others not given here, were taken. The intensity of the discharge was so large that fifteen to thirty seconds were sufficient for a normal exposure.

ANALYSIS OF THE SPECTROGRAMS

The spectrograph used in this experiment had two large flint glass prisms, and the quartz Fabry-Pérot etalon was placed between the collimator lens and the prisms. The jaws of the slit were controlled by screws mounted horizontally on each side of the slit. This made it possible to close the slit from one side after one exposure and to open the slit again from the other side for the second exposure. The two spectrograms are then side by side and no adjustment has been made that would affect the position of the interference patterns on the photographic plate. In all cases, the center of the interference rings was so aligned that it fell on the line between the two spectrograms, since this arrangement is best for comparing two spectrograms.

The line 5461 was studied, as it is the one most suitable for a primary standard of wavelength. Figure 1 shows the etalon spectrogram of Hg^{198} and of ordinary mercury, the latter having been excited in an air-cooled d.c. arc. The positions of all the lines appearing in the spectrogram of ordinary mercury were measured on the original plates by means of a com-

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[15] J. H. Wiens, Phys. Rev. **65**, 58 (1944).

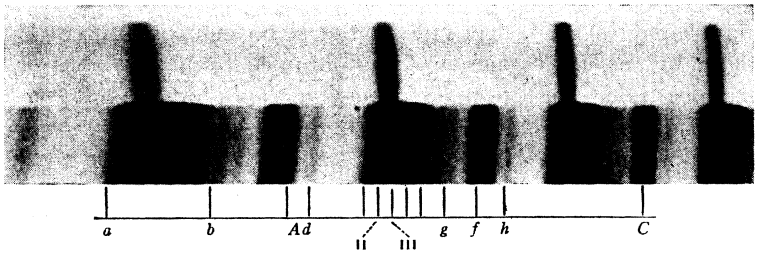


FIG. 2. Resolution of 5461A with designations as given by Schüler.

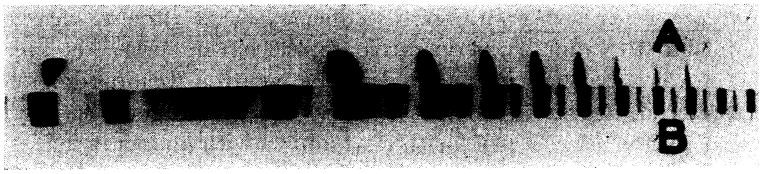


FIG. 3. Same as Fig. 1, except that *A* is more heavily exposed.

parator. From these measurements the wave-number separations were calculated, and by comparing them with the separations given by Burger,[13] assignments were made according to the notation originally given by Schüler and Keyston.[10] Figure 2 is an enlargement of Fig. 1 and these assignments are indicated.

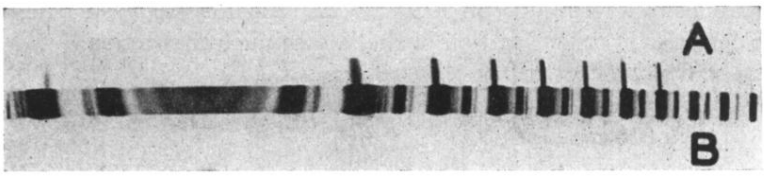
The position of the line due to Hg^{198} was computed from the measurements, and the average of several values gave its position as -33×10^{-3} cm^{-1}. The wave number separation of line III due to Hg^{200} and line II due to Hg^{198} is given[13] as -29×10^{-3}cm^{-1}. Because of the large experimental error in determining the center of the broad component, which contains the five main lines, this difference is within the experimental error. Thus the line 5471A obtained from mercury that is produced by the nuclear transformation of gold coincides, within the experimental error, with the line due to Hg^{198} as assigned by Schüler and other observers. This also verifies the correctness of the mass number of the stable daughter element of radioactive gold as predicted from the observed β-emission of the parent substance. Figure 3 is similar to Fig. 1 except that the spectrogram due to Hg^{198} has been greatly overexposed in an effort to find any other weak lines. No additional lines could be observed in any of the spectrograms.

DISCUSSION OF RESULTS

The results obtained in this experiment indicate that it is both possible and feasible to use the nuclear transmutation of gold to mercury to produce sufficient mercury for use in developing a better primary standard of wave-length. The quantity of mercury can obviously be greatly increased by the use of a larger mass of gold, and since the reaction utilizes stray neutrons produced by the reactions in the cyclotron, no special operation of the cyclotron is required. With the development of other neutron sources many times stronger than the cyclotron, it should very soon be possible to produce enough mercury to make the more standard types of d.c. arcs. 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 from Hg^{198} as a new primary standard of wave-length.

ACKNOWLEDGMENTS

I wish to thank Professor L. W. Alvarez who first saw the feasibility of this problem and encouraged me to start the experiment. Furthermore, I am grateful for the suggestions and help that Professor F. A. Jenkins gave me during the course of the experiment. I wish to thank Professor H. E. White, under whose supervision this problem was completed, for his guidance and encouragement. Lastly, I wish especially to thank the Radiation Laboratory and the operators of the cyclotron for making it possible to bombard the gold samples during the four years occupied by these experiments.



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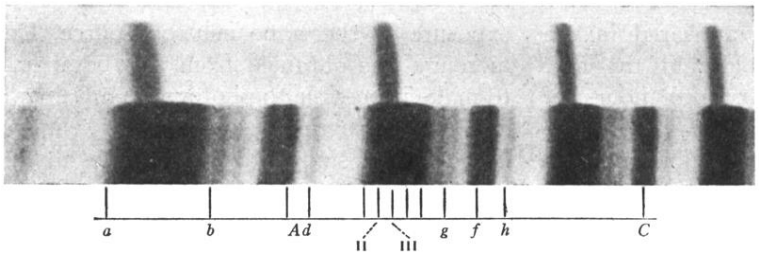

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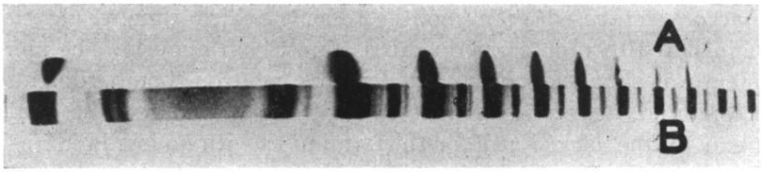

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