Report BIPM-72/2

BUREAU INTERNATIONAL DES POIDS ET MESURES

Alpha Spectrometry - Progress report

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This is a report of all the work done between October 1968 and the beginning of January 1972. These details, although partially contained in the various publications, appeared to be important enough to be collected in a special report for use in Section IV of the Consultative Committee on Standards for Measuring Ionizing Radiations.

1. Magnetic field

The gradients of the field have been reduced in the region of a radius $\rho > 43$ cm and an azimuth $45° < \varphi < 150°$. Further a diaphragm has been introduced at $\varphi = 90°$ which intercepts particles which have left the homogeneous field region. An improved line shape for ^{212}Po has been observed.

The field topography depends among other things on the "history" of magnetization. A stable state may be reached more rapidly if one first maintains the current at too high a value, then too low, and so on, approaching gradually the final value. This value of the current is 372 A in the case most frequently used (B = 0.909 T). Thus, the current was maintained for 15 or 20 minutes at 400, 350, 390, 360, 380, 365, 372 A, successively; of course, the field stabilization was put in only at 372 A.

The undesired low frequency modulation of the field has been measured with the aid of a coil with 450 turns and a mean diameter of 8 cm. This coil was connected to the input of a highly sensitive oscilloscope. Low frequency components at 50 and 300 Hz could be observed, but their amplitude was always less than 10^{-6} of the main field.

The pilot frequency of the field stabilizer (4.3 MHz) is multiplied by an appropriate integer so as to make possible the stabilization at 0.606, 0.707, 0.808, 0.909 or 1.01 T. All these multipliers have been constructed and are available as plug-in units.

One of the amplifiers of the stabilizer was found to be over-loaded. After eliminating this fault, the performance of the stabilizer improved considerably and made the alarm system almost superfluous.

The field measuring apparatus has also been improved and works now perfectly well. Frequency modulation has definite advantages over field modulation. The same device could also be used for the field direction measurements.

The moving nmr probe contains a quartz ampoule filled with a well defined aqueous solution of $MnSO_4$. The same ampoule which has been used in all the measurements published so far is now conserved separately in a safe of the BIPM and could be used for remeasurement if in the future this should become necessary. A new quartz ampoule with an optimal number of windings of the RF coil has been mounted. The fixed probe for the stabilization, however, is provided with a glass ampoule which is much simpler to manufacture and to fill.

The frequency meter has been checked with the standard wave of Droitwich and with a quartz clock of the BIPM. An amplifier was added permitting the frequency of the RF oscillators to be measured more easily.

We have measured the field degree by degree on an arbitrarily chosen semi-circle. From these measurements Hartree-corrections have been evaluated using different samples of 31 values out of 180. The dispersion of the results provides an estimate of the sampling error which is smaller than 10^{-6}. Special measurements were carried out for determining the radial field gradients and the corresponding correction. Field direction measurements were made for various values of the field.

The magnet was kept running without interruption from September 4 to December 21, 1970. This was a test of the high quality of material and installation. The manufacturer overhauled the power supply in February 1971. Since then the magnet has been running during about 4 000 h without difficulty.

2. Spectrograph, length standard

The temperature of the spectrograph can be maintained constant within a few tenths of a kelvin during each run. This is done with a closed water circuit and a HAAKE thermostat. To accomplish this it was necessary to fit the spectrograph with teflon supports and to improve the isolation of the outer circuit. A leak of the water jacket called for improving the soldering of a pipe. Fortunately, the heating of the spectrograph did not modify permanently the distances between the entry slit and the reference markers. These distances have been repeatedly measured with the aid of the universal comparator and the standard bar 122R. The latter has in turn been compared with another standard ruler (SIP 129 24), the length of which is known directly in terms of the ^{86}Kr wavelength. In July 1971 the distances have been remeasured. No significant change was found. The markers at the four ends of the plate holder have been turned so as to give longitudinal marks on the plates.

Further, various measurements have been carried out for evaluating the effective thermal expansion coefficient of the spectrograph and the change in length due to the water pressure while in vacuum.

Finally, a complete calculation has been carried out for determining the effective solid angle, i.e. the ratio of the number of observed particles used in the extrapolation and the number of particles emitted by the source.

3. The cooling system of the magnet

In order to diminish the water consumption, an evaporation cooling system was installed in October 1968. However, even after a thorough cleaning of the external circuit, the cooling was insufficient. Finally it was recognized that the heat exchanger was very inefficient. The manufacturer replaced it by a better construction (October 1969). Since then, the cooling was satisfactory until March 1971. Careful rinsing of the magnet coils and the replacement of a part of the outer circuit tubing made the cooling even more efficient than at the beginning.

4. Vacuum system

The turbo-molecular pump PFEIFFER has been washed internally with freon (TF 113). The belts have been replaced by new ones and the motor has been fixed so as to reduce the vibrations interfering with the field stabilization. The sudden appearance of a leak in the water circuit of the spectrograph made necessary a complete cleaning of the pumping system (October-November 1971).

A pneumatic shutter has been constructed which stops the particles when the stabilization runs with reduced sensitivity. The shutter is intended to close whenever the modulation amplitude is increased in order not to loose the proton signal. However, after repairing the stabilizer, this case became so rare that the shutter seems now almost useless.

5, Nuclear track plates

After many attempts, a recipe has been found which provides a good contrast between alpha tracks and background grains, a high detection efficiency, very small deformation of the emulsion, and a fair reproductibility. A hardener is added to the first bath and the plates are dried by immersion in alcohol solutions of increasing concentration. Much attention has been paid to the cutting of the plates, since the manufacturer refused to supply the dimensions which we need. A cutting apparatus has been constructed and gives satisfactory results.

An Abbe type comparator of ZEISS has been furnished with a NACHET microscope and an appropriate illuminating system. A dry objective (magnification 60) is used together with eye pieces magnifying 11 times.

The person who scans the plates uses a magnetophone from which he can transcribe the results later. In a few cases test scans have been made on the same plate by two different observers. The results were in excellent agreement.

6. Sources and source preparation laboratory

In a trailer outside the buildings we have installed a source preparation laboratory. The roof has been reinforced and protected against bad weather. An evaporator for α-sources has been installed and the first sublimated source was prepared in May 1969.

The trailer was first located beside the building for neutron measurements. However, the two ^{228}Th sources (70 mCi, together) gave too large and variable a γ-background for certain experiments currently carried out in the neutron building. Thus we decided to move the trailer to another place. Since October 1969 it has been located beside the big hall for X-ray measurements. In January 1971 the trailer was cleaned inside and much of the contamination was eliminated.

Several laboratories have sent samples of α-emitters and let us benefit from their competent help and advice.

- The Institute of Atomic Energy I.V. Kourtchatov (Moscow) sent us solutions of

$$\left.\begin{array}{ll} & 4.5 \text{ mCi of } {}^{244}Cm \\ \text{and} & 2.6 \text{ mCi of } {}^{238}Pu \end{array}\right\} \quad \text{(October 1969).}$$

-The Lawrence Radiation Laboratory (Berkeley, Calif.) sent

$$\left.\begin{array}{l} 4.5 \text{ mCi of } {}^{242}Cm \\ 18 \text{ mCi of } {}^{228}Th \end{array}\right\} \quad \text{(November 1967)}$$

$$3 \text{ mCi of } {}^{253}Es \quad \text{(May 1970)}$$

$$\left.\begin{array}{l} 1 \text{ mCi of } {}^{232}U \\ (\text{free of } {}^{228}Th) \\ 10\,\mu Ci \text{ of } {}^{240}Pu \end{array}\right\} \quad \text{(March 1971).}$$

- The Centre of Nuclear and Mass Spectroscopy (Orsay) prepared for us about ten sources of various natural α-emitters. It lent us an activation vessel for ^{222}Rn and prepared the solutions of ^{242}Cm and ^{253}Es from LRL.

- The Union Minière (Brussels) lent us an emanating ^{226}Ra preparation (50 mCi).

Further, we bought solutions of ^{242}Cm, ^{226}Ra and ^{241}Am from Amersham and from Saclay.

A special apparatus has been constructed for using the emanating ^{226}Ra source and for extracting the radon. Thus sources containing ^{218}Po and ^{214}Po have been prepared.

The first experiments were carried out with the thorium active deposit and with ^{242}Cm (Summer 1968). However, these results were wrong because of an incorrect source position. Therefore, a new source-holder was constructed. It is equipped with a sweeping mechanism which slowly moves the source, during the runs, from side to side a few tenths of a mm, in order to eliminate irregularities of the source. This movement is obtained by a piezo-electric bar to which a potential is applied varying linearly in time between + 200 V and - 200 V. This construction is very fragile. Several bars have been broken during source changing. However, we could not find a better yet simple solution.

The mean position of the source had to be aligned with the entry slit. A solid state detector with a slit was first fixed behind the entry slit. The two slits were adjusted to lie on a line perpendicular to the spectrograph axes and the spectrograph was put in its normal position. A strong source was fixed on the source-holder and the vacuum chamber was evacuated. Now the position of the source was varied electrostatically and the count rate was observed without magnetic field. Taking into account the curvature of the trajectories in the field, the optimal position of the source could be determined.

7. Results, publications

A complete description of the experimental set-up as well as all the results obtained before 1971 (20 α-emitters, 31 energy values) have been reported in

- B. Grennberg and A. Rytz, Absolute measurements of α-ray energies, Metrologia 7, 65-77 (1971).

Two further papers describe special problems in the stabilization and the measurement of the magnetic field:

- P. Bréonce and B. Grennberg, A proton resonance magnetic field stabilizer using a quartz stabilized reference frequency, Nucl. Instr. Meth. 84, 83-89 (1970),

- B. Grennberg and A. Rytz, Corrective terms to the Hartree correction formula, Nucl. Instr. Meth. 84, 83-89 (1971).

The result concerning ^{242}Cm, published with reservations in "Procès-Verbaux des séances du Comité International des Poids et Mesures", 2e série, 36, p. 77 (1968), is low by 1.6 keV for reasons explained above. Unfortunately it has already been quoted in "Nuclear Data Sheets".

The first accurate results (^{212}Bi) obtained in 1969 and those of ^{253}Es have also been published in "Comptes Rendus des séances de l'Académie des Sciences de Paris",

- B. Grennberg et A. Rytz, Nouvelles déterminations absolues de l'énergie de particules α, C.R. B-269, 652 (1969),

- B. Grennberg, A. Rytz et F. Asaro, Mesure absolue de l'énergie α du ^{253}Es, C.R. B-272, 283 (1971).

Most of the measurements were carried out between 1st July and 30th November 1970, during which period 43 runs were made.

Further measurements on ^{212}Bi and ^{212}Po

After the departure of B. Grennberg in January 1971 and the arrival of D. Gorman on 1st February 1971 a series of new measurements were made on the nuclides ^{212}Bi and ^{212}Po in order to try to improve the precision of these measurements, and as a check on our method. Determinations of the ^{212}Bi were made at fields of 0.808 and 0.909 T and the results are given below along with the earlier values of B. Grennberg.

α Energies of ^{212}Bi (keV)

	B. Grennberg	New determinations	
	0.9 T	0.8 T	0.9 T
α_0	6 090.06 ± 0.08	6 090.34 ± 0.11	6 089.99 ± 0.12
α_{40}	6 050.77 ± 0.07	6 051.06 ± 0.07	6 050.75 ± 0.07
ΔE_α	39.29	39.28	39.24

Three exposures were made with the ^{212}Po at a field of 1.01 T, and the weighted mean value obtained was 8 784.30 ± 0.07 keV. The precision is approximately a factor of 10 better than that previously reported.

Three more papers have been written since:

- A. Rytz, B. Grennberg and D.J. Gorman, New alpha energy standards, Proceedings of the 4th International Conference on Atomic Masses and Fundamental Constants, Sept. 6 to 10, 1971, Teddginton, England; in the press,

- D.J. Gorman, A. Rytz et F. Asaro, Mesures absolues de l'énergie de particules α émises par le ^{232}U et le ^{240}Pu; preprint, to be submitted to C.R. Acad. Sci. Paris, 1972,

- A. Rytz, A compilation of recalibrated α -particle energy and intensity values, 1972, B.I.P.M.