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DISSEMINATION OF THE « PLANCK-KILOGRAM »

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ABSTRACT

Metrologists all over the world are looking at the activities of the Metre Convention concerning the New International System of Units on the basis of fundamental constants. Especially the redefinition of the kilogram will have consequences in nearly all fields of physics, engineering and trade. A new definition of a base unit imperatively implies thoughts about the future dissemination of the unit. After a short glance at the potential of the Kibble(Watt)-Balance the presentation will describe the strategy of the Physikalisch-Technische Bundesanstalt to use silicon spheres of different qualities for the dissemination of the quantum based kilogram to the macroscopic world. Beside metrological aims the availability of realisations is of crucial importance within a future dissemination chain. Aspects like the connection to the established system as an important aspect for acceptance, the applicability of the developed tools and procedures for using such spheres as mass standards or the current state of activities to proof the expected excellent long term characteristics of silicon spheres in use will be presented.

Index Terms Redefinition of the Kilogram, Dissemination, Silicon Sphere, Avogadro-Project

1. INTRODUCTION

Since 1889 an artefact defines the unit Kilogram in the International System of Units. In 2018 metrologists worldwide are awaiting the decision about a change to a new system of international units defined via fundamental constants. Even the kilogram will be defined by a natural quantum based constant, the Planck constant. The new kilogram will overcome the gap to the electrical units measured based on quantum effects like the Josephson effect and the quantum Hall effect. But for daily life macroscopic realisations of this quantum based unit will be needed. Up to now only the experiments providing data for the redefinition of the kilogram – the Kibble-Balance, the Avogadro-Sphere (as result of the X-ray crystal density (XRCD) experiment) and the Joule-Balance – are expected to allow macroscopic realisations with the necessary small uncertainties. But, can these tremendously expensive and highly sophisticated experiments be used in the sense of disseminating the kilogram?
At the IMEKO World Congress in September 2015 PTB presented for the first time the ideas of setting up a dissemination system for the “Planck-Kilogram” based on silicon spheres [1]. Most of the aspects presented there are still valid. The current paper will provide an overview of the status quo of these activities within the framework of the current mass metrological developments.

2. CURRENT STATUS

In 1999 the General Conference on Weights and Measure (CGPM) recommended that the national metrology institutes shall continue their experiments to “link the unit of mass to fundamental or atomic constants” [2]. Since then a lot of effort was spent to improve the existing experiments and to develop new methods with the aim to redefine the kilogram and other base units in a universal manner, i.e. independent of artifacts.
The Consultative Committee for Mass and Related Quantities (CCM) of the International Committee of Weights and Measures (CIPM) summarized the requirements necessary for a redefinition of the base unit kilogram at its 2005 meeting for the first time. These requirements were discussed and refined several times. In 2013 the following final version of the requirements was recommended by the CCM [3]:

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,
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

Following the “Joint CCM and CCU roadmap for the new SI” [4] the CGPM in 2018 may decide about the future of the SI-units. If the listed requirement will be fulfilled also the kilogram may be redefined.

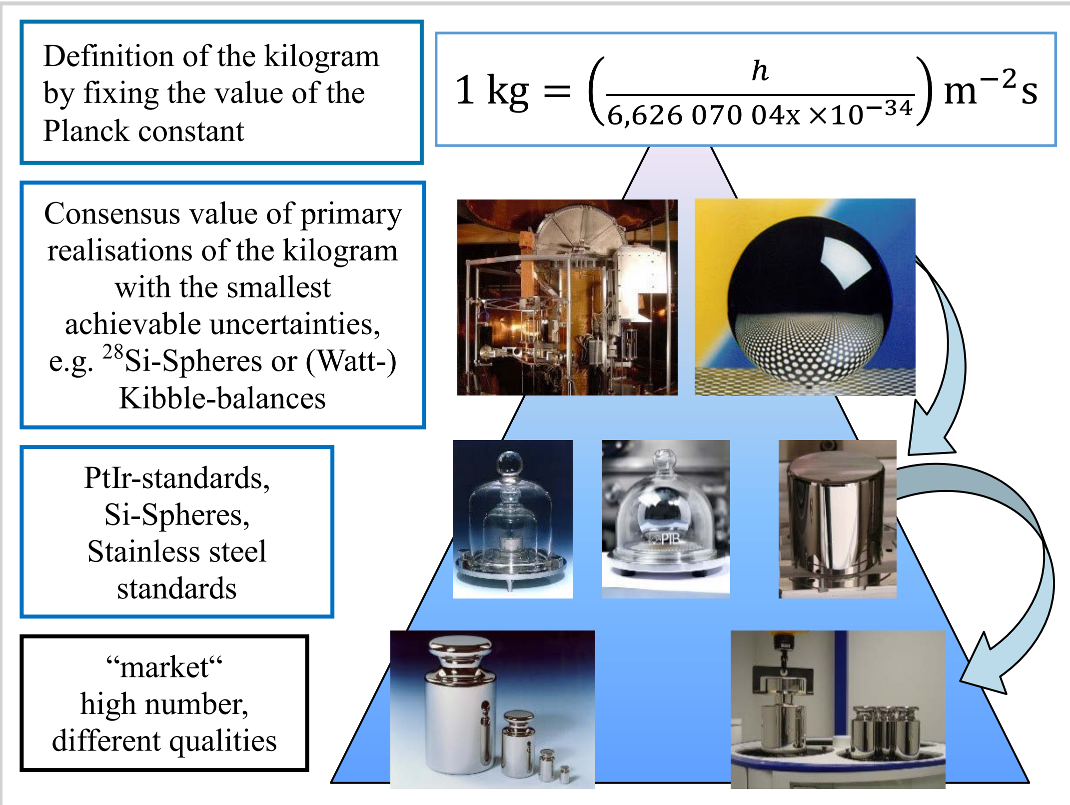


Figure 1: Change of dissemination after redefinition of the kilogram

In the current version of the drafted *Mise en Pratique* of the definition of the kilogram only two methods are described to be capable to realize the kilogram definition with acceptable relative uncertainties – Kibble(Watt)-balances and ^{28}Si-spheres [5]. Of course, other primary realizations may reach the same magnitude of uncertainties in future.
In CCM recommendation G 1 of 2017 [6] the need of “an on-going key comparison of primary realizations of the kilogram” at the level of national metrology institutes as a possible “procedure for applying corrections relative to the consensus value” was mentioned. Thus, the future dissemination structure for the kilogram may finally change as presented in figure 1.

3. KIBBLE/WATT-BALANCE FOR DISSEMINATION

At the moment, many institutes worldwide are developing their own systems balancing mechanical with electrical power or energy. The resulting systems are so called Kibble(Watt)-balances or Joule-balances. They were constructed with the aim to determine the Planck constant as good as possible. Only a few of them – the systems of NIST, NRC, NIM and LNE – provided measurement results for recognition within the CODATA value of the Planck constant. The different systems work at different target values, but, most of the balances work at a mass value of 1 kg. The existing systems are mainly of large geometry and expensive in use because of their construction – e.g. used materials, large coils, temperatures and pressures. The development of table top systems using a defined value of the Planck constant to realize mass values will offer alternative ways to use this method in future (see e.g. [7]). It is the aim to develop small, easy to use and accurate systems that can measure not only at a single mass value but continuously in a mass range. The so-called “Planck balance” – a cooperation between TU Ilmenau and PTB – as an example of such a system is far beyond theoretical considerations and will be described by the project colleagues elsewhere.
Alternatively, to standard measuring systems like Kibble-balances well characterized silicon spheres could be used for the realization of the unit kilogram. Possible qualities of such mass standards and the respective framework will be described in the following chapters.

4. THE «SILICON WAY» OF KILOGRAM DISSEMINATION

4.1 Motivation

During the process of establishing silicon spheres for the determination of the Avogadro constant and the Planck constant using the so-called X-Ray Crystal Density (XRCD) method a lot of experience arose in how to manufacture, characterize and handle silicon spheres of superior quality. During the years of activities of the International Avogadro Coordination (IAC) the Si-spheres were not only measurement objects but as well transfer standards showing a very good stability during transport and over time.
In order to determine the Planck Constant with as small measurement uncertainty as possible it was necessary to use highly isotopic enriched monocrystalline silicon (^{28}Si) to minimize influences due to the material, e.g. molar mass determination or point effects. However, such spheres are very expensive because of the rare material and time consuming manufacturing. Otherwise, they can be used as primary kg realizations with very low measurement uncertainties. During discussions on how to disseminate the kilogram via silicon spheres it became clear, that within the traceability chain different quality levels of spheres may be reasonable. As an outcome of this discussion three categories of 1 kg-Si-spheres were defined (see table 1):

- “**^{28}Si**” – “primary” Si-spheres, made of ^{28}Si with surface and roundness of highest quality, well characterized and ready to use as a primary mass standard with relative measurement uncertainties in the range of $1{,}2 \cdot 10^{-8}$.

- "$^{nat}Si_{qp}$" – "quasi-primary" Si-spheres, made of natural monocrystalline silicon with surface and roundness of highest quality, well characterized and ready to use as a mass standard. Even the "quasi-primary" spheres could act as primary mass standards depending on the methods used for their characterization but with a slightly higher measurement uncertainty than the spheres made of ^{28}Si. The expected relative measurement uncertainty of the mass is in the range of $3 \cdot 10^{-8}$ when calibrated by magnetic flotation method.
- "$^{nat}Si_{sc}$" – "secondary" Si-spheres, made of natural almost monocrystalline silicon with surface and roundness of high quality, manufactured by industry, calibrated and ready to use as secondary mass standards in calibration labs. The relative measurement uncertainties will be in the magnitude of $5 \cdot 10^{-8}$.

Table 1: Categories of Si-spheres proposed as standards for dissemination

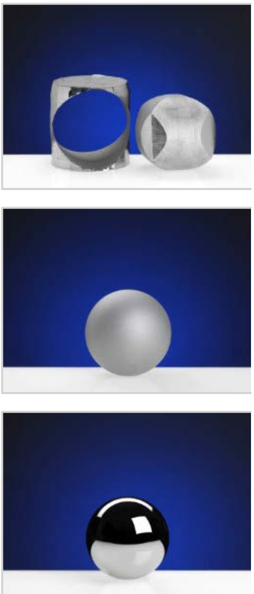
	"^{28}Si"	**"$^{nat}Si_{qp}$"**	**"$^{nat}Si_{sc}$"**
category	primary	"quasi primary"	secondary
u_{rel} (k=1) of mass	$2 \cdot 10^{-8}$	$3 \cdot 10^{-8}$	$3 \cdot 10^{-8}$
form error RONt	< 30 nm	< 20 nm	< 80 nm
average roughness Ra	< 0,3 nm	< 0,3 nm	< 1 nm
expected price	> 1 Mio. €	> 100 k€	> 10 k€
Availability	limited, prepared by PTB	prepared by PTB on inquiry	industrial product
PTB recommended transport packaging	transport container in an aluminum case covered with bespoke polystyrene material	transport container in an aluminum case covered with bespoke polystyrene material	sphere protected by a microfiber tissue in an aluminum case covered with bespoke polystyrene material
Delivered data (see 4.3)	mass, volume, density, molar mass, crystal quality, oxide layer	mass, volume, density, crystal quality, oxide layer	volume/density, mass

4.2 Manufacturing of the Si-spheres

First step in the manufacturing of a Si-sphere that can possibly be used as a mass standard is the preparation of a mono-crystal of natural or highly isotopic enriched monocrystalline silicon. The enrichment of ^{28}Si is a scientific and technological challenge now solved by Russian colleagues of the Electrochemical Plant in Zelenogorsk and the Russian Academy of Science in Nishniy Novgorod. From the poly-crystalline material (natural silicon or isotopic enriched material) the Institute for Crystal Growth in Berlin prepares mono-crystals of a diameter of about 100 mm [8, 9].

A mono-crystal made of natural silicon or ^{28}Si of about 6 kg is departed into blanks for two spheres and test material after the part containing crystal misalignments was cut off. From a coarse cut blank, the first rough ingot is drilled out. After several scientific investigations and experiments it came out, that not edging but classical turning, lapping and polishing procedures allow the best and even manageable results in form and surface preparation as

well as for the sensitive process of silicon oxide formation. Finally, the developed procedures allow the manufacturing of a sphere with a form error RONt less than 30 nm and an average roughness value Ra of less than 0,3 nm [10, 11] (see table 1).
Not only the visible surface quality is determining the quality of the sphere. It is important that the boundary layer resulting of processing the mono-crystalline material is rather uniform and small because here the silicon oxide forms. For mass metrology, finally the maximum deviation of the target mass value is important as well. The maximum admissible deviation of the mass is ± 10 mg. This quality is only achievable by in-process measurements of geometry and mass (see figure 2).
PTBs manufacturing expertise and the developed processing machines allow a stable production of Si-spheres of highest quality within 3 month. At the moment, this technology is transferred to industrial partners to ensure sphere production independently of PTB in industrial scale number. In future, the manufacturing of "secondary" spheres should be handled by industry only.



1. *Cutting processes:*
   in-process inspections;
   - caliper gauge
   - balance
2. *Lapping processes:*
   in-process inspections;
   - balance
   - form measurements
3. *Polishing processes:*
   in-process inspections;
   - form measurements
   - mass determination
   - roughness measurements

Figure 2: In-process measurements during Si-sphere development at PTB [1]

4.3 Characterization of Si-spheres

For many years, the colleagues of the International Avogadro Coordination (IAC) investigated what the determining parameters of a Si-sphere are and how these parameters can be obtained. For the determination of the Planck-Constant the following aspects were identified as the most important ones - volume, molar mass, lattice parameter, point defects, surface (here especially the silicon oxide layer, contaminations (carboneous, metallic, others), and scratches or other marks), and last but not least the mass [12, 13].
To determine the mass value of a ^{28}Si sphere or a $^{nat}Si_{qp}$ sphere the same quantities as listed above should be determined. Additionally, at PTB the density of the sphere will be determined by the so-called magnetic flotation method [18] with respect to well characterized silicon samples. The method is still under development and will probably be available in 2019. The values for mass and density of the sphere have more or less the function of validation data – their relation to the volume should stay stable for each sphere. All measurements, except mass and density are independent to any mass referred quantity. Thus, these results allow a primary realization of the mass unit.

When using silicon spheres as standards for disseminating the kilogram (^{28}Si or $^{nat}Si_{qp}$) it will not be necessary and even not be possible to determine all parameters day by day. Taking into mind that the composition and structure of the material will not change during use of the spheres, lattice parameter, point defects and molar mass will not change. As long as the spheres are not mechanically damaged (e.g. scratches), re-polished or re-lapped the determined volume will be stable over a long time too.
Thickness of the oxide layer, surface contaminations and the surface quality will change over the time. These parameters may have influence on the mass value of the sphere and thus, must be checked regularly. Different methods for characterization may be used for different levels of quality (see figure 3). Every sphere delivered by PTB has a marking on its surface that allows a repeated positioning of the spheres in different instruments with respect to the orientation of the spheres crystal. This enables a topographic mapping of a sphere and thus, the determination of changes at the spheres surface. Scratches could be identified visually by using a strong torch (see methods referred to in chapter 5). For regular check of the oxide layer and surface contaminations the spectral ellipsometry is recommended because it allows a fast monitoring of the spheres surface with acceptable effort. The necessary calibration for the use of this method may be externally (proposed for $^{nat}Si_{sc}$) or internally as calibration points at the surface of the sphere (recommended for ^{28}Si and $^{nat}Si_{qp}$). The “internal calibration” is done with an XPS/XRF-measuring system developed at PTB [14]. Of course, other measuring systems realizing these methods could be used as well.
Based on today’s experience PTB proposes to validate the surface layer stability every two years and the volume every 10 years for ^{28}Si and $^{nat}Si_{qp}$ spheres.

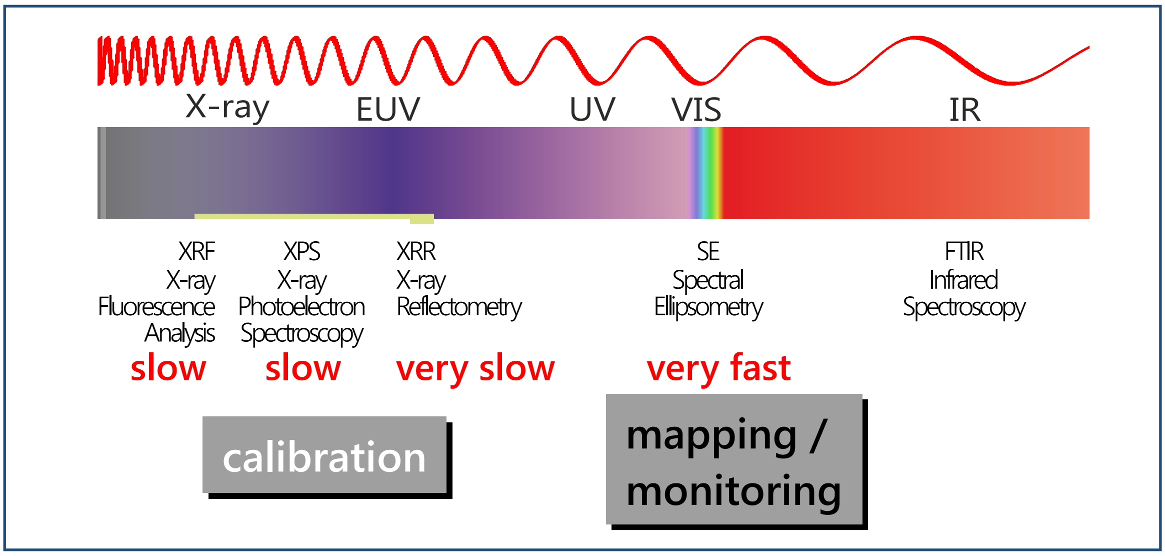


Figure 3: Methods of surface analysis used for characterizing Si-spheres [15]

Thus, for using silicon spheres as mass standards in the dissemination system it will be needed to obtain regularly possible changes at the surface of the respective sphere (e.g. scratches) and to determine changes of the oxide layer. The determination of the other parameters (e.g. volume, molar mass) is only needed at the initial characterization of the sphere or after a re-manufacturing. All spheres may be delivered with material pieces not used for sphere manufacturing. It allows a later independent determination of all material measures needed for a determination of a primary mass value based on Planck’s constant. Thus, all relevant parameter may be determined independently of PTB measures.
The “secondary” silicon spheres ($^{nat}Si_{sc}$) may be characterized in the same way. But, because of the targeted use and the slightly worse manufacturing quality, spheres of this category will

be characterized like "conventional" mass standards by mass, volume (via density) and the visible surface quality only.

5. SILICON SPHERES AS MASS STANDARDS

In principle, silicon spheres can be used in the same way as other mass standards at the highest level (E1 according to OIML R111 [16] or better). Because of the sphere's geometry, it is necessary to apply specific measures. Manufacturers of mass comparators considered this aspect still when constructing modern instruments. Silicon spheres are not sensitive against magnetic or electromagnetic fields what make them probably attractive as mass standards for specific measurement conditions. Because of the smaller density, the effect of air buoyancy will be larger than for stainless steel standards. Thus, it is recommended to use buoyancy artifacts to obtain changes of air buoyancy experimentally during the measurement [17].
The biggest advantage of silicon spheres is their stability. If they are handled with care they show a stable behavior even after several vacuum-air-changes. If there are some contaminations due to normal use the mass value of the spheres could be "reset" within a short time (24 h stabilization time seems to be enough) by relatively simple cleaning. At the "Round and Ready" Workshop at PTB in 2016 respective tools and methods for handling and use of silicon spheres were presented. The posters and videos are available via Internet at www.ptb.de/si-kg-2016.
At the moment 23 volunteering institutes from all over the world are testing the practicability of the "toolbox" developed at PTB. PTB is lending silicon spheres ($^{nat}Si_{qp}$ or $^{nat}Si_{sc}$) to the members of the so-called "Si-Trust" up to two years. These institutes will execute some tests discussed beforehand, in any case mass determinations, and get the possibility to do additionally some experiments of their own. Just now there are already 3 laboratories having spheres for tests and probably 11 others will get spheres within 2017. Nine other laboratories are planned to follow within 2018. As results of this measurement campaign not only conclusions concerning the stability of the spheres and the usability of the handling procedures are expected but as well comments, questions and proposals for future developments in this field. Additional partners to join Si-Trust are welcome (contact: si-trust@ptb.de).
One of the points coming up regularly is the manufacturing and characterization of smaller silicon spheres as mass standards. In density metrology, the use of smaller silicon spheres is still common. Currently, these spheres are not characterized at a level to become primary realizations of mass units. But, because the uncertainty of density comparisons decrease importantly using magnetic flotation [18] the uncertainty of the mass value of such spheres could be smaller in future than the uncertainties of (other) primary realizations. Just now, the manufacturing of such spheres is not intended at PTB.

6. SUMMARY

When the SI unit kilogram will be redefined based on a fundamental constant there will be not only one primary unit realization with zero uncertainty but several primary realizations with uncertainties determined following the rules of the Guide to the Expression of Uncertainty in Measurement [19]. The methods being capable to reach relative uncertainties smaller than $5 \cdot 10^{-8}$ are, just now, only the two methods used for the determination of the Planck constant. They are expensive and rare. Beneath the highest level in principle nothing needs to be changed. But, the existing experiences with manufacturing and use of silicon spheres open the possibility to use silicon spheres as stable mass standards in the laboratories too. Finally, the spheres are surprisingly simple to handle, robust and stable. Industrial manufactured spheres of natural silicon will be available in bigger numbers in near future. Finally, because of the expected excellent stability in combination with the achievable high accuracy silicon spheres will play a major role in future dissemination structures.

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