Homogeneity Characterization of Lattice Spacing of Silicon Single Crystals by a Self-Referenced Lattice Comparator at BL-3C

The lattice spacing of a perfect silicon crystal is critical when determining the Avogadro constant by the X-ray crystal density Rietveld method [1]. In the XRCO method, the Avogadro constant, $N_A$, is derived from the mean molar mass, $M$, the density, $\rho$, and the lattice spacing of the (220) plane, $d_{220}$, of a perfect silicon crystal using the following equation:

$$N_A = \frac{M}{\rho \times \sqrt{8} \times d_{220}^2}$$

The International Avogadro Coordination (IAC) project started in 2004 has performed various measurements using a silicon crystal highly enriched with $^{28}$Si isotope with the aim of achieving an uncertainty of $2 \times 10^{-9}$ for $N_A$.

A very important precondition for achieving this goal is to have a perfect silicon crystal or at least an imperfect silicon crystal with a known lattice spacing distribution. This is because $M$, $\rho$, and $d_{220}$ are measured from samples obtained from different locations in the ingot. Impurity and defect measurements of the crystal are performed for crystal characterization.

The lattice spacing of silicon is determined by combining the lattice spacing measured using a technique that involves X-ray and optical interferometry under standard conditions (i.e., 20°C and 0 Pa). The lattice spacing is required to have an expanded uncertainty of $3 \times 10^{-9}$. Impurities measured on samples throughout an ingot are used to derive the compensations at the positions of the X-ray interferometer (XINT) and spheres.

We discovered a strain pattern whose magnitude was of the order of $10^{-6}$, which is too small to be observed by X-ray topography, in an ingot of a high-purity silicon crystal with natural isotopic abundances [2].

This strain distribution has a high spatial frequency in the ingot and thus cannot be compensated by solely determining impurity concentrations at a few sampled positions. Crystals used in X-ray interferometry are also susceptible to the very small strains caused by defects introduced during crystal processing. Damage caused during fabrication and self-weight (gravity-induced) deformation of the crystal must be checked by a sensitive method. Strain measurements were performed [3] using a Self-Referenced Lattice Comparator (SRLC) installed at BL-3C, on single crystals of silicon with natural isotopic abundances, and silicon crystals highly enriched with the $^{28}$Si isotope, which are all used to determine the Avogadro constant.

The measurement capability, i.e., the standard deviation of repeated measurements, of the system is about $3 \times 10^{-9}$ [4]. Samples from crystals with natural isotopic abundances exhibited clear pattern of striations, whereas almost no pattern was observed for crystals enriched with $^{28}$Si isotope (see Figs. 2, 3, 4, and 5).

The standard deviation of the lattice spacing of the silicon single crystal highly enriched with $^{28}$Si isotope was $4.7 \times 10^{-9}$, which enabled the lattice spacing to be determined with an expanded uncertainty of $3 \times 10^{-9}$.

The Avogadro constant $N_A$ is determined from the measurements, the lattice parameter, the mass and volume of the sphere, and the molar mass to be $N_A = 6.022\times 10^{23}$. This value differs by $16 \times 10^{-9}$ $N_A$ from the CODATA 2006 adjusted value. This value is midway between the $N_A$ values derived from Planck’s constant obtained by NIST and NPL watt-balance using the molar Planck constant $h = 3.990 \times 10^{-24}$ J s/mol [5].

REFERENCES


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