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# Avogadro, Planck and the kilogram redefinition

E. Massa

## Abstract

In 2018, the General Conference on Weights and Measures reformed the International System of Units by adopting fixed values for some fundamental constant of physics. The kilogram, a unit previously defined by a material artefact, is now established and realised from the knowledge and the technology of the modern world.

## 1 A brief history of the kilogram

The physicists' mass appears firstly in Newton's Principia. In Definition I - *quantitas materiae est mensura ejusdem orta ex illius densitate et magnitudine conjunctim* - Newton grounded the mass on the concept of density, which was assumed primitive.

In 1790 a group of experts, appointed by King Louis XVI, established a system of measurement units which would subsequently be a foundation of the International System of Units. The proposed length and mass units were based on fixed values of the length of the Paris meridian and the mass of a cubic decimeter of water (at the melting temperature of ice and at atmospheric pressure). Finally, two platinum artefacts representing the metre and the kilogram were manufactured and stored in the *Archives de la Republique* in 1799.

On 20<sup>th</sup> of May 1875, the representatives of seventeen nations signed an international treaty, "The Metre Convention" [1], which established the International Bureau of Weights and Measures (BIPM). Subsequently, at the 1<sup>st</sup> General Conference on Weights and Measures (CGPM) in 1889 were sanctioned and declared the first definition of the kilogram: "this prototype shall henceforth be considered to be the unit of mass". In 1901, the 3<sup>rd</sup> CGPM confirmed that: "the kilogram is the unit of mass; it is equal to the mass of the international prototype of the kilogram". This declaration intended to end the ambiguity in the widespread usage of the word "weight". With this definition followed that the mass of the international prototype of the kilogram (IPK) was always 1 kilogram exactly or  $m(\mathfrak{K}) = 1 \text{ kg}$ .

The international prototype of the kilogram is a platinum-iridium cylinder kept by the International Bureau of Weights and Measures at Sèvres, France. The reference mass thus defined was used to calibrate national standards of platinum-iridium alloy.

Verifications have been carried out in 1889, 1946, and 1991. They show a drift of up to 50  $\mu\text{g}$  per century between the mass of the IPK (which was 1 kg by

definition) and the mass of some of the official copies (*témions*) [2,3]. It cannot be determined how much of this mass change could be due to the IPK itself. The continued dependence of the kilogram definition on a physical prototype made metrologists uneasy. Although the international prototype served science and technology well, it had limitations as a material artefact:

- it is not linked to an invariant of nature and hence could be changed physically;
- it collects contaminants on its surface and must be washed before use;
- it is only accessible at the BIPM and could not be used routinely because of the risk of mechanical wear.

As shown in fig.1, the *témions* were re-measured in 2014, after cleaning and washing, during the calibration campaign against the IPK in anticipation of the redefinition of the kilogram (Extraordinary Calibrations) [4, 5]. As a consequence, the BIPM “as-maintained” mass unit was found to be offset by 35  $\mu\text{g}$  to the IPK. This correction evidenced a further weakness of the kilogram definition based on an artificial prototype.

In the framework of the international efforts to redefine the International System of Units according to our understanding of the world, the Kibble balance (formerly watt balance) and the X-Ray Crystal Density method (XRCD, also called the Avogadro project) produced the most accurate determinations of the Avogadro and Planck constants.

## 2 Mass metrology

In 2005, a paper published in the international journal *Metrologia* declared that the “time for the redefinition of the kilogram had come” [6]. At the same time, the Consultative Committee for Mass and Related Quantities (CCM) of the International Committee for Weights and Measures (CIPM) presented recommendations for the conditions to be met before the redefinition.

These events started the process for the changing of the International System of Units. In 2005, one of the main difficulties to redefine the kilogram was the discrepancy of 1 part in  $10^6$  between the measurement results obtained from the Kibble balance and the XRCD experiments. The mass community expected both agreement and low uncertainties of the measurements. To ensure continuity to mass metrology, any change of the kilogram definition must be invisible to the users. Also, all the already existing mass and mass related data and measurement results had to keep the same numerical value when expressed in the new unit. Therefore, the CCM, in its 15<sup>th</sup> meeting, made the Recommendation G1 (2013) “On a new definition of the kilogram” sanctioning that:

1. at least three independent experiments, including work from watt balance and XRCD experiments, yield consistent values of the Planck constant with relative standard uncertainties not larger than 5 parts in  $10^8$ ,

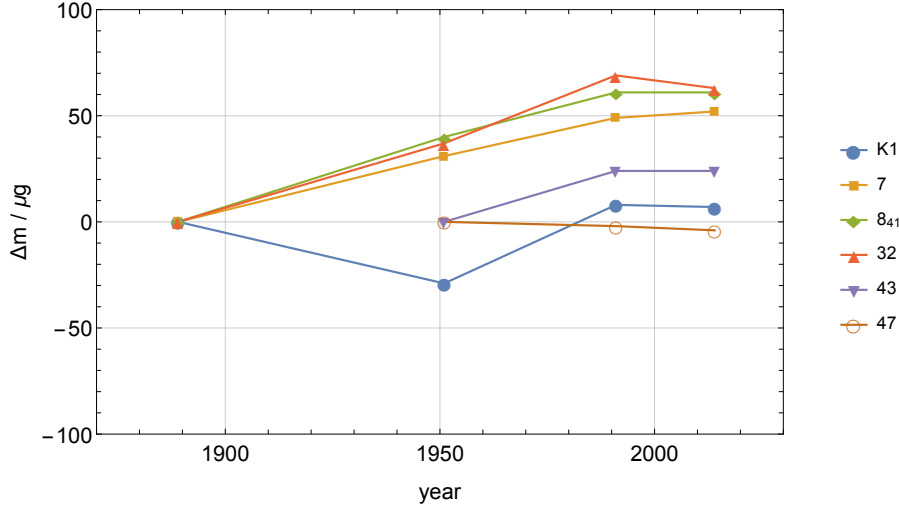


Figure 1: Mass variation of the official copies K1, 7, 8<sub>41</sub>, 32, 43, 47 with respect to the mass of the international prototype of the kilogram. The official copy number 8 was erroneously marked as 41. This comparison has been carried out four times, in 1889, 1946, 1991 and 2014.

2. at least one of these results should have a relative standard uncertainty not larger than 2 parts in  $10^8$ ,
3. the BIPM prototypes, the BIPM ensemble of reference mass standards, and the mass standards used in the watt balance and XRCD experiments have been compared as directly as possible with the international prototype of the kilogram,
4. the procedures for the future realization and dissemination of the kilogram, as described in the mise en pratique, have been validated in accordance with the principles of the CIPM-MRA (Mutual Recognition Arrangement).

These requirements were met at the time of the 16<sup>th</sup> meeting of the CCM in 2017. The scientific path for the redefinition of the kilogram was thus fulfilled. The CGPM, at the 26<sup>th</sup> meeting, considered the recommendations of the CIPM and reformed the International System of Units. The kilogram, ampere, kelvin, and mole, previously defined in terms of material artefacts or physical properties of matter, were re-defined by adopting fixed values for some fundamental constant of physics. Today, the new definition of the kilogram is as follows:

**The kilogram, symbol kg, is the SI unit of mass. It is defined by taking the fixed numerical value of the Planck constant  $h$  to be**

$h = 6.62607015 \times 10^{-34}$  when expressed in the unit J s, which is equal to  $\text{m}^2 \text{kg s}^{-1}$ , where the metre and the second are defined in terms of  $c$  and  $\Delta\nu_{Cs}$ .

So the Planck constant  $h$  is precisely  $h = 6.62607015 \times 10^{-34}$  J s or  $\text{m}^2 \text{kg s}^{-1}$ , where the second is defined by fixing the frequency of the light emitted in a transition of the caesium 133 atoms and the meter by fixing the speed of light. The numerical value of the Planck constant sets the size of the kilogram. Today, the mass of the IPK is 1 kg with a relative uncertainty equal to the recommended value of the Planck constant just before the redefinition [7]. In the future, the mass of IPK will be determined experimentally.

### 3 The Planck constant – CODATA 2017 adjustment

The stipulated value of the Planck constant is the result of the special least-squares adjustment (LSA) of the values of the fundamental physical constants provided by the Committee on Data for Science and Technology (CODATA) through its Task Group on Fundamental Constants (TGFC). The TGFC periodically provides the scientific and technological communities with a self-consistent set of internationally recommended values of the basic constants and conversion factors of physics and chemistry [8–14]. The fig.2 shows the values of the Planck constant inferred from both the Kibble balance and XRCD experiments. The square dot is the recommended CODATA 2017 [15,16]. This figure also summarises the best determination of the Avogadro and Planck constants.

### 4 The *mise en pratique* of the kilogram

Experts of the CCM prepared a document, the *mise en pratique*, indicating how the definition of the kilogram is realised in practice [17]. The definition does not imply any particular experiment for its practical realisation: any procedure capable of deriving a mass value traceable to the set of defined fundamental constants realises the kilogram. Therefore, the *mise en pratique* is not an exhaustive list of recommended primary methods having the highest metrological properties.

The two experiments described in the following, the Kibble balance [18–33] and the XRCD method [34–40], are the state-of-the-art to realise the unit of mass at the one-kilogram level. These experiments played a central role in the redefinition of the kilogram, in the determination of both Avogadro and Planck constants, and allowed reforming the entire International System of Units.

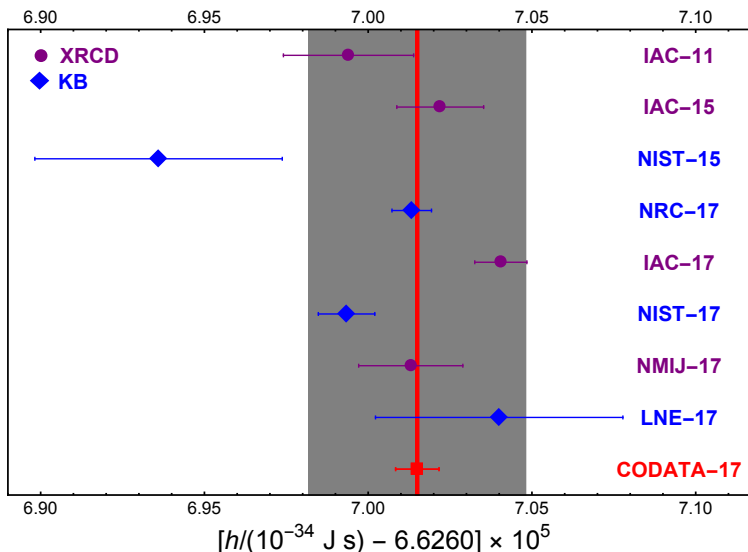


Figure 2: Values of the Planck constant  $h$  used by CODATA 2017 special adjustment. The inner grey band is 5 parts in  $10^8$ . KB: Kibble balance; XRCD: X-Ray-Crystal-Density method.

#### 4.1 The Kibble balance - $h$ measurement

The direct way of access to the  $h/m$  ratio, where  $m$  is the mass of a prototype, is by a Kibble balance. The Kibble balance was first proposed in 1975 by Dr Bryan Kibble (1938–2016), a metrologist at the National Physical Laboratory (NPL) in the UK. It was developed to realise the unit of current, the ampere, and, after the discovery of the quantum Hall effect, paved a new way to the measurement of the Planck constant. This experiment compares virtually the mechanical and electrical powers produced by the motion of a mass in the Earth’s gravitational field and by the motion of the supporting coil in a magnetic field. As shown in fig.3, the comparison is carried out in two steps.

In the first step, static or force mode, a balance is used in substitution mode, to compare the prototype weight,  $mg$ , with the Lorentz force generated by the interaction between the electrical current  $I$  flowing in a coil having length  $L$  immersed in an external magnetic field  $B$ . Hence, by leaving out a vector notation,

$$mg = BLI. \quad (1)$$

In the second step, dynamic or velocity mode, the coil is moved and the induced electromotive force is measured at its ends. The equation that links the coil velocity  $u$  to the electromotive force  $\mathcal{E}$  is

$$\mathcal{E} = BLu. \quad (2)$$

The geometric term  $BL$  appearing in both (1) and (2) cannot be measured

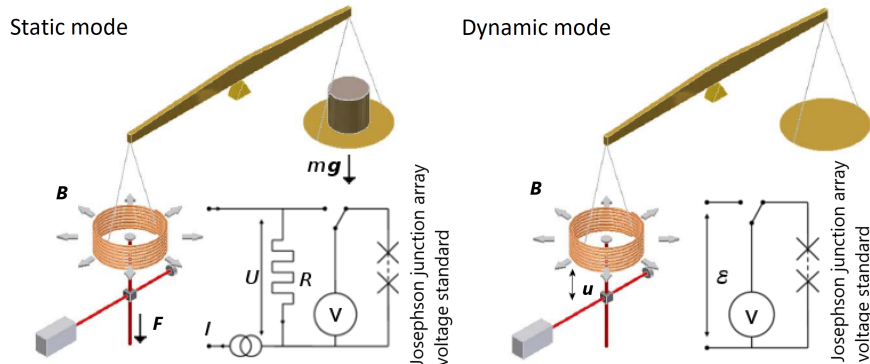


Figure 3: Scheme of a Kibble balance. Static mode: the force  $F$  acting on the current-carrying coil is balanced against the weight  $mg$  of the test mass. The current  $I$  flowing in the coil is measured in terms of Josephson voltage and quantum Hall resistance. Dynamic mode: the coil is moved at velocity  $u$  in the vertical direction through the magnetic field  $B$  and the induced voltage  $\mathcal{E}$  is measured in terms of Josephson voltage.

with  $10^{-8}$  fractional accuracy. By eliminating it, we obtain the so-called watt equation

$$mgu = \mathcal{E}I. \quad (3)$$

This name stems from the fact that both the mechanical (on the left-hand side) and the electrical (on the right-hand side) powers are measured in the watt unit. The electromotive force is measured via the Josephson effect  $\mathcal{E} = n_1(h/2e)\nu_1$ , where  $n_1$  is an integer and  $\nu_1$  is the frequency irradiating the device. The current measurement,  $I = U/R$ , is based on both the Josephson and the quantum Hall effects. Hence,  $U = n_2(h/2e)\nu_2$ ,  $R = r(h/e^2)$ , where  $n_2$  is an integer,  $r$  is a real number and  $\nu_2$  is again a frequency irradiating the Josephson device. Eventually, the  $h/m$  ratio is obtained in units of the international system from

$$\frac{h}{m} = \frac{4r}{n_1 n_2} \frac{gu}{\nu_1 \nu_2}. \quad (4)$$

The measurement equation (4) relates mass to the Planck constant. All the quantities in the right-hand side are measured with uncertainties small enough to give the  $h/m$  ratio to within a relative uncertainty of  $1 \times 10^{-8}$ . In the practical execution of the measurement, there are several other sources of uncertainty, alignments, unwanted motion, parasitic forces and torques. A detailed description of how they are made harmless can be found in [41].

## 4.2 Counting $^{28}\text{Si}$ atoms – $N_A$ measurement

In 1811, Amedeo Avogadro, (9 August 1776 - 9 July 1856), introduced the molecular hypothesis to explain the Gay-Lussac's observations. The molecule

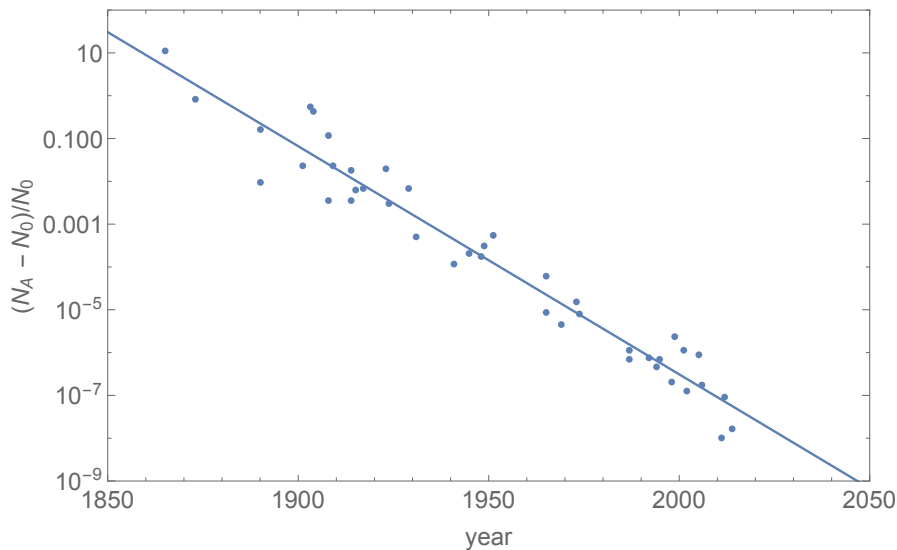


Figure 4: Reduction of the  $N_A$  uncertainty from Loschmidt’s measurement (1865) to today. The data are from [11–14, 43]; the reference value,  $N_0 = 6.02214076 \times 10^{23} \text{ mol}^{-1}$  is from CODATA 2017 special adjustment [15].

concept (a small amount of substance) allowed him to advance the hypothesis that “equal volumes of gases under the same conditions of temperature and pressure will contain equal numbers of molecules”. This statement is known as Avogadro’s law and sometimes referred to as Avogadro’s hypothesis or Avogadro’s principle. In tribute to him, the number of elementary entities (atoms, molecules, ions or other particles) in one mole of a substance is known as the Avogadro constant and represented by  $N_A$  symbol.

The Avogadro constant is a fundamental physical constant that relates a quantity at the atomic scale to its corresponding macroscopic amount. Its determination fascinated generation of scientists and triggered the developments of many different experiments. Reviews of the  $N_A$  measurements can be found in [42, 43]. As shown in fig.4, in the last century, the accuracy of the  $N_A$  values increased by about five orders of magnitude.

The first value (1865,  $N_A = 72(1) \times 10^{23} \text{ mol}^{-1}$ ) was a fallout of Loschmidt’s estimates of the diameter and mean free path of air molecules [44]. Other estimates were proposed by Planck (1901,  $N_A = R/k = 6.16(1) \times 10^{23} \text{ mol}^{-1}$ ), from the black-body determination of Boltzmann and gas constants [45]; Millikan (1917,  $N_A = F/e = 6.064(6) \times 10^{23} \text{ mol}^{-1}$ ), from the Faraday constant and his measurement of the electron charge [46]; Rutherford (1909,  $N_A = 6.16(6) \times 10^{23} \text{ mol}^{-1}$ ), from the rate of production of helium in the radium decay [47]; du Noüy (1924,  $N_A = 6.004(8) \times 10^{23} \text{ mol}^{-1}$ ), from the estimate of the size of molecules in mono-layers films on the water surface [48]. The numbers in brackets are the



value of the uncertainties referred to the last digits of the measurement results.

A breakthrough in the  $N_A$  measurement was the determination of X-ray wavelengths by diffraction by calibrated gratings [49]. Before this, the X-ray wavelength  $\lambda$  was determined via crystal diffraction and the Bragg law

$$n\lambda = 2a \sin(\Theta_B), \quad (5)$$

where the diffraction order  $n$  is an integer,  $\Theta_B$  the diffraction angle, and the lattice parameter  $a$  the spacing of  $\langle 100 \rangle$  diffracting planes that, following Bragg [50] and assuming a cubic cell, was calculated as

$$a = \sqrt[3]{\frac{qM}{\rho N_A}}, \quad (6)$$

where  $q$  is the number of atoms per unit cell, and  $M/\rho$  is the molar volume, and  $M$  and  $\rho$  are the mean molar mass and density. Grating diffraction allowed  $N_A$  to be determined by reversing (6) and (5).

In the years from 1925 to 1965, the lattice parameter measurements were carried out via (5), where ruled gratings calibrated the X-ray wavelength  $\lambda$ . The principle of this calibration is the same as for the diffraction of visible light, but the diffraction angles are tiny, only a fraction of a degree. The limiting factor was that of measuring these small angles to a high degree of accuracy.

From 1930 onwards, instead of determining the X-ray wavelengths from the lattice parameter and the lattice parameter from the  $N_A$  and molar-volume values, as Bragg did,  $N_A$  was calculated from the measured values of the lattice parameter and molar volume. Since crystallisation acts as a low-noise amplifier and makes the lattice parameter accessible to macroscopic measurements, atoms were counted by exploiting their ordered arrangement.

Historically, a remarkable attempt to count the atoms was made by Johann Magnenus, who, in 1646, used the observation that the scent of burned incense chickpea fills a church to propose a primitive estimate of the atom size. From the ratio between the church volume and that of the minimal amount of evenly dispersed incense that one can sense, which he estimated to be to one-thousandth of a chickpea and guessed to contain at least a million atoms, Magnenus arrived at the lower bound of the number of atoms in an incense chickpea  $7.776 \times 10^{17}$  [51, 52]. This result is the first example of estimating the number of atoms in a macroscopic volume.

The development of the semiconductor industry made macroscopic crystals available, and in 1963, Egidi thought about realising an atomic mass standard by counting the atoms in a crystal scaled to result in the form of a cube with its faces parallel to the lattice planes [53]. The technology of his time was unable to do that to the necessary accuracy, but, in 1965, Bonse and Hart operated the first X-ray interferometer, paved the way towards absolute measurements of the atom distance in crystals [54], and bypassed the accuracy limitations due to the X-ray wavelength calibration via artificial gratings.

Provided the crystal and the unit cell volumes are measured, and the number of atoms per unit cell is known, the counting requires their ratio to be calculated

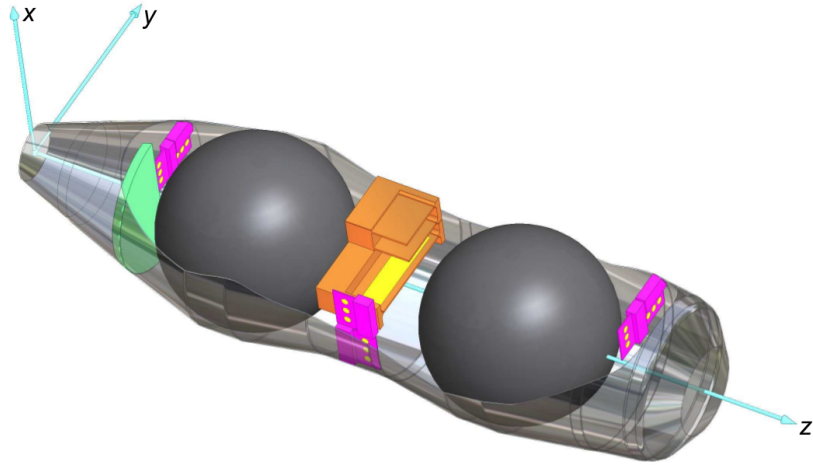


Figure 5: Sketch of the float-zone AVO28 crystal showing the two spheres S5 and S8. The enrichment is higher than 0.99995. In the middle (orange): X-ray interferometer for the lattice spacing determination (in the yellow rectangular area). From seed to tail (purple and green): samples for lattice-parameter homogeneity and lattice strain tests.

as

$$N_A = \frac{8M}{\rho a^3}, \quad (7)$$

where 8 is the number of atoms per unit cell, and  $M$ ,  $\rho$  and  $a$  are the molar mass, density and lattice parameter of the crystal.

Deslattes completed the first count of the atoms in a silicon crystal and determined the Avogadro constant (1974,  $N_A = 6.0220943(63) \times 10^{23} \text{ mol}^{-1}$ ) [55]; further measurements soon followed [56–60]. These measurements used silicon single-crystals because they are grown highly pure, large, and quasi-perfect. At the end of the last century, the measurements came to a halt because of an unsolved discrepancy of more than  $10^{-6}N_A$  to the value (1998,  $N_A = 6.02214199(47) \times 10^{23} \text{ mol}^{-1}$ ) estimated by the adjustment of the fundamental-constant [10]. Besides, insuperable difficulties impaired the accuracy of molar-mass measurements of natural silicon. The last natural-silicon measurement,  $N_A = 6.0221353(18) \times 10^{23} \text{ mol}^{-1}$ , was completed in 2005 [60].

In 2004, to get around the problem of the molar-mass measurement and following an idea outlined by Zosi in 1983 [61], an international project named

International Avogadro Coordination (IAC) <sup>1</sup> combined resources and competence to grow a silicon crystal highly enriched by <sup>28</sup>Si [62]. Isotope enrichment made accurate molar-mass measurements possible by isotope dilution mass spectroscopy combined with multi-collector inductively coupled plasma mass spectrometry.

To turn Egidi's idea into practice, two slices were taken from the enriched ingot and shaped as quasi-perfect 1 kg spheres by the Australian Centre for Precision Optics. The spheres' composition, mass, volume, density and lattice parameter were accurately determined, and their surfaces were geometrically, chemically, and physically characterised at the atomic scale. Since the molar mass, density, lattice parameter, and contaminant concentrations were not measured in the same sample, the values used in (7) had to be obtained by extrapolation. The most critical aspects of the Avogadro constant remain the perfection and homogeneity of the <sup>28</sup>Si ingot.

The most accurate values of the Avogadro constant so far obtained [40]

- $N_A$  (IAC-11) =  $6.02214095(18) \times 10^{23} \text{ mol}^{-1}$ ,
- $N_A$  (IAC-15) =  $6.02214070(12) \times 10^{23} \text{ mol}^{-1}$ ,
- $N_A$  (IAC-17) =  $6.022140526(70) \times 10^{23} \text{ mol}^{-1}$ ,
- $N_A$  (NMIJ-17) =  $6.02214078(15) \times 10^{23} \text{ mol}^{-1}$ .

were included in the CODATA 2017 adjustment.

In the revised SI, the XRCD is a primary method for the realisation of the unit of mass. As shown in fig.6, the Si-sphere mass is obtained by adding the mass of the pure-silicon sphere-core,  $m_{\text{core}}$  and that of the surface layer,  $m_{\text{SL}}$

$$m_{\text{sphere}} = m_{\text{core}} + m_{\text{SL}}, \quad (8)$$

where

$$m_{\text{core}} = Nm(\text{Si}) = \frac{8V_{\text{core}}}{a^3}m(\text{Si}) \quad (9)$$

is the counted number of atoms  $N$  times the mean mass of the Si atoms,  $m(\text{Si})$ . Also, since silicon consists of three stable isotopes, in order to determine  $m(\text{Si})$ ,

$$m(\text{Si}) = \sum_{k=28}^{30} f(^k\text{Si})m(^k\text{Si}), \quad (10)$$

the amount-of-substance fraction,  $f(^k\text{Si})$ , of each isotope ( $^k\text{Si}$ ) has to be measured.

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<sup>1</sup>International Avogadro Coordination – IAC: Bureau International des Poids et Mesures (BIPM), Institute for Reference Material and Measurements - European Commission Joint Research Center (IRMM - Belgium), Istituto Nazionale di Ricerca Metrologica (INRIM - Italy), National Measurement Institute of Japan (NMIJ), National Measurement Laboratory (NML - Australia), National Physical Laboratory (NPL - UK), Physikalisch-Technische Bundesanstalt (PTB - Germany)

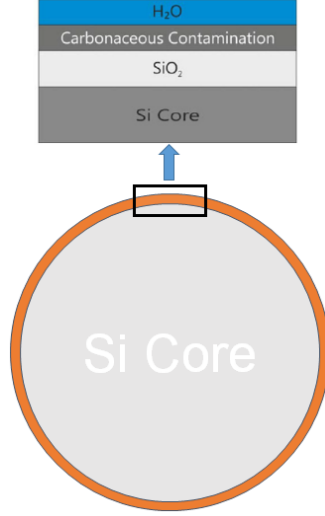


Figure 6: Model of the silicon surface.

Taking the equivalence of the ratios between atomic masses and relative atomic masses into account and considering the silicon isotopes and electron, the mass of  ${}^k\text{Si}$  is given in terms of the electron mass as

$$m({}^k\text{Si}) = \frac{A_r({}^k\text{Si})}{A_r(e)} m(e), \quad (11)$$

where the electron mass can be obtained via the measurements of the Rydberg and fine-structure constants as

$$m(e) = \frac{2hR_\infty}{c\alpha^2}. \quad (12)$$

Combining (9), (11), and (12), the mass of a silicon sphere is related to the Planck constant by

$$m_{\text{sphere}} = \frac{8V_{\text{core}}}{a^3} \frac{2hR_\infty}{c\alpha^2} \left[ \sum_{k=28}^{30} f({}^k\text{Si}) \frac{A_r({}^k\text{Si})}{A_r(e)} \right] - m_{\text{deficit}} + m_{\text{SL}} \quad (13)$$

where  $m_{\text{deficit}}$  and  $m_{\text{SL}}$  are small correction taking the influence of the point defects (i.e., impurities and self-point defects) and surface layer into account.

## 5 Conclusions

This manuscript does not want to be exhaustive, but an introduction to the challenges provided by the fascinating world of precision measurements. Many

informative articles on the kilogram redefinition are available in the literature, but I would like to point out the Metrologia papers, collected in three special issues [63–65]. They are a source of information on the efforts made to determine the Avogadro and Planck constants. The development of the new SI and, eventually, the redefinition of the International System of Units on fundamental constants of nature depended on these measurements.

Many NMIs and researchers, post-docs, and students contributed to the works here described; all were dreaming “*for all times, for all peoples.*”<sup>2</sup>

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