Far-infrared elastic scattering proposal for the Avogadro Project's silicon spheres

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ABSTRACT

Avogadro constant determines the number of particles in one mole of a substance, thus relating the molar mass of the substance to the mass of this substance. Avogadro constant is related to Système Internationale base units by defining the very concept of chemical quantity. Revisions of the base units created a need to redefine the Avogadro constant, where a collaborative work called the Avogadro Project is established to employ optical interferometry to measure the diameter of high quality 100 mm silicon spheres. We propose far-infrared spectroscopy for determining the Avogadro constant by using elastic scattering from the 100 mm Avogadro Project silicon spheres. Similar spectroscopic methods are already in use in the near-infrared, relating whispering gallery modes of the 1 mm silicon spheres to the diameter of the spheres. We present numerical simulations in the far-infrared and the near-infrared, as well as spatially scaled down elastic scattering measurements in the near-infrared. These numerical and experimental results show that, the diameter measurements of 100 mm single crystal silicon spheres with elastic scattering in the far-infrared can be considered as an alternative to optical interferometry.

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1. Introduction

The kilogram is the Système Internationale (SI) unit of mass, which is defined with the mass of a material artifact kept at Bureau International des Poids et des Mesures (BIPM) [1,2], where Avogadro constant (N_A) relates the number of atoms in the artefact to the molar mass of the material. However, for the definition of a base unit, it is imperative that, the defining quantity does not experience a change in its physical state with the passage of time. Thus, a need of redefinition for the kilogram standard has arisen [3]. A global effort has been ongoing to redefine the mass of the prototype in terms of fundamental constants or the mass

of an atom, which can have small enough uncertainty to replace the current definition of the kilogram [4]. In this case, recalculating N_A with sufficient accuracy became the focus. In the past 40 years, using interferometric methods to measure the diameter of an extremely well-shaped and pure silicon (Si) sphere with low uncertainty became a fundamental approach [5].

The use of optical interferometry for the diameter measurement of spheres was first setup by Saunders [6]. Over the years, the improvement and the novelty in interferometry resulted in more precise and accurate measurements. The material choice for the sphere was Si, where increasing production purity of the Si spheres helped the researchers. Diameter measurements for Si spheres by using interference yielded an uncertainty on the orders of 10^{-8} [7,8], and use of different interferometry methods such as scanning type and Fizeau interferometer helped improve the measurement conditions [9–12]. Recently, laser interferometry with a flat etalon on two spheres [13] combined with the modern mathematical methods [14] helped bringing down the uncertainty in diameter measurements to less than 1 nm. Recently, relative standard

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uncertainty in these spheres is reported to be on the order of 2.2×10^{-8} [15].

Avogadro spheres, with their 100 mm diameters, are macroscopic objects. On the microscopic scale, Si microspheres [16] provide confinement of light in all three dimensions (3D), and can act as optical fibre coupled [17] 3D optical microcavities with high quality factor (Q-factor) whispering gallery modes (WGMs), which sensitively depend on the diameter of the sphere [18–20]. Size measurement by using light scattering for microparticles is well-known [21–23]. Dielectric spheres with dimensions much larger than the wavelength of light can sustain high Q-factor WGMs [24]. Schiller et al. performed resonance experiments on a 38 mm diameter fused-silica sphere [25].

Additionally, far-IR time domain WGM spectroscopy of a Si cylinder with a diameter of 5 mm and excited by a Si waveguide with a thickness of 100 μm at 1 THz [26] has been reported. Here we propose, for the first time to the best of our knowledge, far-IR elastic scattering WGM spectroscopy to measure the diameter of the Avogadro Project Si spheres. Our proposal offers the benefit of using macroscopic Si spheres with the excitation of their WGMs to measure their diameter with far-IR spectroscopy. We structure the paper as follows. We demonstrate our approach experimentally with the elastic scattering measurements from a 1 mm single crystal Si sphere in the near-IR. We perform Gaussian beam elastic scattering simulations for the 1 mm Si sphere and for the 100 mm Avogadro sphere. Finally, we propose a far-IR resonance spectroscopy setup for the elastic scattering diameter measurements for the 100 mm Avogadro sphere.

2. Avogadro Project

The Avogadro Project proposes to redefine the kilogram in terms of one of the Avogadro constant (N_A) as [27]:

$$1\text{kg} = \{N_A\} \frac{m(^{12}\text{C})}{0.012} \quad (1)$$

where $m(^{12}\text{C})$ is the molar mass of carbon. The conditions for redefinition of the kilogram have been formulated, and a relative uncertainty of 2 parts in 10^8 is required for redefinition of unit of mass [28]. For finding N_A , a single crystal ^{28}Si isotope is used [29]. Si crystal was chosen due to the presence of already mature semiconductor industry based on Si with well-known mechanical, thermal, electrical, and chemical properties, so that, highly pure and almost perfect Si spheres can be manufactured [30,31]. Counting the atoms in a single crystal Si sphere for determining N_A for redefinition of the mass unit was previously outlined [32]. X-ray crystal density molar mass (XRCDMM) method can be used to find N_A by utilizing the macroscopic density ρ of the ideal Si crystal, the molar mass $m(^{28}\text{Si})$ of Si, and the unit cell volume a^3 of the Si crystal as follows [33,34]:

$$N_A = nV_{\text{mol}}/a^3 \quad (2a)$$

$$V_{\text{mol}} = m(^{28}\text{Si})/\rho \quad (2b)$$

where V_{mol} is the molar volume of Si, and n the number of atoms in the unit cell. Thus, XRCDMM method facilitates the determination of N_A in terms of macroscopic parameters instead of counting a large number of atoms with a very small uncertainty. The density of the Si can be found by dividing the mass of the sphere M by the volume V as,

$$\rho = M/V \quad (3)$$

The volume of the Si sphere can be found by measuring its diameter. Uncertainty in the diameter measurement decides the uncertainty in the volume calculations. To achieve an accuracy of 0.01 ppm in volume, the diameter must be known to a range

Fig. 1. Scanning electron micrograph (SEM) of the 1 mm diameter silicon sphere.

Fig. 2. Experimental setup for near-IR 90° elastic scattering and 0° transmission spectroscopy of the 1 mm Si sphere.

of 0.6 nm, i.e., within one atomic spacing. In diameter measurements the effect of the surface native and non-native oxides (with a thickness on the order of 1.07 nm) has a relative uncertainty of 3.4×10^{-8} [35].

3. Near-IR elastic scattering of the 1 mm silicon sphere

Fig. 1 shows a scanning electron micrograph (SEM) of the 1 mm diameter Si sphere (Ball Semiconductor). The 1 mm Si sphere was produced by the solidification of melted droplets of Si falling through a tube. After the rapid solidification, single crystal silicon spheres are evidenced by the formation of 8 horns in the 111 crystal directions. These horns are attributed to the volume expansion of Si from liquid to solid phase, and the difference of the solidification speed in crystal orientation, as the 111 direction has the slowest solidification speed, the excess liquid is maintained in 111 direction, until the end of the solidification with the formation of the horns. Later, the Si beads were lapped and polished mechanically to make a mirror-like finish with no surface defects and damages [36,37].

Fig. 2 shows our near-IR WGM excitation setup. Tunable distributed feedback (DFB) diode lasers (operating at 1428 and

Fig. 3. Near-IR (1428 nm) 90° elastic scattering and 0° transmission spectra from the 1 mm silicon sphere.

Fig. 4. Near-IR (1472 nm) 90° elastic scattering and 0° transmission spectra from the 1 mm silicon sphere.

1472 nm) are used to excite the WGMs of the Si sphere. The DFB lasers are driven by a laser diode controller (LDC) connected to a computer, where the output wavelength of a DFB laser is controlled by the LabView (National Instruments) software. The DFB laser output is fed to the single mode fibre (SMF), which is connected to the optical fibre half coupler (OFHC). The OFHC is manufactured from polished SMF, which couples 99.4% of the light passing through the coupler, when an index-matching oil is placed on the OFHC [38]. The SMF cladding thickness on the OFHC surface is on the order of a micrometer.

The Si microsphere is placed on the top of the OFHC, where the light in the SMF is evanescently coupled to the Si microsphere [39,40]. An optical microscope collects the elastically scattered light from the Si microsphere at 90°. This collected light is incident on the InGaAs photodetector (PD), which is connected to a digital storage oscilloscope (DSO). The 0° transmitted signal is collected by the InGaAs power head (PH) of an optical multimeter (OMM). General purpose interface bus (GPIB) protocol is used to connect the DSO and the OMM to the data acquisition computer.

4. Near-IR elastic scattering and transmission spectra for the 1 mm silicon sphere

Fig. 3 shows the 90° elastic scattering and the 0° transmission for near-infrared (near-IR) light (with a central wavelength at 1428 nm). Fig. 4 shows the 90° elastic scattering and 0° transmission for near-IR light (with central wavelength at 1472 nm). The

elastic light scattering spectra in Figs. 3 and 4 show the expected dips in the 0° transmission and the corresponding peaks in the 90° elastic scattering. In Fig. 3, the experimentally measured mode spacing of the WGMs is 0.244 nm for the DFB laser at 1428 nm, and correlates well with the calculated mode spacing of 0.249 nm in the theoretical model. In Fig. 4, the experimentally measured mode spacing of the WGMs is 0.252 nm for the DFB laser with central wavelength 1472 nm, and correlates well with the 0.265 nm of the theoretical model. Using Eq. (8), the Q-factor is found to be 6×10^4 for the WGMs of Fig. 3, and $\approx 5 \times 10^4$ for the WGMs of Fig. 4. These relatively broad WGM linewidths can be reduced by several orders of magnitude with improved experimental conditions such as using narrower linewidth lasers.

5. Comparison of 100 mm Avogadro Project Si sphere and 1 mm Si sphere

The most recent determination of the Avogadro constant, $N_A = 6.022\,140\,76(12) \times 10^{23} \text{ mol}^{-1}$ achieved the target of relative standard uncertainty of 2.0×10^{-8} . This is currently the most accurate determination of a fundamental constant, which can be used for a new definition of the kilogram. Table 1 contains the x-ray crystal density (XRCD) method results of this recent work. In Table 1, we also compare the properties of the 100 mm Avogadro sphere and the 1 mm Si sphere.

By taking the refractive index of Si at 1500 nm as 3.48 [41], and using Eqs. (6) and (7) on Fig. 3 results, the diameter of the 1 mm sphere was calculated to be 1022 μm. From the SEM (ZEISS Ultra plus field emission SEM) image of our 1 mm Si sphere (Fig. 1), the diameter was found to be 1017 μm, agreeing to within 0.5% with our spectral measurement. Additionally, by taking the values of the molar mass $m(^{28}\text{Si})$, the lattice constant, and the N_A from CODATA [42], the mass of the 1 mm sphere was calculated to be 1.222 mg (Table 1). The mass (Mettler Toledo Excellence Plus XP6 Micro Balance) of the 1 mm sphere was found to be 1.257 mg agreeing with the calculated value to within 3%.

If the size obtained is the correct one, it should be correct, when measured in different spectral regions, i.e., near IR, mid-IR, far-IR, or in the same spectral region at different spectral bands, different thicknesses of the cladding on the SMF, different positions of the collection optics, and different polarizations of the excitation laser. All these potential multiplexed spectroscopic measurements will contribute to the precision of the measurement of the diameter of the Avogadro sphere.

In addition, the sphericity measurements can be performed by rotating the sphere in various directions and acquiring the respective spectra. If an excitation laser close to the absorption bands of silicon is used, it would be possible to heat up the silicon sphere, and study the thermal tuning of the elastic scattering spectra.

6. GLMT simulations for the 1 mm Si sphere and the 100 mm Avogadro Project Si sphere

Our numerical simulations of the 1 mm Si sphere and the 100 mm Avogadro sphere uses Lorenz–Mie theory [43]. An incident electromagnetic wave can be trapped inside the surface of a dielectric sphere with the help of total internal reflection (TIR) and result in WGMs [44]. WGMs manifest themselves as peaks in the elastic scattering spectrum as a function of the size parameter:

$$x = \frac{\pi \phi \mu_o}{\lambda} \quad (4)$$

where ϕ is the diameter of dielectric sphere, λ the vacuum wavelength of light, and μ_o the refractive index of the surrounding medium. The variation of the wavelength as a function of the vari-

Table 1

Experimental parameter comparison of the 100 mm far-infrared Avogadro sphere and the 1 mm near-infrared silicon sphere.

parameter	symbol	100 mm silicon sphere	reference	1 mm silicon sphere	reference
silicon molar mass (g mol ⁻¹)	m(²⁸ Si)	27.976 970 09(15)	15	27.976 970 09(15)	42
silicon atoms per unit cell	n	8	15	8	42
silicon lattice constant (pm)	a	543.099 6219(10)	15	543.102 050 4(89)	42
silicon density (kg.m ⁻³)	ρ	2320.070 943(46)	15	2320.070 959(49)	42
Avogadro constant (10 ²³ mol ⁻¹)	N _A	6.022 140 72(13)	15	6.022 140 82(18)	42
standard uncertainty (10 ²³ mol ⁻¹)	δN_A	2.0 × 10 ⁻⁸	15	3.0 × 10 ⁻⁸	42
silicon sphere diameter (mm)	ϕ	93.710811 11(62)	15	1.020(03)	this work
silicon sphere volume (cm ³)	V	430.891 2891(69)	15	0.004 445 1776(09)	this work
silicon sphere mass (g)	M	999.698 359(11)	15	0.001 222(05)	this work
silicon resistivity (k Ω cm)	σ^{-1}	10	51	10	53
silicon refractive index	μ	3.40	57	3.48	41
silicon absorption coefficient (cm ⁻¹)	α	0.02	51	0.01	42
silicon sphere quality factor	Q	10 ⁸	proposed work	5 × 10 ⁴	this work
excitation wavelength (nm)	λ	150,000	proposed work	1500(50)	this work
excitation frequency (THz)	ν	2	proposed work	200(07)	this work
silicon sphere size parameter	x	2094	proposed work	2094	this work
silicon sphere WGM spacing (nm)	$\Delta\lambda$	25.5	proposed work	0.253(11)	this work

ation of the size parameter x is given as:

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta x}{x} \quad (5)$$

The mode spacing Δx between two WGMs, with consecutive polar angular mode numbers and with the same radial mode order as a function of size parameter x , can be calculated as [45]:

$$\Delta x = \frac{\tan^{-1}\sqrt{m^2 - 1}}{\sqrt{m^2 - 1}} \quad (6)$$

where $m = \mu / \mu_o$ is the relative refractive index ratio, and μ is the refractive index of the sphere. We have to keep in mind that, Eq. (6) is an estimate to the overall mode spacing, and more accurate determination of the Avogadro Number can be obtained by fitting the measured spectra for different combinations of mode numbers. Then, Eqs. (4)–(6) can be used to find the diameter of dielectric sphere as [46]:

$$\phi = \left[\frac{\lambda^2}{\Delta\lambda} \right] \left[\frac{\Delta x}{\pi} \right] = \left[\frac{\lambda^2}{\Delta\lambda} \right] \left[\frac{\tan^{-1}\sqrt{m^2 - 1}}{\pi \sqrt{m^2 - 1}} \right] \quad (7)$$

One of the main parameters of the spherical resonator is the Q-factor of the WGMs, which is a measure of the sensitivity of the resonator. The Q-factor is spectrally defined as:

$$Q = \frac{\lambda_R}{\delta\lambda} \quad (8)$$

where λ_R is the resonant wavelength, and $\delta\lambda$ the linewidth of the resonance peak [47]. For high precision measurements, high Q-factor Si spherical resonators, with minimum optical losses due to absorption and scattering, are desirable. High Q-factors can be obtained, if the sphere is protected from the atmosphere, and the losses are controlled during the manufacturing and the storage stage. The limit for achieving high “material Q-factor” is defined by [48]:

$$Q = \frac{2\pi\mu}{\lambda\alpha} \quad (9)$$

where α the absorption coefficient of material. The highest Q recorded for Si is on the order of 10⁶ for planar Si microdisk resonators [49], and 10⁸ for chemically etched wedge-resonators on Si chip [50].

The quality factor of the WGMs for the Si spheres should be affected with non-negligible absorption of the Si. This intrinsic material loss should limit the unloaded quality factor. The lowest absorption coefficient of the Si reported at 150 μ m is around 0.02 cm⁻¹ at room temperature for Si resistivity more than 10 k Ω cm [51]. This absorption coefficient is on the same order of

magnitude as the absorption coefficient of Si, i.e., 0.01 cm⁻¹ [52] in near-IR at 1.5 μ m at room temperature for Si resistivity on the order of 10 k Ω cm [53].

Elastic scattering simulations were performed using a Gaussian beam excitation algorithm [54] of generalized Lorenz–Mie theory (GLMT) with transverse electric (TE) and transverse magnetic (TM) polarization incident beams on the 1 mm Si sphere and the 100 mm Avogadro sphere. The half-width-at-half-maximum (HWHM) of the Gaussian beam ω_o , was chosen as 56 μ m, and the impact parameter b as 556 μ m for the 1 mm sphere. The impact parameter b defines the vertical distance of the microsphere centre to the excitation source as defined by [55]:

$$b = (2n + 1) \frac{\phi}{x} \quad (10)$$

The impact parameter associates the angular momentum quantum number n of the excited WGMs to the lateral distance of the beam to the centre of the sphere through the localization principle [56].

The HWHM of the Gaussian beam ω_o , was chosen as 5.513 mm and impact parameter b as 55.513 mm for the 100 mm Si sphere. The rather high HWHM values were chosen due to computational limitations. To assure SMF excitation, these values of ω_o should be reduced by an order of magnitude. The far-IR wavelength of operation is chosen as 150 μ m (2 THz), i.e., 100 times greater than the near-IR wavelength of 1.5 μ m (200 THz). The refractive index at 150 μ m is taken as 3.4 [57].

Figs. 5–8 show the GLMT elastic scattering simulations for transverse electric (TE) and transverse magnetic (TM) polarizations for the 1 mm Si sphere and the 100 mm Avogadro sphere. The size parameter was kept at the same value of $x=2094$ for both Si spheres. The spectral mode spacing of 0.25 nm was found for the 1 mm Si sphere, which correlates well with the experimentally observed value (Figs. 3 and 4). The 25.5 nm spectral mode spacing found for the 100 mm Avogadro sphere is 100 times greater, due to the 100 times scaling of the Si sphere sizes and the spectral ranges. Numerically simulated elastic scattering calculations of the Avogadro sphere show promise for the proposed far-IR elastic scattering diameter measurement method.

7. Proposed far-IR elastic scattering setup for the 100 mm Avogadro sphere

Fig. 9 shows our proposed far-IR elastic scattering setup for measuring the diameter of the Avogadro sphere. The tunable laser to be used as the excitation source can operate at a wavelength of 150 μ m (2 THz), therefore keeping the size parameter of both

Fig. 5. Near-IR (1427 nm) TE 90° elastic scattering and 0° transmission simulation spectra for the 1 mm silicon sphere.

Fig. 6. Near-IR (1427 nm) TM 90° elastic scattering and 0° transmission simulation spectra for the 1 mm silicon sphere.

Fig. 7. Far-IR (142 μm) TE 90° elastic scattering and 0° transmission simulation for the 100 mm sphere.

spheres the same, as the Avogadro sphere size is 100 times bigger than the 1 mm Si sphere.

Tunable quantum cascade (QC) lasers and methanol lasers are available in the far-IR [58–61]. Waveguiding is also possible with far-IR fibres [62–64]. Quantum dot (QD) THz detectors can be used for detection of the far-IR signal [65]. The elastically scattered signal from the Avogadro sphere can be collected by a far-IR photodetector (PD), which is connected to a digital storage oscilloscope (DSO). The 0° transmitted signal can be collected by a far-IR power

Fig. 8. Far-IR (142 μm) TM 90° elastic scattering and 0° transmission simulation for the 100 mm sphere.

Fig. 9. Proposed far-IR 90° elastic scattering and 0° transmission spectroscopy setup for the Avogadro Project silicon sphere.

head (PH) of an optical multimeter (OMM). The far-IR PD and the far-IR PH can be manufactured from InGaAs heterostructure photoconductors [66].

8. Conclusions

Whispering gallery modes (WGMs) observed in elastic scattering from dielectric and semiconductor microspheres can provide diameter information of the sphere. Avogadro spheres are 100 times larger than the Si microspheres used in this work. Our proposal is to use elastic scattering in the far-IR, while keeping the same size parameter as the near-IR. This spectroscopic technique will provide an alternative to the optical interferometry used for diameter measurements of the Avogadro Project's Si sphere. We used elastic scattering in the near-IR for 1 mm Si spheres as a scale down model for the Avogadro sphere. Numerical simulations were performed using a GLMT algorithm for both the 1 mm Si sphere and the 100 mm Avogadro sphere. The spectral mode spacing found using the numerical simulations agreed with the calculated and measured (for the near-IR) spectral mode spacing. The

mass of the 1 mm Si microsphere was measured using a microbalance, and agreed within 3% with the mass of the microsphere estimated using the equations linking the Avogadro constant, N_A , with molar volume to atomic volume ratio. The diameter of the 1 mm Si sphere was also measured using SEM, and agreed within 0.5% with the value found using the spectral mode spacing formula Eq. (7). The GLMT simulation for the Avogadro sphere was performed using a 150 μm wavelength far-IR excitation source to keep the size parameter the same as the near-IR excitation source at 1.5 μm . Based on the numerical simulations and the scaled down near-IR spectroscopy model, the diameter measurement of the Avogadro Project Si sphere with far-IR elastic scattering is well poised as an alternate spectroscopic method for the characterization of these roundest and purest single crystal Si spheres.

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