# Volume Measurement of a $^{28}$Si-Enriched Sphere for a Determination of the Avogadro Constant at NMIJ

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*Abstract*—**For a determination of the Avogadro constant by the X-ray crystal density method, the volume of a Si sphere manufactured from a Si crystal isotopically enriched in $^{28}$Si, named Si28-23Pr11, was measured at the National Metrology Institute of Japan (NMIJ). The volume was determined on the basis of diameter measurement in 2175 directions by an optical interferometer. The surface layers on the sphere were characterized by X-ray photoelectron spectroscopy and spectroscopic ellipsometry. Taking into account the effect of the surface layers on the diameter measurement, the volume of the Si core excluding the surface layers was determined with a relative uncertainty of $2.0 \times 10^{-8}$. The results were used to determine the Avogadro constant in 2017 by the International Avogadro Coordination project, which was used as an input datum to determine the CODATA 2017 adjusted value of the Planck constant $h$. This $h$ value will be used in the new definition of the kilogram. This paper provides details of the measurement apparatus and the uncertainty analysis of the Si core volume measurement. In addition, an approach to reduce the uncertainty in the Si core volume determination is proposed for the accurate realization of the kilogram based on the new definition at NMIJ. Details of the approach are also provided.**

*Index Terms*—**Avogadro constant, diameter measurement, optical interferometer, silicon crystal, volume measurement.**

## I. INTRODUCTION

**T**HE International Committee for Weights and Measures (CIPM) has proposed a change in the definition of the unit of mass, the kilogram [1]. The new definition will be based on a fixed value of the Planck constant $h$ and is anticipated to be accepted in 2018 [2]. Toward the change in the definition, national metrology institutes have been requested to continue attempts to link the kilogram to $h$ with a relative uncertainty of a few in $10^{-8}$. The Avogadro constant $N_A$ is an important input datum in changing the definition of the kilogram because $h$ can be calculated from $N_A$ via the molar Planck constant $N_A h$. Since the relative standard uncertainty of $N_A h$ has been estimated to be as small

as $4.5 \times 10^{-10}$ by the CODATA Task Group on Fundamental Constants [3], the accurate and independent measurement of $N_A$ played a crucial role in the determination of $h$ in the new definition.

The X-ray crystal density (XRCD) method was an approach to determine $N_A$ value with a relative uncertainty of $10^{-8}$. In this method, absolute measurements of the lattice constant, density, and molar mass of a silicon crystal are performed. In 2011, 2015, and 2017, the International Avogadro Coordination (IAC) project succeeded in determining $N_A$ with relative standard uncertainties of $3.0 \times 10^{-8}$, $2.0 \times 10^{-8}$, and $2.4 \times 10^{-8}$, respectively, using a Si crystal isotopically enriched in $^{28}$Si, named AVO28 [4]–[6]. To determine $N_A$ using different $^{28}$Si-enriched crystals, a new crystal, named Si28-23Pr11, was prepared by the Physikalisch-Technische Bundesanstalt (PTB, Germany) [7]. In 2017, the IAC project determined $N_A$ using this new crystal [8]. The relative uncertainty of the $N_A$ determination was estimated to be $1.2 \times 10^{-8}$, which is the smallest uncertainty achieved by the XRCD method to date. The four $N_A$ values determined by the XRCD method were used as input data for the CODATA 2017 special adjustment to provide the $h$ value in the new definition of the kilogram [9].

For the new $N_A$ determination in 2017, the density was determined from the volume and mass of two $^{28}$Si-enriched spheres, Si28kg01a and Si28kg01b, manufactured from Si28-23Pr11 [8]. The spheres were covered with surface layers having silicon dioxide as their main constituent, as shown in Fig. 1, and the volume of the Si core excluding the surface layers should be determined for accurate $N_A$ determination. To determine the sphere volume, optical interferometers of PTB and the National Metrology Institute of Japan (NMIJ) were used. Since the surface layers cause a phase shift upon the reflection of the light beam, the volume measured by the optical interferometers was, therefore, the "apparent volume," which was different from the core volume. To derive the core volume from the apparent volume, the surface layers were characterized using various analysis techniques at PTB and NMIJ [8]. Since the core volume determination was the largest uncertainty source in the previous $N_A$ determinations [4]–[6], the high-accuracy measurements of the core volume at the two national metrology institutes played a crucial role in determining $N_A$ with the smallest uncertainty achieved by the XRCD method to date [8]. Details of the measurement results by PTB were, therefore, provided in [10]–[12]. On the other hand, only a concise summary of the results concerning the core volume measurement at NMIJ was provided in [8]
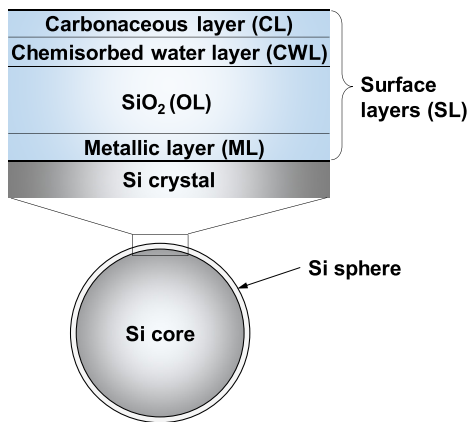
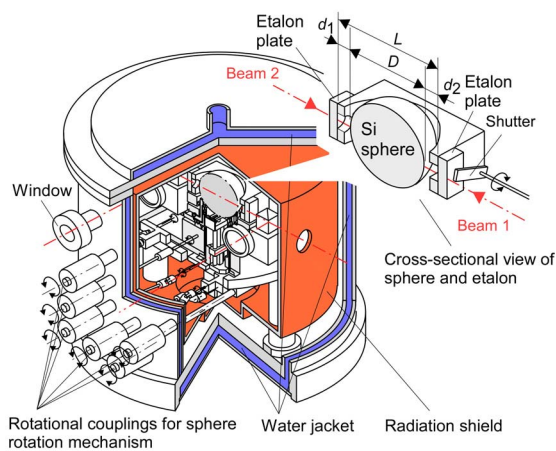Fig. 1.　Model of the surface layers of Si spheres in vacuum.



Fig. 2.　Schematic of the optical interferometer used to measure the volume of Si28kg01a. The sphere and the etalon are installed in a vacuum chamber equipped with an active radiation shield to control the sphere temperature.

and [13]. Details of the measurement at NMIJ including the derivation of the core volume from the apparent volume are, therefore, provided in this paper.

After the change in the definition of the kilogram, NMIJ will realize the new kilogram definition by the XRCD method on the basis of the value of $h$ [14]. The largest uncertainty source in the realization is the Si core volume determination using the interferometer. For the accurate and reliable dissemination of mass standards, the interferometer is, therefore, currently being improved to reduce the uncertainty in the Si core volume determination. Details of the approaches for the improvement are also introduced.

## II. Optical Interferometer

Fig. 2 shows a schematic drawing of the optical interferometer used to determine the volume of the [28]Si sphere by optical frequency tuning [6], [15], [16]. A Si sphere is placed in a fused-quartz Fabry–Perot etalon. The sphere and etalon are installed in a vacuum chamber equipped with an active radiation shield and a water jacket to control the sphere temperature. The pressure in the chamber is reduced to $10^{-2}$ Pa. The light source of the interferometer is an external cavity

diode laser with a center wavelength of 633 nm. A laser beam from the light source is split into two beams (Beam 1 and Beam 2). They are reflected by mirrors toward opposite sides of the etalon. The light beams reflected from the inner surface of the etalon plate and the adjacent surface of the sphere interfere to produce concentric circular fringes. These are projected onto Charge Coupled Device (CCD) cameras. Measurements of the fractional fringe order of interference for the gaps between the sphere and the etalon, $d_1$ and $d_2$, are carried out by phase-shifting interferometry [15], [16]. The sphere diameter $D$ is calculated as $D = L - (d_1 + d_2)$, where $L$ is the etalon spacing. To determine $L$, the sphere is removed from the light path by a lifting device installed underneath the sphere, and Beam 1 is interrupted by a shutter. Beam 2 passes through a hole in the lifting device, and the beams reflected from the two etalon plates produce fringes on a CCD camera. The fringes are also analyzed by phase-shifting interferometry. The volume is determined on basis of the diameter measurement in many directions. The required phase shifts are produced by changing the optical frequency of the diode laser. Its optical frequency is phase locked and controlled based on a phase-stabilized frequency comb referring the time standard UTC(NMIJ), which is used as the national length standard in Japan. Details of the interferometer are given in [14]–[17].

## III. X-ray Photoelectron Spectroscopy System

X-ray photoelectron spectroscopy (XPS) is a surface-sensitive technique for identifying the chemical elements present on a surface. It also provides information on the chemical binding state of the elements. The main component of the XPS system of NMIJ is ULVAC-Phi 1600C equipped with a monochromatic Al K$\alpha$ X-ray source. The pressure in the chamber is reduced to $1.5 \times 10^{-6}$ Pa. A manipulator with five-axis freedom is installed in the chamber to rotate the sphere around the horizontal axis and vertical axis for the mapping of the entire surface. The sphere is placed on two rollers of the manipulator during the measurement. The rollers are made of polyimide to protect the sphere surface from damage during the rotation. Details of the XPS system are given in [18] and [19].

## IV. Spectroscopic Ellipsometer

Spectroscopic ellipsometry (SE) is widely used to measure the thickness of a thin oxide layer (OL) on a Si substrate, since the measurement time is very short and the reproducibility is very high. SE is, therefore, used for rapid topographical measurements of the SiO$_2$ layer thickness on Si spheres [20]. The main component of the spectroscopic ellipsometer of NMIJ is Semilab GES5E. Its spectral bandwidth ranges from 250 to 990 nm. The ellipsometer has a vacuum chamber to characterize the surface layers in vacuum, where the pressure is reduced to $1 \times 10^{-3}$ Pa. A sphere rotation mechanism is also integrated into the vacuum chamber, and the ellipsometric measurement can be carried out over the entire sphere surface. Details of the ellipsometer are provided in [21].
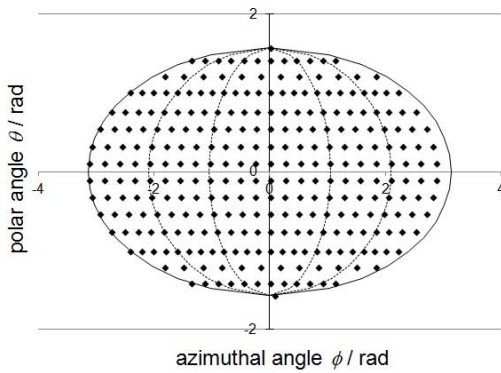
Fig. 3. Mollweide projections of the 145 measurement directions used for the diameter measurement. The sphere surface is divided into small cells with equal areas, and the measurement directions are distributed individually in each cell [15].

## V. Measurement Results

### A. Measurement Sequence

The core volume of Si28kg01a was measured at NMIJ. This $^{28}$Si sphere is one of the two spheres manufactured from the new crystal [8]. The $^{28}$Si sphere was transported from PTB to NMIJ in November 2016 by hand carry. The surface of the sphere was characterized by the XPS system from November to December 2016 and by the spectroscopic ellipsometer in December 2016. The volume was measured by the optical interferometer from January to February 2017.

### B. Cleaning of the $^{28}$Si Sphere

Before each measurement, the $^{28}$Si sphere was cleaned using the procedure employed in the international comparisons of the mass and diameter measurements of a 1-kg Si sphere in the IAC project [22], [23]. In the first step, the sphere was washed with a dilute aqueous solution of a detergent (deconex OP 164, Borer Chemie AG), during which the sphere surface was rubbed manually with nitrile rubber gloves. Next, the sphere was placed under a stream of water purified by a Milli-Q Integral system (Merck KGaA). Finally, ethanol (Infinity Pure grade, Wako Pure Chemical Industries, Ltd.) was poured over the sphere. After cleaning, the sphere was stored in a clean booth overnight. The gloves were also dried in the booth and were used to install the sphere in each apparatus.

### C. Volume Measurement Using Optical Interferometer

A sphere rotation mechanism installed under the sphere was used to measure the diameter in many different directions [24]. In a set of diameter measurements, the diameter was measured in 145 directions distributed nearly uniformly on the sphere surface as shown in Fig. 3 [15]. The set of diameter measurements was repeated 15 times, and the total number of measurement directions was, therefore, 2175. Between each set, the sphere was rotated to distribute the starting point of each set of measurements to the vertices of a regular dodecahedron. Because the 10 directions defined by the vertices of a regular dodecahedron are uniformly distributed, this procedure, therefore, distributed all of the measurement
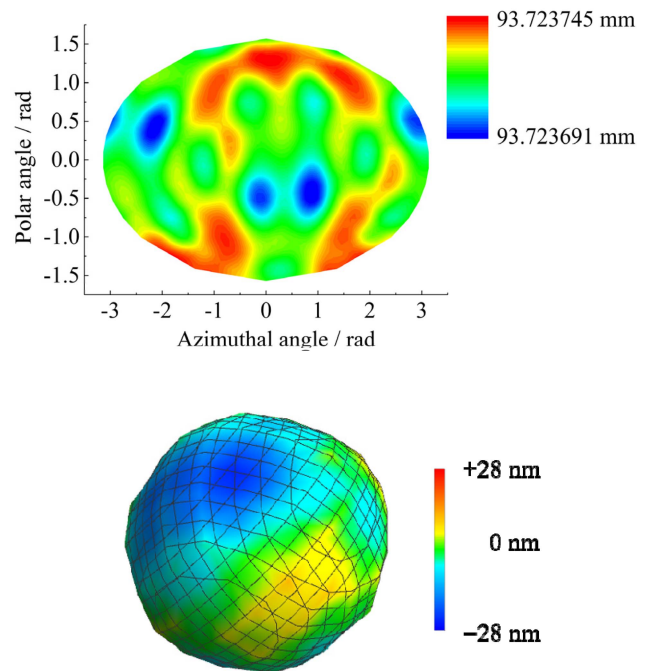


Fig. 4. Mollweide map projection of the diameter (top) and 3-D plot of the diameter (bottom) based on the measurement from the 145 directions. The peak-to-valley value for the diameter is 56 nm.

TABLE I

RESULTS OF THE VOLUME MEASUREMENT OF Si28kg01a
AT 20.000 °C AND 0 Pa BY NMIJ

| Quantity | Value | $u_{c,r}$[a] / $10^{-9}$ |
|---|---|---|
| Apparent diameter[b] | 93.723 723 61(61) mm | 6.5 |
| Actual diameter[c] | 93.723 727 08(65) mm | 7.0 |
| Si core diameter[d] | 93.723 723 70(61) mm | 6.5 |
| Si core volume | 431.069 4323(85) cm$^3$ | 19.6 |

[a] Relative standard uncertainty of each quantity.
[b] Diameter observed by interferometric measurement.
[c] Diameter including the surface layers.
[d] Diameter excluding the surface layers.

points as uniformly as possible [15]. The temperature of the sphere was measured with a standard uncertainty of 0.62 mK using small platinum resistance thermometers (PRTs) inserted in copper blocks in contact with the sphere [25]. The PRTs were calibrated using temperature fixed points in ITS-90. The average measurement temperature was 20.0031 °C. The measured diameters were converted to those at 20.000 °C using the thermal expansion coefficient of an enriched $^{28}$Si crystal [26]. Fig. 4 shows the Mollweide map projection of the diameter based on the measurement from the 145 directions. A 3-D plot of the diameters is also displayed in this figure.

The measured diameter was the apparent diameter, which was not corrected for the phase shift due to the surface layers. The average apparent diameter was 93.723 723 61(61) mm, where the numbers in the parenthesis indicate the standard uncertainty of the last digits. This expression is used throughout this paper. The apparent diameter is given in Table I. The relative standard uncertainty of the volume measurement was
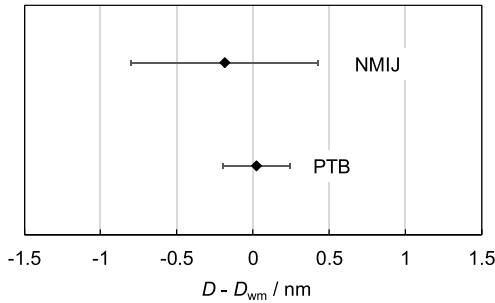
Fig. 5. Comparison of the average apparent diameters of Si28kg01a measured at PTB and NMIJ. The difference from the weighted mean diameter, $D_{wm}$, calculated from the diameters measured at the two national metrology institutes is plotted. Horizontal error bars: standard uncertainty of each value.

$20 \times 10^{-9}$. Table II shows the uncertainty budget for the determination of the apparent volumes. The largest uncertainty source is the diffraction effect [27]. Details of each uncertainty source are given in [15].

The volume of Si28kg01a was also measured at PTB [10]. The average apparent diameter at 20.000 °C and 0 Pa was 93.723 723 82(22) mm. Fig. 5 shows a comparison of the average apparent diameters measured at PTB and NMIJ. The average diameters obtained using the two interferometers with different optical configurations and phase-shifting algorithms show excellent agreement within their uncertainties. The difference between the average diameters is 0.2 nm, showing the high reliability of the diameter and volume measurements in the IAC project.

To rigorously evaluate the results of the comparison of the sphere diameter measurement, the equivalence of the temperature sensors of PTB and NMIJ is essential. The equivalence was confirmed by the following procedure. The temperature sensors used for the volume measurement at PTB and NMIJ were calibrated using temperature fixed point cells at each institute. All the cells were traceable to the primary temperature standards at each institute. The international mutual equivalence of the primary temperature standards was confirmed by the CIPM key comparisons CCT-K7 and CCT-K3 [16]. In addition, the nonuniqueness of the sensors was checked in May 2016 by temperature comparisons in the IAC project [28]. In comparison, the temperature sensor of NMIJ was transported to PTB. The sensors of the two institutes were calibrated using the temperature fixed points at PTB and then were immersed in an electronic temperature reference point apparatus thermostated to 20 °C [29]. The difference in the temperature readings of the two sensors was less than 0.1 mK, showing that the difference in the nonuniqueness of the two sensors was less than 0.1 mK. The Si sphere temperature measurement uncertainty of 0.1 mK corresponds to the sphere diameter measurement uncertainty of 0.03 nm. The temperature sensors used for the volume measurement at PTB and NMIJ were, therefore, sufficiently equivalent to allow the comparison of the diameters.

### D. Surface Analysis by XPS

The XPS spectra were measured at 52 points distributed near uniformly on the sphere surface [8]. The 52 measurement

TABLE II
UNCERTAINTY BUDGET IN THE DETERMINATION OF THE APPARENT VOLUME, $V_{app}$, OF Si28kg01a AT 20.000 °C AND 0 Pa

| Uncertainty source | Standard uncertainty | Relative standard uncertainty in $V_{app}$ |
|---|---|---|
| Frequency of laser | 33 kHz | $0.2 \times 10^{-9}$ |
| Sphere temperature | 0.62 mK | $4.8 \times 10^{-9}$ |
| Interferogram analysis | 0.3 nm | $9.6 \times 10^{-9}$ |
| Effect of Gouy phase shift | 0.5 nm | $16.0 \times 10^{-9}$ |
| Frequency change corresponding to one integer order | 10 MHz | $0.4 \times 10^{-9}$ |
| Standard deviation of the mean volume | | $3.5 \times 10^{-9}$ |
| Relative combined standard uncertainty | | $19.6 \times 10^{-9}$ |

points were defined by dividing the sphere surface into small cells with equal areas [14], and the measurement points were distributed individually in each cell. Fig. 6 shows the distribution of the measurement points, in which the measurement points in XPS are shown by solid diamonds.

*1) Oxide Layer:* The thickness of the OL was determined by analyzing the XPS Si $2p$ core-level spectra, in which peaks corresponding to the $SiO_2$ layer and the interfacial oxide ($Si_2O$) layer were observed. For rigorous evaluation of the OL thickness, accurate values of the attenuation lengths for the Si $2p$ electrons in $SiO_2$ and $Si_2O$ were determined by an SI-traceable X-ray reflectometry system at NMIJ [20] using Si flat samples with different thicknesses of thermal $SiO_2$. The average thicknesses of the $SiO_2$ layer $d_{SiO2}$ and that of the $Si_2O$ layer $d_{Si2O}$ at the 52 points were estimated to be 0.91 (10) nm and 0.14 (1) nm, respectively. The average OL thickness $d_{OL}$ was, therefore, estimated to be 0.95 (10) nm using $d_{OL} = d_{SiO2} + 0.25 d_{Si2O}$. Details of the determination of the attenuation lengths and the thickness of the $SiO_2$ and $Si_2O$ layers are summarized in [8], [14], and [19]. The value of $d_{OL}$ was used to calibrate the spectroscopic ellipsometer. Details of the calibration are provided later.

*2) Carbonaceous Layer:* In the XPS C 1s core-level spectra, peaks corresponding to the C–C/H and C–O bonds were observed. The main constituents were deduced to be both ethanol and a hydrocarbon [8]. By analyzing the spectrum at the 52 measurement points, the average thickness of each sublayer was estimated to be 0.30(6) nm and 0.13(3) nm for the ethanol sublayer and hydrocarbon sublayer, respectively. Consequently, the average carbonaceous layer (CL) thickness $d_{CL}$ was estimated to be 0.43(7) nm.

*3) Metallic Layer:* In the XPS Na 1s core-level spectra, the Na 1s electrons were detected at all 52 measurement points. This was attributed to the metallic Na layer. The average metallic layer (ML) thickness at the 52 measurement points $d_{ML}$ was estimated to be 0.05(2) nm.

### E. Surface Analysis by Spectroscopic Ellipsometry

The spectroscopic ellipsometer was used to evaluate the distribution of the OL thickness. First, the measurement was
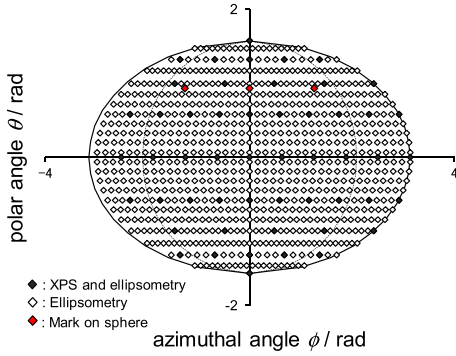
Fig. 6. Distribution of the measurement points in XPS and ellipsometry (◆) and those in only ellipsometry (◇) [14]. The measurement points in XPS were also characterized by ellipsometry. The measurement positions were identified using the three marks present on the sphere.

performed at the same 52 points as those used for the XPS measurement. The measurement points were identified using three marks on the sphere. The ellipsometric data were analyzed on the basis of a surface model assuming a SiO$_2$ layer on a Si substrate. To calibrate the ellipsometer, the following equation was used:

$$d_{\mathrm{OL}} = d_{\mathrm{XPS}} = d_{\mathrm{SE}} + c \qquad (1)$$

where $d_{\mathrm{OL}}$ is the OL thickness, $d_{\mathrm{XPS}}$ is the OL thickness measured by XPS, $d_{\mathrm{SE}}$ is the OL thickness measured by SE, and $c$ is the calibration constant. The value of $c$ was determined from the average values of $d_{\mathrm{XPS}}$ and $d_{\mathrm{SE}}$ measured at the 52 points.

After the calibration, the ellipsometric measurement was performed at 812 points distributed near uniformly on the sphere surface shown in Fig. 6. The measured ellipsometric data were analyzed on the basis of the surface model assuming a SiO$_2$ layer on a Si substrate, and the determined thickness was corrected by $c$ to obtain $d_{\mathrm{OL}}$. The measurement at the 812 points was repeated three times, and the total number of measurement points was, therefore, 2436. Between each set of measurement at the 812 points, the sphere was oriented so as to distribute all the measurement directions as uniformly as possible. The average OL thickness at the 2436 points was estimated to be 0.93 nm, which was used as the average OL thickness $d_{\mathrm{OL}}$ to derive the core volume.

The standard deviation of the mean of $d_{\mathrm{OL}}$ for the three sets of measurements at the 812 points was 0.01 nm. In the calibration of the ellipsometer, the standard deviation of the mean of the difference between $d_{\mathrm{XPS}}$ and $d_{\mathrm{SE}}$ at the 52 points was 0.02 nm. By combining these uncertainties with the uncertainty of the OL thickness determination by XPS, the uncertainty of the average OL thickness determination by XPS and SE was estimated to be 0.10 nm.

Table III summarizes the thickness of each layer. The thickness of the chemisorption water layer $d_{\mathrm{CWL}}$ was derived from the value measured by Mizushima [30], where Si disks were outgassed at 500 °C and the mass of water adsorbed on the disks was evaluated by gravimetry.

Table IV summarizes the refractive index and extinction coefficient of each layer used in the derivation of the Si core

TABLE III

THICKNESS OF EACH LAYER

| Layer | Thickness / nm | Measurement method |
|---|---|---|
| OL | 0.93(10) | XPS, SE |
| CL | 0.43(7) | XPS |
| CWL | 0.28(8) [a] | Gravimetry |
| ML | 0.05(2) | XPS |

OL: Oxide layer, CL: Carbonaceous layer, CWL: Chemisorbed water layer, ML: Metallic layer, XPS: X-ray photoelectron spectroscopy, SE: Spectroscopic ellipsometry.
[a] Value from [8].

TABLE IV

UNCERTAINTY BUDGET IN THE DETERMINATION OF THE PHASE SHIFT CORRECTION $\Delta d_0$ TO OBTAIN THE Si CORE DIAMETER

| Uncertainty source | Value | Standard uncertainty in $\Delta d_0$ |
|---|---|---|
| Thickness | | |
| CL | 0.43(7) nm | 0.009 nm |
| CWL | 0.28(8) nm | 0.009 nm |
| ML | 0.05(2) nm | 0.023 nm |
| OL | 0.93(10) nm | 0.016 nm |
| Refractive index | | |
| CL | 1.39(14) | 0.024 nm |
| CWL | 1.33(1) | 0.001 nm |
| ML | 0.05(1) | 0.000 nm |
| OL | 1.457(10) | 0.004 nm |
| Si | 3.881(1) | 0.000 nm |
| Extinction coefficient | | |
| CL | 0.0(1) | 0.001 nm |
| CWL | $1.54(1.00) \times 10^{-8}$ | 0.000 nm |
| ML | 2.6(1) | 0.004 nm |
| OL | 0.0(1) | 0.000 nm |
| Si | 0.019(1) | 0.014 nm |
| Combined standard uncertainty | | 0.041 nm |

volume from the apparent volume. The optical constants of Na at 633 nm were estimated from those in the wavelength range from 546 to 751 nm given in [31]. The optical constants of the CL were estimated from those of the ethanol layer [14] and hydrocarbon layer [14] taking into account the ratio of the thicknesses of the two sublayers. The optical constants of the other layers were taken from [14].

## VI. CORE VOLUME

To determine the Si core volume, the total phase retardation $\delta$ upon reflection at the sphere surface was calculated on the basis of the procedure described in [16]. To calculate $\delta$, the thickness and optical constants of each layer summarized in Table III were used. The phase shift upon reflection $\delta - \pi$ was $-0.034(6)$ rad. The effect of this phase shift on the gap measurement, $\Delta d$, was estimated to be 1.74(12) nm.

TABLE V

CORRELATION COEFFICIENTS AMONG THE Si CORE
VOLUMES DETERMINED BY NMIJ

| | $V_{2011}$ [a] | $V_{2015}$ [b] | $V_{2017}$ [c] | $V_{28Sikg01a}$ [d] |
|---|---|---|---|---|
| $V_{2011}$ | 1.00 | 0.07[e] | 0.07[e] | 0.07[f] |
| $V_{2015}$ | | 1.00 | 0.17[e] | 0.17[f] |
| $V_{2017}$ | | | 1.00 | 0.17[f] |
| $V_{28Sikg01a}$ | | | | 1.00 |

[a] Si core volume of AVO28-S5 [4]
[b] Si core volume of AVO28-S5c [5]
[c] Si core volume of AVO28-S5c [6]
[d] Si core volume of Si28kg01a in this work
[e] Correlation coefficient given in [6]
[f] Correlation coefficient evaluated in this work

This means that the actual diameter including the surface layers is larger than the apparent diameter by 3.47(24) nm. The actual diameter $D_{actual}$ is, therefore, obtained by $D_{actual} = D_{app} + 2\Delta d$ and is summarized in Table I. The phase shift correction $\Delta d_0$ to obtain the Si core diameter was determined as $\Delta d_0 = 2(\Delta d - d_{total})$, where $d_{total}$ is the sum of the thickness of each layer. The value of $\Delta d_0$ was estimated to be 0.092(41) nm. Table IV shows the uncertainty budget for the determination of $\Delta d_0$. The thickness and optical constants of each layer used in the calculation of $\delta$ are also given in Table IV. The largest uncertainty sources are the thickness and refractive index of the CL and the thicknesses of the OL and the ML. The Si core diameter is obtained by $D_{core} = D_{app} + \Delta d_0$ and is summarized in Table I with the core volume $V_{core}$. The relative uncertainty of the core volume determination is $20 \times 10^{-9}$.

In Table IV, the large uncertainty was attributed to the refractive index of the CL compared with those of the other layers. This is due to the lack of information to clarify the chemical composition of the CL.

## VII. CORRELATION COEFFICIENT WITH THE PREVIOUS Si CORE DETERMINATIONS

The IAC project reported four $N_A$ values in 2011 [4], 2015 [5], and 2017 [6], [8]. The correlation coefficient among the four $N_A$ values was estimated to make the $N_A$ values usable for the least-squares adjustment of the Planck constant by the CODATA adjustment in 2017 [6], [32]. Since the Si core volume determination was the largest uncertainty source in the $N_A$ determinations, the correlation coefficient among the Si core volume determinations should be accurately evaluated. The correlation coefficient of the Si core volume determined in this paper with those in the previous works was, therefore, evaluated using the procedure given in [6]. These are given in Table IV with the correlation coefficient among the Si core volumes determined in the previous works [4]–[6] given in [6]. The correlation coefficients in Table V were used to determine the correlation coefficients among the four $N_A$ values determined by the IAC project [32].

## VIII. AVOGADRO CONSTANT

The relative uncertainty of the weighted mean of the average Si core volumes determined at PTB and NMIJ was estimated to be $6.7 \times 10^{-9}$ [8]. This volume was used to determine $N_A$

in 2017. The relative uncertainty of the $N_A$ determination was estimated to be $1.2 \times 10^{-8}$, which is the smallest uncertainty achieved by the XRCD method to date [8]. In the previous $N_A$ determinations, the relative uncertainties of the Si core volume determinations were in the range from $2 \times 10^{-8}$ to $3 \times 10^{-8}$ and were the largest uncertainty sources [4]–[6]. The fundamental reduction of the uncertainty of the Si core volume measurement, therefore, played a crucial role in determining $N_A$ with the smallest uncertainty.

The $N_A$ value was used as an input datum for the CODATA 2017 special adjustment to provide the Planck constant $h$ in the new definition of the kilogram [9]. The new definition will be effective from May 20, 2019 [33].

## IX. IMPROVEMENT OF THE INTERFEROMETER

After the change in the definition of the kilogram, the NMIJ interferometer will be used to realize the kilogram based on the value of $h$ using the XRCD method [14], [34]. The relative uncertainty of the realization was estimated to be $24 \times 10^{-9}$, corresponding to 24 µg for 1 kg [14]. The largest uncertainty source was the Si core volume determination of a $^{28}$Si-enriched sphere. For the more accurate realization of the kilogram at NMIJ, the interferometer is being improved. As given in Table II, the largest uncertainty source in the diameter measurement is the phase correction due to the diffraction effect [16], [27]. The correction was estimated on the basis of the calculation of the Gouy phases of the beams used for the diameter measurement. The Gouy phases were calculated from the measurement of the beam waist and the ray tracing of the beams and the size of the beam waist was determined from the measurement of the beam size by a beam profiler at several positions in the interferometer [16], [27]. The uncertainty of this correction has already been estimated by simulation to be 0.1 nm [35]. However, the reliability of the simulation result has not been verified experimentally. The uncertainty of the correction is, therefore, conservatively overestimated to be 0.5 nm. The reliability of the uncertainty estimation by the simulation [35] is, therefore, being experimentally examined, in which the change in the apparent diameter introduced by the change in the positions of the optical components in the interferometer is measured. The change in the apparent diameter will be compared with that estimated from the calculation of the Gouy phases.

Another approach to verify the reliability of the uncertainty estimation by the simulation is to significantly change the laser wavelength of the light source. Although the laser wavelength of the light source is presently 633 nm, the effect of changing the laser wavelength was estimated theoretically by changing the laser wavelength in the calculation of the Gouy phases [16], [27] and the results are summarized in Fig. 7. The diffraction effect on the diameter measurement (♦) was estimated by summing the effect on the gap measurement ($\Delta$) and that on the etalon spacing measurement ($\Diamond$). The diffraction effect on the diameter measurement at 633 nm was estimated to be 0.5 nm [15]. If the wavelength is changed to 430 nm, the diffraction effect is changed by 0.5 nm and is estimated to be 0.0 nm as shown in Fig. 7. This estimated
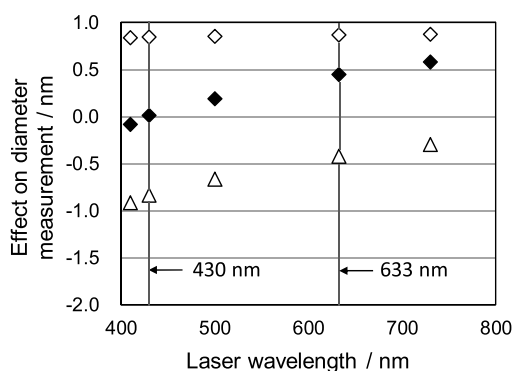
Fig. 7. Calculated diffraction effect on the diameter measurement (♦) for different laser wavelengths. The diffraction effect on the diameter measurement was estimated by summing the effect on the gap measurement (△) and that on the etalon spacing measurement (◇).

change in the apparent diameter will be compared with that determined from the diameters measured by using two lasers operated at 633 and 430 nm to verify the reliability of the uncertainty estimation by the simulation [35]. For this verification, a new laser system with a single-mode tuning range of 20 GHz at a center wavelength of 426 nm is being developed at NMIJ. Its optical frequency also will be phase locked and controlled based on a frequency comb.

After verifying the reliability of the uncertainty estimation by the simulation using the two approaches noted earlier, the uncertainty of the diffraction effect in Table II will be reduced to 0.1 nm, improving the relative uncertainty of the Si core volume measurement to $1.2 \times 10^{-8}$. As a result of this improvement, the relative uncertainty of the realization of the kilogram will be reduced to $1.7 \times 10^{-8}$, corresponding to 17 µg for 1 kg. The verification of the simulation result by the two approaches is currently under way.

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