

Lattice parameter measurement for the determination of the Avogadro constant

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AN X-RAY INTERFEROMETER

(phase contrast microscopy; direct measurements of: thickness, x-ray refractive index, small lattice distortions, dispersion surfaces, electron densities; E/T)

U. Bonse and M. Hart Department of Materials Science and Engineering Cornell University Ithaca, New York (Received 22 February 1965)



This letter describes the construction and application of the first x-ray interferometer in which the principal parts of the paths of interfering beams are widely separated and in air. The instrument can



NBS

NPL

PTB

NBS SPECIAL PUBLICATION 343 (August 1971) Precision Measurement and Fundamental Constants



FIGURE 1. Partial diagram of final stages of x-ray/optical interferometer. The driven point in labeled \otimes .





Figure 2. Construction of traverse units TU used with $\Delta s\text{-}$ and $\Delta x\text{-} traverses.$



FIGURE 3. Layout of assemblies of crystals C_1 , C_2 with conjugate Fabry-Perot plates FP_1 , FP_2 respectively and their provisions for ρ - and Θ -alignments.

R.D.Deslattes - NBS - 1648.3 ± 0.1 @ 25 °C

Separate crystals

Monolitic crystal

Separate crystals



62 CONSIGLIO NAZIONALE DELLE RICERCHE ISTITUTO DI METROLOGIA «G. COLONNETTI»» R 81 DETERMINAZIONE ASSOLUTA DELLA COSTANTE RETICOLARE DEL SILICIO

R 81 Technical Report 15 Nov 1972



E. Massa was 4 years old





Combined x-ray and optical interferometry

- Laser interferometry
- X-ray interferometry
- *d*₂₂₀= (m/n) λ/2
- n number of x-ray fringes
- m number of optical fringes











Fig. 1. – Two-erystal X-ray interferometer. a) Crystallographic orientations. Isthmus I is shown before being out to allow translation of W_a with respect to $W_1 - W_a$: b) W_i beam splitter. W_a mirro W_a analyser. [110] indicates the direction of translation. D_i and D_2 detect the exit beam in the incident direction and in the diffracted direction, respectively.



Preliminary Results in X-Ray Interferometry at the Istituto di Metrologia «G. Colonnetti ».

VOL. 23, N. 9

G. BASILE, A. BERGAMIN, M. OBERTO and G. ZOSI I.M.G.C. - Strada delle Cacce 73, Torino

(ricevuto il 4 Settembre 1978)

LETTERE AL NUOVO CIMENTO





28 Ottobre 1978





FIG. 1. Schematic drawing of the combined x-ray and optical interferometer. X-INT x-ray interferometer: C1 and C2 movable and fixed crystals, O and H transmitted and diffracted beams, D_O (position-sensitive), and D_H photodetectors. O-INT polarization encoding optical interferometer: B polarizing beam splitters, M fixed mirror, P polarizers, EOM phase modulator, D_{σ} (position-sensitive), and D_{π} photodiodes.









Figure 1. Combined IMGC x-ray and optical interferometer. Crystal displacement along the x-axis is achieved by an elastic guide (not shown in the figure) on which the tip-tilt platform rests. TE and PE, target and probe electrodes of the capacitive sensor; C1 and C2, movable (analyser) and fixed crystals of the x-ray interferometer; OINT, optical interferometer; D, position-sensitive detector. The arrow indicates the common baseline of the interferometers and the capacitive sensors.













Pitch and Yaw	nrad
Roll	µrad
Х	pm
Y and Z	nm (fraction of)

Active Controls over six degrees of freedom;

Pitch (ρ), Yaw (θ) and x displacement are sensed via laser interferometry;

Roll (Ψ) and *z-y* transverse motion are sensed via capacitive transducers;







Measurement Procedure

The measurement equation is

$$a = \sqrt{8}d_{220} = \frac{\sqrt{8}m\lambda}{2n},$$

where d_{220} is the spacing of the {220} planes, $\sqrt{8}$ accounts for the different spacings of the {100} and {220} planes, and n is the number of x-ray fringes in a step of m optical fringes having period $\lambda/2$. In practice, d_{220} is determined by comparing the periods of the x-ray and optical fringes. This is done by measuring the x-ray fringe fraction at the ends of increasing steps m $\lambda/2$, where m = 1, 10, 100, 1000, and 3570. We start from $\lambda/(2d_{220}) = n/m \sim 1385,95$ and measure the fringe fractions at the step ends with an accuracy sufficient for predicting the integer number of fringes in the next step.





Experimental upgrade

In 2011, a discrepancy between the values of the Planck constant measured by counting silicon atoms and by comparing mechanical and electrical powers prompted a review of the measurement of the spacing of ²⁸Si {220} lattice planes, in order to confirm the measured value and its uncertainty or to identify errors.

All the past measurements relied on the same optical interferometer, that served us since 1994. In order to exclude systematic effects, we assembled a new one and integrated it in the apparatus. The main novelties of the upgraded system are listed here in below.



Experimental upgrade (1)



A 532 nm frequency-doubled Nd:YAG laser substituted for the previous 633 nm diode laser – stabilised by frequency-offset technique against the frequency of an He-Ne laser which, in turn, was stabilized against component a_{18} of the ¹²⁷I₂ transition 11-5 R(127).

The frequency of the Nd:YAG line is locked to the component R(56) of the 32-0 transition of the $^{127}\rm{I}_2$ molecule.

It was measured better than 10⁻¹⁰ relative uncertainty; therefore, it does not contribute to the measurement uncertainty.

The laser was also better collimated to reduce the correction for diffraction.





Experimental upgrade (2)



A new optical bench is clamped to the vacuum chamber; it collimates the laser beam, modulates the phase of the -polarized component, and delivers it to the interferometer by a pointing mirror and a window of the vacuum chamber. The delivery, collimation, modulation, and pointing systems - optical fiber, beam collimator and polarizer, phase modulator, and injection mirror – have been rebuild to conform to the new wavelength @ 532 nm.







Experimental upgrade (3)



A plate beam-splitter was manufactured *adhoc* and substitutes for the cube beamsplitter previously used to ensure that the difference of the transmitted- and reflectedlight paths is insensitive to the beam translations and rotations. Therefore, the components of the optical interferometer beam splitter, quarter-wave plates, and fixed mirror - were replaced and assembled anew.

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Experimental upgrade (4)





In order to make the interfering beams parallel, the components of the optical interferometer are cemented on a glass plate supported by three piezoelectric actuators. As shown in the picture, a noise at a frequency of about 1 mHz caused phase instabilities between the x-ray and optical fringes. A new power supply was realized, having a sub part-per-million stability over the time scales, from 1 s to 1 h, relevant to the lattice parameter measurement.

Eventually, the phase noise was reduced to the shot noise limit of the x-ray photon count.





The orthogonality between the laser beam and the analyser was only occasionally checked. This was done by observing simultaneously, via a visual autocollimator placed when necessary - outside the vacuum chamber, the analyser and laser beam through the output port of the interferometer. To gain the online control of the beam pointing, an home-made telescope picks up part of the beam delivered to the detector. In order to ensure stability, it is clamped on the same base plate as the x-ray/optical interferometer.







Experimental upgrade (6)

The residual pressure in the vacuum chamber has been reduced by an order of magnitude, below 0.04 Pa. This makes any correction for the refractive index of the residual gas in the vacuum chamber inessential and ensures the calibration of the optical interferometer with a negligible uncertainty.

New instrument FLUKE 1595A super-thermometers for temperature determination.

Contrary to our past measurement, the lattice spacing was surveyed along an horizontal line at 21 mm from the analyser base (instead of the previous 26 mm) and the correction for the self-weigh deformation was recalculated.

The Physikalische-Technische Bundesanstalt found a contamination of the surfaces of the x-ray interferometer by Cu, Fe, Zn, Pb, and Ca caused by the wet etching used by the INRIM to remove any residual stress due to surface damage after the crystal machining. The contamination was removed by cleaning the crystal in aqueous solutions of HF and $(NH_4)_2S_2O_8$.



Combined x-ray and optical interferometry











lattice-plane measurement



Red dots - outliers;

Red line – gradient of the lattice spacing, it is correlated with thermal gradient caused by the injected power of the laser beam (0.75 mW):





A – Statistics

Each measurement is the mean, after eliminating the outliers and the thermal strain, of the survey results. The uncertainty of the mean is dominated by the variations of the measured values, that are supposed to be caused by local effects of the analyser surface.

Therefore, when calculating the uncertainty, we took the residual correlation into account.

B – Laser beam wavelength

The frequency of the Nd:YAG line is locked to the component R(56) of the 32-0 transition of the ${}^{127}I_2$ molecule. It was measured to better than a 10⁻¹⁰ relative uncertainty. To eliminate the influence of the refractive index of air, the experiment is carried out in vacuo. With respect to our past measurements, the residual pressure has been reduced by a factor of ten. Since the air refractivity at the atmospheric pressure is 2.9x10⁻⁴, assuming that the pressure is in the interval from zero to 0.04 Pa with a uniform probability, the relevant correction is 0.058(33) nm/m.





C – Laser beam diffraction

The period of the interference fringes is not equal to the plane-wave wavelength.

The measurements are made from 2010 to 2014 by using laser beams differently collimated.

The correction is calculated by measuring the angular power-spectrum of the beams emerging from the interferometer by using the Fourier transforming properties of the lens.

The correction is calculated from central second moment matrix of the focal plane image recorded by videocamera.

The possible overestimation of the correction, is a wrong estimate of the center of mass of the focal plane image, which implies a correction always larger than true.





D – Laser beam alignment

The laser beam deviation from a normal incidence on the analyser was nullified with the aid of an autocollimator looking at both the analyser and beam from the interferometer output-port.

We carried out a number of d_{220} measurements where

the angles (pitch an yaw) were purposely changed along two orthogonal directions and its variations were recorded by an on-line telescope.

The telescope is mounted, inside the vacuum chamber, on the same base plate of the x-ray/optical interferometer and picks up part of the output beam. After two parabola were fitted to the measured d_{220} values, the beam direction corresponding to the maxima - hence, to a supposed permal incidence.

to a supposed normal incidence.





E – Laser beam walk

The beam walk refers to the transverse motion of the interfering beams through the optical components. It originates from different effects causing the beams to move across imperfect surfaces or wedged optics. The beam-splitter imperfection, combined with tilts of the apparatus baseplate with respect to the laser beam, caused systematic differential variations of the optical paths through the interferometer that required corrections. In the new apparatus, we made this problem harmless by using a plate beam-splitter, having a parallelism error less than 10 μ rad, and by controlling electronically the baseplate level and tilt to within 25 nm and 70 nm/m.

It is difficult to estimate if there is a residual systematic differential-walk of the interfering beams. If, over 1 mm analyser step, the systematic walk is in the [-0.1, 0.1] mm interval, a 10% of what observed, and the differential wedge-angle between the end surfaces of the separate paths through the interferometer is in the [-10, 10] µrad interval, the beam walk contribution to the uncertainty budget is 0.577 nm/m.





F – Abbe's error

The vertical offset was nullied by carrying out off-line measurements of the variations of the x-ray fringe phase in different detector pixels while the pitch component is purposely changed in order to keep the analyser displacement null. We identied the virtual pixel having a zero offset.

The horizontal offset was set to zero to within the same uncertainty by rotating the analyser about the vertical and by shifting horizontally the laser beam up to no phase variation is detected.

We discovered instabilities of the relative levelling between the x-ray source and interferometer which affect the pixel looking at the analyser point having zero vertical offset.

We estimated that the Abbe's error was nullified to within a total uncertainty 6.11 x $10^{-10} d_{220}$.

Analysis of the error budget





G – Movement direction

The analyser moves orthogonally to the front mirror of a trihedron, straightness errors are nullified to within nanometers by servoing the motion with the signals of capacitive transducers that sense the transverse displacements of the trihedron.

The misalignment between the optical and x-ray interferometers causes them to measure different components of the displacement.

$$d_{220} = \frac{m\lambda}{2n} \left[1 + \hat{\mathbf{s}} \cdot (\hat{\mathbf{n}} - \hat{\mathbf{h}}) \right]$$

s displacement,

n unit vector normal to the optical surface,h unit vector normal to the diffracting planes.

The dashed line is the locus of the **s** direction where the projection error is null.

The circles indicate the measurement uncertainties.







H – Analyser temperature

The linearity of 1595A was checked better than $10\mu\Omega$ (from 90Ω -120 Ω);

Stability of the 100Ω internal reference;

The thermometer self-heating was identified by repeating d_{220} measurements with varying currents;

Non uniqueness @ 20 °C set to 0.1 mK;

The calibration history, shows a linear drift of 14(5) $\mu\Omega$ /month;

Each d₂₂₀ was extrapolated to 20 °C, all measurement were carried out between 19,9 °C to 20.3 °C

I – Thermal strain

A linear approximation of thermal strain due to the optical power injected into the analyser by the laser beam. The filled area was found by a least-squares adjustment of the d_{220} gradients and the results of repeated d_{220} measurements carried out with varying optical powers.

The red line is the numerical calculations of the thermal strain. To correct for the thermal strain, we trusted the value given by LSA of the linear approximation, but increased its uncertainty to the one half of the gap between the minimum and maximum strain predicted by the numerical calculation.





J – Self-weight deformation

The simulation of the gravitational bending allowed the analyser to be optimally designed, the residual lattice strain to be predicted, and the contribution of the self-weight deformation to the uncertainty budget estimated. Residual mean self-weigh strain of the analyser calculated at 21 mm from the base as a function of the distance between the support points. The filled curve is the distance probability-distribution, given three contact points uniformly distributed in the (5 x 5) mm² support areas.

K – Aberrations of X-ray interferometer

Geometric aberrations contribute to the phase of the x-ray fringes (i.e. analyser thickness or focusing).

A stress exists in the crystal surfaces even if the bulk material is stress-free. If the tensile stress is 1 N/m, the measured lattice spacing is $6 \times 10^{-9} d_{220}$ smaller than the value in an unstrained crystal. We do not propose a correction and associate to a null stress an uncertainty of 0.1 N/m. This term is the relevant contribution for the uncertainty.



Experiments are under way....



TABLE I. Relative correction and uncertainty, in parts per 10^9 , of the $2014/02/12 \ d_{220}$ value.

Contribution	Correction	Uncertainty
data averaging	0.000	0.722
wavelength	-0.058	0.033
laser beam diffraction	3.978	0.597
laser beam alignment	-0.110	0.480
beam walks	0.000	0.577
Abbe's errors	0.000	0.611
movement direction	0.699	0.214
temperature	-0.500	0.497
thermal strain	-0.948	0.641
self-weigh	-0.543	0.377
aberrations	0.000	0.642
total	2.52	1.75

PRELIMINARY RESULT



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Thank you for your attention

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