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LETTER TO THE EDITOR

The silicon route to a primary realization of the new kilogram

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Abstract
The General Conference on Weights and Measures (CGPM), at its 24th meeting, took note of the intention of the CIPM to revise the SI such that the new definition of the kilogram will be based on a fixed value of the Planck constant. Thus, any experiment providing a direct link between the Planck constant and a macroscopic mass, and not including any traceability to another primary kilogram standard, can be considered as a primary method for the realization of the new kilogram. In this letter we show that a silicon single crystal, as used in the Si-Avogadro experiment, is suitable as a primary kilogram standard according to the new proposed definition. We further discuss its potential with regard to the achievable relative uncertainty, which has to be 2×10^{-8} or better to serve all dissemination needs for the unit kilogram. This silicon route to the primary realization of the new kilogram represents a method which is completely independent of the alternative watt balance approach.

1. Introduction

The General Conference on Weights and Measures (CGPM), at its 24th meeting, took note of the intention of the CIPM to revise the SI such that the new definition of the kilogram will be based on a fixed value of the Planck constant, h [1]. In an analogue manner, the new definition of the mole, kelvin and ampere shall be based on fixed values of the Avogadro constant N_A, the Boltzmann constant k_B and the electron charge e, respectively.

Thus, the direct relation of a unit to one fixed constant is systematically implied. However, all base units with the exception of second and mole, and most of the derived units take reference to more than one constant of nature, e.g. the ampere needs the definition of the elementary charge and the second. The Planck constant quantifies the quantum of action and has the unit $\mathrm{J\,s} = \mathrm{kg\,m^2\,s^{-1}}$, containing the unit kg among other base units. Consequently, the draft chapter 2 of the new SI brochure includes the description of the more general, fundamental concept behind the redefinition: the whole system of units is scaled such that a set of constants of nature is fixed to specific values, irrespective of base or derived units, and without specified relations between units and specific constants.

In this letter we show that a macroscopic single-crystal sphere of highly enriched ^{28}Si is suitable as a primary realization of the unit kilogram according to the envisaged new definition. From a point of view of principle we show that the determination of the mass of the silicon sphere does not include any traceability to another primary kilogram standard but only takes reference to exact constants of nature—among them the Planck constant—and includes relative measurements.

2. Relation between the watt balance and the Avogadro experiment

The so-called watt balance experiment [2][1] is an approach which provides a direct link between the Planck constant and a macroscopic mass via virtual comparison of electrical and mechanical power induced by a test mass. The electrical quantities are measured in terms of the Josephson and von Klitzing constants, K_J and R_K, respectively. The determination of h from the measured product of K_J^2 and R_K is based on the assumption that the values of the Josephson and von Klitzing constants are exactly $2e/h$ and h/e^2, respectively.

The Avogadro experiment [3, 4] (sometimes also referred to as XRCD, 'x-ray crystal density' experiment) is aimed at

[1] For a summary of recent watt balance experiments see [7].

the determination of the Avogadro constant N_A using a highly isotopic-enriched single crystal of silicon. It was originally set up in order to realize an alternative way of redefining the kilogram on the base of an exactly defined atomic mass such as that of the ^{12}C atom, the ^{28}Si atom or the mass of an elementary particle.

As pointed out already by Mills *et al* [5] the Avogadro constant and the Planck constant are related via

$$N_A = \frac{\alpha^2 c}{2R_\infty h} A_r^e M_u, \tag{1}$$

with the fine structure constant $\alpha = e^2/2c\varepsilon_0 h$, the speed of light in vacuum c, the Rydberg constant $R_\infty = m_e e^4/8c\varepsilon_0^2 h^3$, the relative atomic mass of the electron A_r^e, and the molar mass unit M_u, respectively.

The watt balance and the Avogadro experiments are completely independent approaches tied via equation (1), and provide a fundamental consistency check for the measurement of the Planck constant or the Avogadro constant, respectively. Furthermore, the molar Planck constant hN_A can be independently measured by γ-spectrometry [6] and is known with a relative uncertainty of 7×10^{-10} [7].

3. The silicon route for a primary kilogram standard

The Avogadro experiment relates the mass m of a macroscopic silicon single crystal, to its volume V, molar mass M_{Si}, lattice parameter a_0 and the Avogadro constant according to

$$m = \frac{8V}{a_0^3} \frac{M_{Si}}{N_A}. \tag{2}$$

The molar mass M_{Si} has to account for all three isotopes of silicon: $M_{Si} = \sum_i f_i M_{Si}^i$, where i stands for three isotopes ^{28}Si, ^{29}Si and ^{30}Si, and f_i are the respective mole fractions. While the original Avogadro experiment used natural silicon with abundances of about $f_{28} = 0.922$, $f_{29} = 0.047$ and $f_{30} = 0.031$, the latest version [3, 4] used highly enriched ^{28}Si ($f_{28Si} \approx 0.999\,95$). Combining equations (1) and (2) and using $\sum_i f_i M_{Si}^i = \sum_i f_i A_r^i M_u$, with A_r^i being the relative atomic mass of isotope i, yields

$$m = \frac{8V}{a_0^3} \frac{2R_\infty h}{c\alpha^2} \sum_i f_i A_r^i A_r^e. \tag{3}$$

This is the basic relation for the silicon route to realize the new kilogram.

Equation (3) links the macroscopic mass m to the Planck constant h according to the definition of the new kilogram. None of the further input quantities requires a traceability to another primary kilogram standard, neither directly nor indirectly. Thus, such a configuration itself can be considered a primary kilogram standard. In fact, despite the name 'Avogadro experiment', the Avogadro constant does not enter explicitly.

The fraction $8V/a_0^3$, which is equal to the number of atoms in the macroscopic body, includes the measurement of the volume and the silicon lattice parameter. The factor 8 accounts for the number of atoms in the specific unit cell of silicon. The chosen geometry of a macroscopic silicon body is the sphere, which only requires a set of diameter measurements scanning the whole surface. The uncertainty of the volume determination is limited by deviations from the ideal sphere, which leads to wavefront deformations and imperfect overlap of the light beams forming the interferograms. With shape deviations of the order of 100 nm, relative uncertainties of 2.3×10^{-8} for the volume have been achieved [4]. With improvements to the polishing procedure, shape deviations of the order of 20 nm and uncertainties for the volume of parts in 10^{-9} seem feasible. Other, less symmetric geometries, such as a cube, will be inferior due to extra sources of error and uncertainty mostly at the edges and corners. The lattice parameter a_0 of the enriched ^{28}Si crystal is known with a relative uncertainty of 3.5×10^{-9} [8].

The factor $2hR_\infty/c\alpha^2 = m_e$ of equation (3) has the unit kilogram; it contains the speed of light c and the Planck constant, which will both be exact after the redefinition of the base units. The fine structure constant α is known with a relative uncertainty better than 10^{-9}. After redefinition, e and h will be exact, but the value of the dielectric constant will have an uncertainty, leaving the uncertainty of α unchanged, however. The best experimental values for α are derived from the measurements of the g-factor of the electron in Penning traps, which are not affected by the redefinition. The Rydberg constant corresponds to a frequency measurement in the hydrogen atom and will also not be affected by the redefinition. It is one of the best-known measured constants of nature with a relative uncertainty of 6.6×10^{-12} [7].

The factor $\sum_i f_i A_r^i / A_r^e$ of equation (3) is the ratio of the atomic mass of silicon with its specific isotope composition and the electron. The relative atomic masses are measured by a comparison of cyclotron frequencies in Penning traps. They do not require traceability to another primary kilogram standard. Even if the relative atomic mass of ^{28}Si [9] and the electron are measured versus ^{12}C [10], the exact definition of the atomic mass of ^{12}C is not needed and its uncertainty after redefinition will cancel out.

The mole fractions f_i are determined using mass spectroscopy. In order to reach the desired uncertainties for the highly enriched samples with large differences in the mole fractions ($f_{28Si} \approx 0.999\,95$), a method based on isotope dilution mass spectrometry has been developed [11]. Combined with multicollector detection in an inductively coupled plasma mass spectrometer, a relative uncertainty of molar mass of 8×10^{-9} has been achieved [3].

4. Further constraints

The relative uncertainty of a primary kilogram standard must be better than 2×10^{-8} in order to serve all dissemination needs of the unit. At this level of uncertainty the surface layer, the impurities and voids in the single crystal have to be analysed carefully in addition to the determination of the quantities entering explicitly in equation (3).

A silicon single crystal exposed to air will be covered by an oxide layer and possibly contaminants such as carbon

and water. Their contributions to the total mass must be determined. The sphere is either thermally oxidized after cleaning the surface by chemical etching or is oxidized under normal conditions. The thickness of the oxide layer and its composition are determined by x-ray reflectometry and optical spectral ellipsometry and with the help of various x-ray analytical tools such as XRF (x-ray fluorescence), NEXAFS (near edge x-ray absorption fine structure) and XPS (x-ray photoelectron spectroscopy). The overall contribution of the silicon surface coverage to the uncertainty currently amounts to 1.5×10^{-8}, relatively, but can be further decreased with a cleaner polishing procedure [3]. For a primary standard this contribution must use the atom densities of the contaminants and their atomic masses relative to that of silicon, but not mass densities in order to avoid a traceability to another primary kilogram standard.

Even though silicon single crystals can be grown with very high purity and crystal perfection, the concentration of residual impurities and defects has to be determined quantitatively. The major impurities to be considered are carbon, oxygen and boron. Their concentrations have been determined by infrared spectroscopy [12]. The vacancy density was measured by positron lifetime spectroscopy [13]. The overall contribution of point defects to the total uncertainty amounts to 3×10^{-9}, relatively [3].

5. Conclusions

We have shown that a sphere of pure silicon is suitable to be used as a primary kilogram standard according to the envisaged new definition. It links a macroscopic mass m to the Planck constant h according to the definition of the new kilogram, does not involve traceability to another primary kilogram standard to determine the total mass, and reaches uncertainties which are sufficient to provide traceability at an uncertainty level requested by the CGPM.

Currently, the major contributions to the published uncertainty of the determination of the Avogadro constant arise from the determination of the volume (66%), surface properties (18%) and lattice parameter (9%) [3]. These measurements and their uncertainties will contribute similarly when the silicon sphere will be used as the primary kilogram standard. However, as pointed out briefly in this letter, considerable improvement seems possible and thus a primary kilogram standard using the silicon sphere with a relative uncertainty below 10^{-8} seems feasible.

Redundancy of the silicon route as the primary kilogram standard independent of other routes such as the watt balance can be achieved by producing several spheres, at best from different bulk materials, and determining their masses according to equation (3) with independent instrumentation. The weighing of these masses must then be consistent when compared using high-precision mass comparators. Such an initiative is currently under way. It will provide a significant contribution to resolving the still existing discrepancies between the various experiments to determine the Planck constant before redefinition. After redefinition, it can form the backbone of the internationally distributed system of primary kilogram standards.

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