THE MARATHON RACE TO A NEW ATOMIC KILOGRAM

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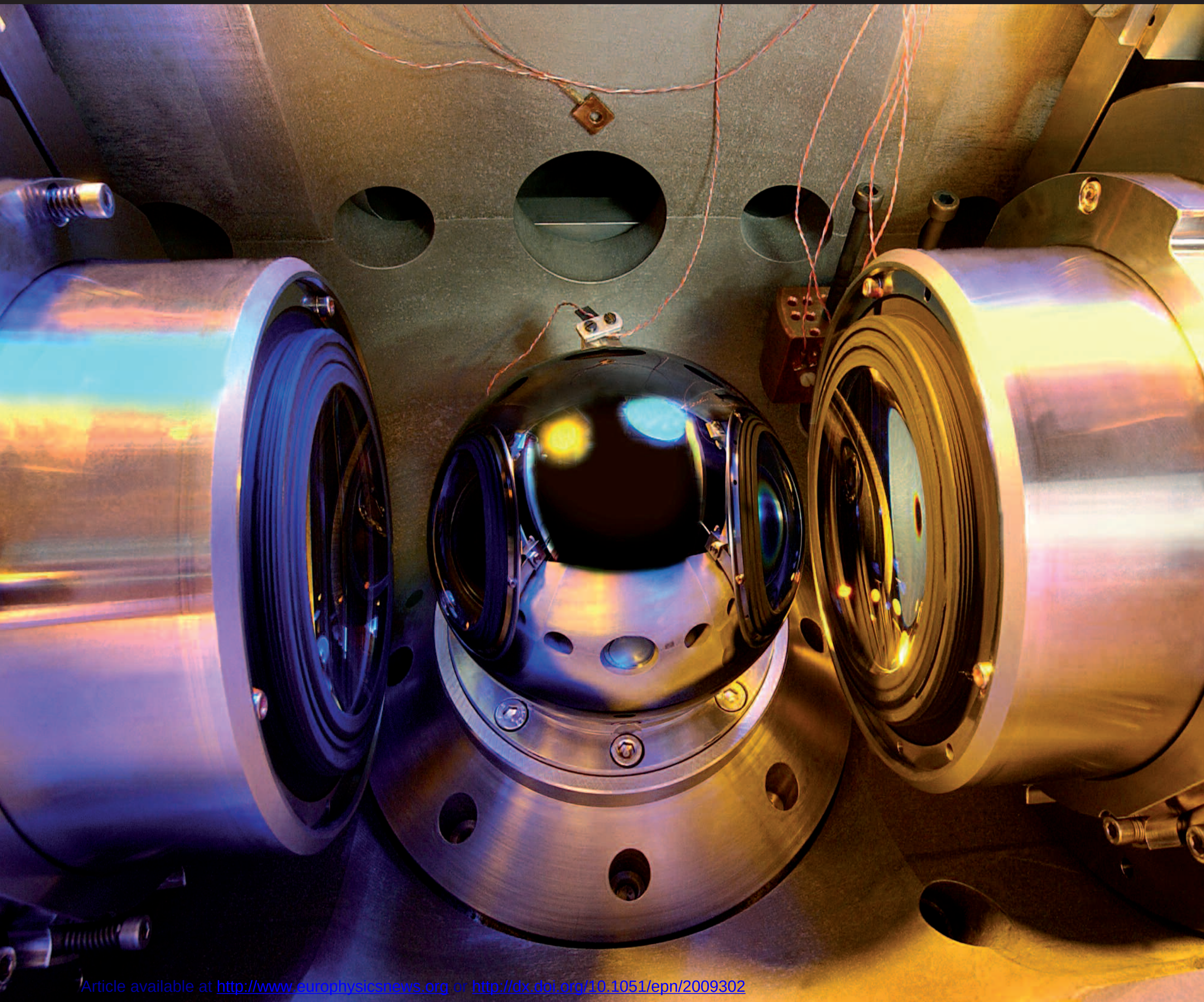
Almost all of the base units of the International System of Units (SI) have meanwhile been defined by atomic constants or fundamental constants of physics. Only the kilogram is still represented by a prototype, the prototype kilogram. For a long time now, experiments to also link the kilogram to fundamental constants have been running worldwide. Metrologists worldwide have advanced an important step with their so-called Avogadro experiment.



"The kilogram is the unit of mass; it is equal to the mass of the international prototype kilogram." This prototype, a platinum-iridium cylinder, has been stored since 1875 in a safe of the International Bureau of Weights and Measures in Paris (Figure 1). Only at this site is it possible for the national metrology institutes to assure themselves that their national mass scale is still correct – a disadvantage of this unit. A further disadvantage of this definition is the "vulnerability" of this prototype. As a result of frequent use and interaction with the environment, it changes its mass. Furthermore, it has been demonstrated in the last hundred years that the mass differences between the prototype and the various national copies have not remained constant [1].

Other base units of the SI have meanwhile been defined by atomic constants or fundamental constants of physics. Thus, the meter is linked to the second via the speed of light in vacuum. The second, in turn, is defined via the frequency of a hyperfine transition in the caesium atom.

Since about the beginning of the 1970s, test facilities have been set up in order to also link the kilogram to a fundamental constant. The objective of such an experiment for a redefinition is to attain a relative measurement uncertainty of a few times 10^{-8}. A recommendation of the CIPM, the *Comité International de Poids et Mesures*, where a redefinition of the kilogram, the ampere and the Kelvin is envisaged by 2011 in case the experimental measurements are indeed acceptable, has initiated a "race" among the national metrology institutes to tie the kilogram to a constant of nature. The general idea is to link this unit to exactly known values of the Planck constant or the Avogadro constant, which defines the number of atoms in a molar volume. The Avogadro Project, too, has its origins in the 70s, when it proved possible for German researchers to determine for the first time the lattice spacing in a silicon crystal X-ray interferometrically, without needing to know the X-ray wavelength used [2]. Thus it was possible to link the kilogram to the atomic mass unit. The Avogadro constant N_A, which specifies the number of atoms in one mole, serves as a link between a macroscopic mass and the mass of an atom:

$$N_A = M_{mol} / m_{Si} = (M_{mol} V) / (v_o m)$$

where M_{mol} is the molar mass, m_{Si} the mass of an atom, V and m are volume and mass of a silicon sphere, and v_o is the space taken up (the 'volume') of an atom.

In order to determine N_A, first V and m are determined. Then from specimens of the same crystal, the molar mass M_{mol} and the volume v_o of an atom are measured. When N_A is known, then it is easy to infer the number of atoms in a kilogram of the same material. All of the measured quantities must naturally be traced back to the existing Si base units.

Volume

The volume of the silicon sphere is determined by measuring the diameter of the sphere with a double ended Fizeau interferometer. The interferometer mainly consists of two optical surfaces in the form of spherical segments - the concave faces facing each other and so building a spherical etalon (Fig. 2). In order to fit the wave fronts of the interferometer to the curvature of the sphere, light beams from a fibre exit are first collimated by high-precision collimators and then transformed to spherical waves by means of special Fizeau objectives also containing the reference faces of the etalon. All rays of the beam therefore hit both the spherical reference plates and the surface of the sphere perpendicular to it, such that equidistant interference fringes are formed. Using a camera which can be switched to low spatial resolution (128x128 pixel) about 16000 optical phase values are measured simultaneously. For the diameter

P.23 ▶ FIG. 2: Optical characteristic of the Fizeau interferometer for determining the diameter of a sphere made of enriched silicon.

▼ FIG. 1: The prototype kilogram in Paris: the unit of mass.

measurement of the sphere two consecutive interferometric measurements are to be performed: first, the empty etalon is measured, yielding the inner diameters *D*. Then the sphere is inserted. Two new interference systems evolve now between the left arm reference face and the corresponding part of the sphere and accordingly between the right arm reference face and the opposite segment of the sphere. After measuring these distances d_{right} and d_{left} the diameter of the sphere *d* can be calculated from $d = D - d_{right} - d_{left}$. About 30 measurements at different orientations have to be performed to get a complete map of the diameters of the sphere (Fig. 3). For the best spheres they vary about 30-50 nm. The volume is then calculated from about 400 000 diameters. Measurement uncertainties of less than 1 nm for the diameter measurements could be achieved.

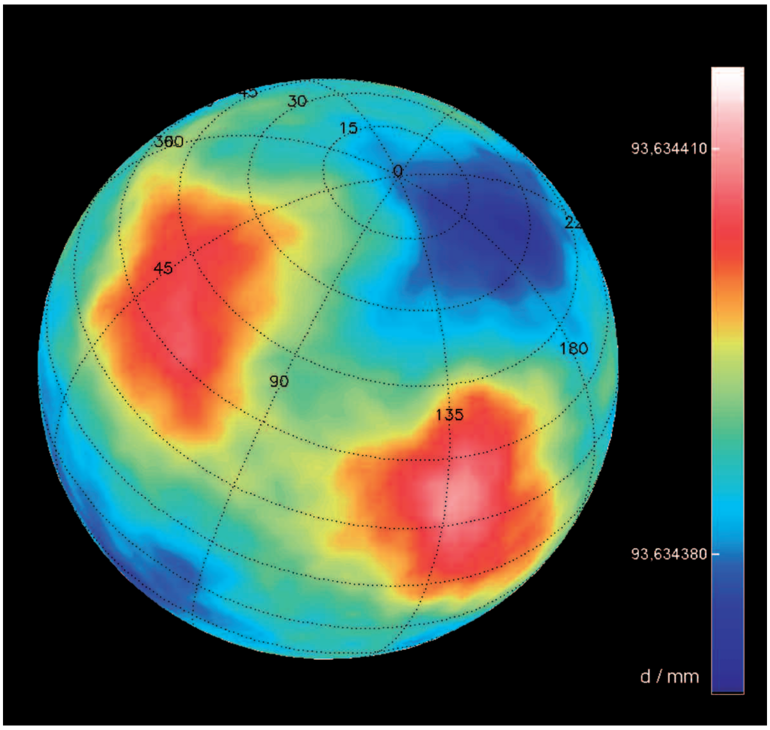
The spheres were polished in the Australian Centre for Precision Optics (ACPO). In the polishing process, the silicon sphere is overlaid with a thin oxide skin of some few nanometres, whose chemical structure and density also must be characterised. Its thickness is mapped by ellipsometry, giving a narrow net of the relative thickness. At some locations the relative values were combined with absolute layer thickness measurements by X-ray reflectometry measurements, thus providing uncertainties for the oxide layer thickness of some tenth of a nanometre.

Mass

The mass of the silicon sphere is linked to the prototype kilogram by weighing. Since the two materials have different densities, extensive buoyancy corrections are necessary. Volume and mass determinations must then still be adjusted in terms of the oxide layer.

Lattice spacing

The atomic volume is determined from the lattice plane spacing a of the crystal lattice with the aid of an X-ray scanning interferometer. The X-ray interferometer consists of three thin crystal lamellae with rows of atoms well orientated with respect to one another. The X-ray beams reaching the entrance surface of the third lamella form a periodic X-ray standing wave-pattern, reproducing the lattice period. Moving this lamella perpendicularly through the pattern results in a sinusoidal intensity modulation behind the interferometer. Measuring the travel distance using the metre as the unit of length (for instance by optical laser interferometry) and simultaneously counting the X-ray intensity maxima leads to a calibrated - mean - silicon lattice parameter a, averaged over the cross-section of the X-ray beam. Assuming the unit cell to be cubic in shape and to



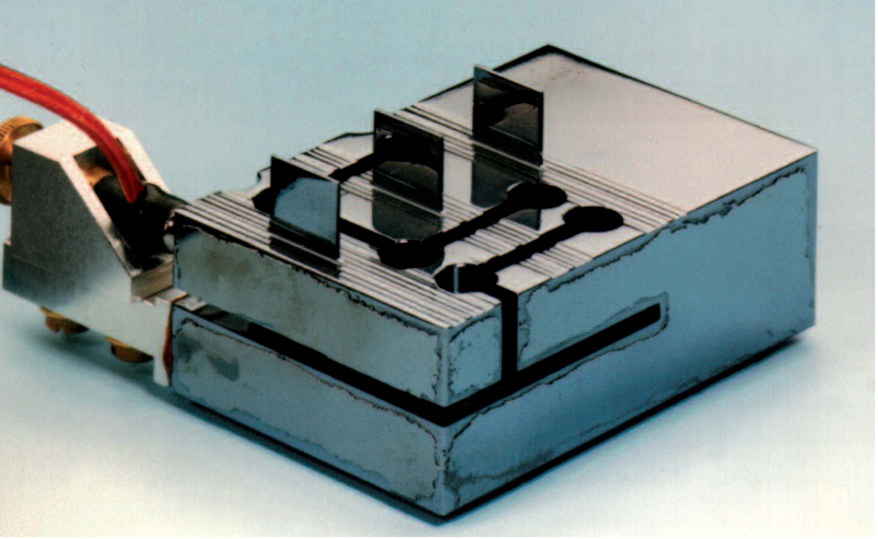
▲ **FIG. 3:** Diameter topography of a 1 kg sphere, peak to valley difference 40 nm

always contain eight atoms, the atomic volume is $v_0 = a^3/8$, with a the edge length of the cubic unit cell. Sufficient knowledge of the contaminations and of the crystal construction defects is a necessary precondition for this purpose.

The molar mass

The molar mass *M*(Si) is obtained from the measurement of the isotope abundance ratios *Ri*/28 of the three stable Si isotopes ^{28}Si, ^{29}Si, and ^{30}Si which are then combined with the molar mass values $M(^i\mathrm{Si})$, all available with an uncertainty $< 10^{-8}\cdot M(^i\mathrm{Si})$, for ^{28}Si with $< 10^{-9}$: $M(\mathrm{Si}) = \Sigma f(^i\mathrm{Si}) M(^i\mathrm{Si}) = [\Sigma M(^i\mathrm{Si})\, R_{i/28}] / \Sigma R_{i/2}$. The atomic masses of the silicon isotopes are linked to the mass of the carbon isotope ^{12}C by means of Penning traps. To calibrate the measurement of $R_{i/28}$, synthetic mixtures prepared gravimetrically from highly enriched ^{30}Si, ^{29}Si, and ^{28}Si were used. These were kept in the form of $BaSiF_6$, carrying the isotope amount ratios $n(^i\mathrm{Si})/n(^{28}\mathrm{Si})$ synthesized gravimetrically. The abundance ratio measurements are performed by means of an isotope ratio gas mass spectrometer, which compares the number of atoms of the various isotopes with each other through precise measurements of the ratios of the ion currents of the isotopes. In order to apply this method, the single crystals are converted to the gaseous compound SiF_4.

The idea is to **link the kg** to exactly **known values** of the **Planck** constant or the **Avogadro** constant



▲ **FIG. 4:** A scanning x-ray interferometer for nanometre calibration

In the case of natural silicon, however, technical limits were encountered in determining the molar mass. Therefore some years ago, the investigations of the Avogadro constant were stopped at an attained measuring uncertainty of approx. 3×10^{-7}, which was not sufficient for a redefinition of the kilogram however [3]. Unfortunately, in addition to the insufficient accuracy, also the observed difference of the measurement value of approx. 1×10^{-6} relative to other fundamental constants caused the researchers to rack their brains. Had they miscounted or are there hidden inconsistencies in the system of the physical constants?

The Avogadro Project received a new incentive with the possibility of using enriched ^{28}Si on a large scale for the production of large samples. Assessments had yielded, namely, that in this way the problems in measuring the isotopic composition, which is carried out in the European Institute for Reference Materials and Measurements (IRMM) in Geel near Brussels, were distinctly reduced. However, it was necessary to also repeat all other tests with this material.

An international Avogadro coordination IAC was agreed between national metrology institutes of Japan, Italy, Germany, Australia, Great Britain, USA, the IRMM and the BIPM, the *Bureau International des Poids et Mesures*, to bundle the existing capabilities and knowledge for the race finish. In a cooperation with research institutes in Russia in 2003, the IAC started the ambitious plan to have approx. 5 kg of highly enriched ^{28}Si (99.99%) produced as single crystal and to attain a measuring uncertainty until 2010 of approx. $2 \cdot 10^{-8}$. Meanwhile, the production of the material was successfully concluded with the growing of a perfect dislocation-free single crystal at the *Helmholtz Institute of Crystal Growth* in Berlin, see Figure 3. Two ^{28}Si spheres, each weighing one kilogram, were already polished in the Australian ACPO and arrived at the metrology laboratories of Germany and Japan in April last year. Here they will be measured in the hope of attaining the objective of a more accurate determination of the Avogadro constant within the prescribed time span. In the process, it should also be possible to uncover the cause of the observed difference with other fundamental constants.

A first inspection of the material has been started and the following characteristics of the material have been checked: impurity, vacancy and void contents, isotope enrichment, lattice parameter and density differences, presently compared with silicon of natural composition. All data measured so far meet the expectations of the scientists. For example, the carbon and oxygen content in the Avogadro crystal – usually the dominant impurities in commercially available FZ crystals – is astonishingly small.

The Avogadro constant is not the only candidate in the running for a redefinition of the unit of mass. The so-called watt balance experiments, which are being set up at several metrology institutes, *e.g.* in the USA, England and Switzerland, are determining the Planck constant and are given better odds in expert circles of winning the competition. But matters have not progressed so far yet. ■

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▲ **FIG. 5:** The highly enriched Avogadro ^{28}Si single crystal