γ-Ray Wavelength Standard for Atomic Scales

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The wavelength of the ^{57}Fe Mössbauer radiation is measured with a relative uncertainty of 0.19 ppm by using almost exact Bragg backscattering from a reference silicon crystal. Its value is determined as $\lambda_{\rm M} = 0.860\,254\,74(16) \times 10^{-10}$ m. The corresponding Mössbauer photon energy is $E_{\rm M} = 14\,412.497(3)$ eV. The wavelength of the ^{57}Fe Mössbauer radiation is easily reproducible with an accuracy of at least $10^{-11}\lambda_{\rm M}$ and could be used as a length standard of atomic dimensions.

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γ radiation of natural linewidth from nuclear transitions, the Mössbauer radiation, could be used as a length standard of atomic dimensions that is easily reproducible with unique accuracy. For example, the wavelength $\lambda_{\rm M} \simeq 0.86$ Å ($E_{\rm M} = 14.4$ keV) of the Mössbauer radiation of ^{57}Fe nuclei is reproducible with an accuracy of at least $10^{-11}\lambda_{\rm M}$ without any special precautions regarding temperature, pressure, and chemical composition of the environment in which the nuclei are placed. Such a high reproducibility is due to the weak coupling of nuclei to atomic shells. The hyperfine interaction shifts the photon energy at most by 0.5 μeV. The uncertainty of the photon energy due to the finite lifetime $\tau_0 = 141$ ns of the 14.4 keV nuclear excited state is even less: $\hbar/\tau_0 = 4.8$ neV. Such a wavelength standard would be important for metrology, crystallography, x-ray optics, x-ray spectroscopy, nuclear spectroscopy, etc. However, the wavelength, or, respectively, the energy of the ^{57}Fe Mössbauer radiation is acknowledged to be known with a relative accuracy of only 10 ppm: $E_{\rm M} = 14\,413.00(15)$ eV [1].

Thirty-five years ago Bearden considered the possibility of introducing a γ-ray wavelength standard, attempting to measure the wavelength of 14.4 keV Mössbauer radiation of ^{57}Fe nuclei with a relative accuracy of 1 ppm [2]. His conclusion was the following: to make the Mössbauer standard experimentally feasible the brightness of the 200 mCi source used would have to be increased by a factor of 100 [2].

Since that time the progress in growing perfect silicon single crystals and the progress in the determination of the silicon lattice constant in terms of an optical wavelength standard was considerable [3–5]. The lattice constant of silicon became the most accurate length standard in use. The 1998 CODATA recommended value of the lattice constant of silicon [6] is given with a relative uncertainty of 0.029 ppm as $5.431\,020\,88(16) \times 10^{-10}$ m.

However, lattice constants are sensitive to the ambient temperature and pressure, as well as to the chemical purity and the perfection of crystals. To reproduce the standard value with utmost precision the absolute values of these parameters have to be very accurately controlled. An alternative length standard easily reproducible with high accuracy is highly desirable.

The brightness of radioactive sources is limited by self-absorption. This limitation can be overcome if one uses synchrotron radiation as a source. Mössbauer photons are filtered from the synchrotron radiation spectrum by coherent nuclear resonant scattering. Since the pilot experiment [7] in 1984 the rate of the 14.4 keV Mössbauer photons available has increased from 0.1 Hz to $\simeq 10^4$ Hz within a solid angle of $20 \times 20\ \mu{\rm rad}^2$ at modern synchrotron radiation facilities of the third generation. The synchrotron-based sources of Mössbauer photons are already $\simeq 10^6$ times brighter than the 200 mCi radioactive source used by Bearden and will be much brighter with the coming x-ray free-electron laser facilities. Thus the conditions are favorable to return to the problem of a Mössbauer wavelength standard.

In the present paper we introduce an experimental technique which enables a comparison of crystal lattice constants with x- or γ-ray wavelengths with a relative accuracy of $\simeq$0.2 ppm. In particular, results of recent experiments are presented in which the ratio of the crystal lattice constant of silicon a to the wavelength $\lambda_{\rm M}$ of the ^{57}Fe Mössbauer radiation is measured with an uncertainty of 0.19 ppm; i.e., with the same uncertainty the value of $\lambda_{\rm M}$ can be determined using the so far more precisely known value of a.

The idea underlying the experimental technique exploits the fact that Bragg's law for exact backscattering

$$\lambda = 2d_{hkl} \tag{1}$$

establishes a direct relation between the interplanar distance $d_{hkl} = a/\sqrt{h^2 + k^2 + l^2}$ of the reflecting atomic planes (hkl) in silicon and the wavelength λ of the reflected radiation. This property is used to calibrate in units of a the instrument for measuring wavelengths, the λ-meter. By selecting the indices h, k, l the calibration can be performed in any region of interest where wavelengths are then measured in units of a.

Figure 1 shows the scheme of the experimental setup. As reference a silicon single crystal (Si) was used, which lattice constant $a = 5.431\,020\,30(36) \times 10^{-10}$ m has been calibrated against the silicon standard crystal [5] with a relative uncertainty of 0.07 ppm at 22.5 °C in vacuum. To reproduce this value of a in the present experiment the crystal is kept in an evacuated thermostat at 22.500 °C (295.650 K) with an accuracy of $\leq$5 mK and a stability of $\leq$1 mK [8]. The thermostat is installed on a four-circle goniometer and can be oriented to allow backreflections of x rays having passed the λ-meter (λ). We use backreflections with $2d_{hkl}$ close to λ_M: (11 5 3), (12 4 0), and (9 9 1). Any pair of these reflections allows one to calibrate the λ-meter.

If the wavelength of the radiation coincides with λ_M, it excites coherently the ^{57}Fe nuclei in an α-Fe foil (F). The foil is 6 μm thick, enriched to 95% in ^{57}Fe. The excited nuclei emit Mössbauer radiation in forward direction, with an average delay of $\tau_0 = 141$ ns. This delay allows one to discriminate easily Mössbauer quanta from the prompt radiation pulse.

The detector, an avalanche photodiode, with a time resolution of $\simeq$1 ns [9], consists of a silicon wafer of 100 μm thickness which is semitransparent for 14 keV x rays. It detects $\simeq$50% of the radiation pulse after the λ-meter and the backscattered radiation pulse after its 40 ns time of flight to and back from the reference silicon crystal [10,11].

The λ-meter in our experiment is a silicon channel-cut crystal. Its temperature is kept at 303.8 K with a stability of 2 mK. The symmetric Bragg reflection (777) is used. The wavelength λ at the exit of the λ-meter varies as a function of the angle of incidence θ of the beam to the (777) atomic planes. Variation of the wavelength is performed by rotation of the channel-cut crystal mounted on a high-angular resolution rotation stage (KOHZU KTG-15) with a step of 25 nrad. The rotation angle ψ is measured with a Heidenhain ROD800C angle encoder and AWE1024 interpolation electronics, which has a resolution of 0.175 μrad. Figure 2 shows the detected backreflected radiation as well as the Mössbauer radiation on the ψ scale.

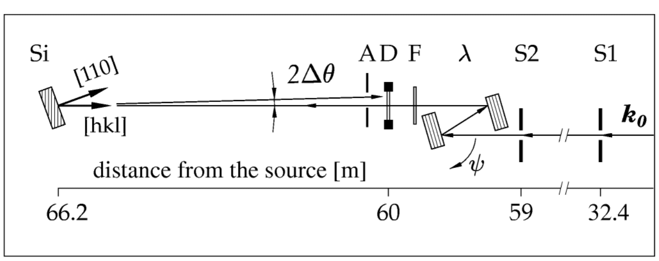

FIG. 1. The scheme of the experiment: the radiation after a high-heat-load premonochromator (not shown) passes through the vertical slits $S1$ and $S2$, λ: λ-meter; F: ^{57}Fe foil used as a source of Mössbauer radiation of high brightness; D: semitransparent avalanche photodiode with 1 ns time resolution; A: 4 mm aperture, Si: reference silicon single crystal with (110) surface in an evacuated thermostat on a four-circle goniometer. The distances from the APS source are given.

The intrinsic relative width of the (777) reflection on the wavelength scale for x rays with $\lambda \simeq 0.86$ Å is $\Delta\lambda/\lambda \simeq 3.5 \times 10^{-7}$, thus allowing one to measure wavelengths with a relative accuracy better than 10^{-7}. To achieve this precision two conditions have to be fulfilled: (i) the divergence of the incident beam in the plane of dispersion should be less than the angular width of the (777) reflection—1.2 μrad, and (ii) the direction of the incident beam should be kept constant *independent* of the wavelength of the x rays. To ensure this, a system of two slits $S1$ and $S2$ at a large mutual distance is used.

In the following the theoretical background and sources of possible uncertainties of the measurements are discussed. Weakly absorbing crystals, like silicon, reflect x rays incident at the angle θ to atomic planes within a wavelength region centered at λ_c. λ_c is determined by

$$\frac{\lambda_c}{2d}\left[\frac{\lambda_c}{2d} - \sin\theta\right] = -\delta(\lambda_c). \quad (2)$$

Here

$$\delta(\lambda) = \frac{r_e\lambda^2}{2\pi V}N[Z + f'(\lambda)] \quad (3)$$

is the real part of the correction to the complex refractive index $n(\lambda) = 1 - \delta(\lambda) - i\beta(\lambda)$ of x rays in silicon; r_e is the classical electron radius, V is the unit-cell volume, and N, Z, and f' are the number of atoms in the unit cell, the atomic number, and the real anomalous correction to the forward scattering amplitude, respectively. A kind of the relation (2) is derived in the dynamical theory of Bragg diffraction [12]. In the form of Eq. (2) it is valid at any angle θ including normal incidence [10,11].

We measure the variation of the rotation angle ψ rather than of θ. They are identical only if the rotation axis is perfectly perpendicular to the incident beam $\boldsymbol{k}_0$

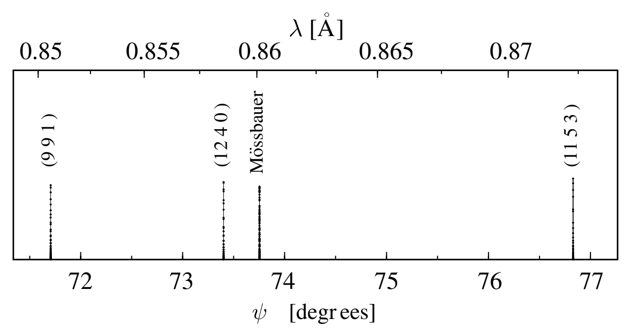

FIG. 2. Backreflected and Mössbauer radiation in the detector vs the rotation angle ψ of the λ-meter. The wavelength scale is calculated according to (4).

and parallel to the (777) reflecting planes. If ξ and η are the angles of deviation from perfect alignment, then the relation between θ and ψ reads as follows: $\sin\theta = \sin\psi \cos\eta \cos\xi + \sin\eta \sin\xi$. Combining this expression with Eqs. (2) and (3) one obtains the relation between the rotation angle ψ and the wavelength λ_c:

$$\sin\psi = \frac{\lambda_c}{2d^*} - \zeta \,. \tag{4}$$

Here $\zeta = \tan\eta \tan\xi$ allows for a nonperfect alignment of the λ-meter. d^* is another instrumental parameter of the λ-meter given by $d^* = d \cos\eta \cos\xi/(1 + w)$ with $w = (2r_e d^2 N/\pi V)[Z + f'(\lambda_c)]$. We assume it to be constant in the spectral range $\lambda_M - 0.01$ Å $\le \lambda_c \le \lambda_M + 0.01$ Å investigated in our experiment. This is justified as $f'(\lambda_c)$ varies in this range at most by ± 0.003 about its average value of 0.119 [13], and thus w varies by less than 10^{-8} compared to the leading term Z. The instrumental parameter d^* is determined in the experiment.

If the radiation is reflected backwards from the (hkl) planes in the silicon crystal then, according to Eq. (2), its spectral region of total reflection is centered at

$$\lambda_c = 2d_{hkl}\left[1 - \frac{1}{2}\Delta\theta^2 - \delta(\lambda_c)\right]. \tag{5}$$

The relative spectral widths of the backreflections are $\simeq 3 \times 10^{-7}$, i.e., similar to the spectral width of the λ-meter. Equation (5) is a more general condition for Bragg backscattering than Eq. (1). It includes not only the deviation from normal incidence $\Delta\theta = \pi/2 - \theta \ll 1$, but also the effect of refraction. The refractive index $1 - \delta$ in silicon is known with a relative accuracy of 2×10^{-9} [13]. The value of δ extrapolated for $\lambda_M = 0.860\,25$ Å from the data of Ref. [13] is $\delta = 2.340 \times 10^{-6}$ and varies by 5×10^{-8} in the region of interest.

Because of the $\Delta\theta^2$ dependence in (5) a deviation from normal incidence by $\Delta\theta \simeq 0.3$ mrad changes the wavelength by only $4.5 \times 10^{-8}\lambda_c$. This weak dependence is favorable, since at exact Bragg backscattering in silicon, multiple-beam scattering effects take place and more complex equations in place of (5) should be used [14,15]. To avoid this we performed the measurements 0.3 mrad off normal incidence where the multiple-beam effects vanish [11,15] and apply Eq. (5) with the precisely known small $\Delta\theta^2$ correction. The $\Delta\theta = 0.3$ mrad offset is adjusted with the help of a 4 mm aperture (A) installed behind the detector.

The rotation angle ψ_{hkl} of the λ-meter at which it transmits x rays matching the backscattering reflection (hkl) is calculated by using Eqs. (5) and (4). Equation (4) allows one to determine the rotation angle ψ_M at which the λ-meter transmits the Mössbauer radiation. For the difference $\psi_{hkl} - \psi_M = \Delta\psi_{hkl}$, which is measured in the experiment, we obtain

$$\Delta\psi_{hkl} = \arcsin\left[\frac{2\tilde{a}x}{\sqrt{h^2 + k^2 + l^2}} - \zeta\right] - \arcsin[x - \zeta]. \tag{6}$$

Here $\tilde{a} = a[1 - \Delta\theta^2/2 - \delta(\lambda_M)]/\lambda_M$, $x = \lambda_M/2d^*$, and ζ are free parameters. The λ-meter was adjusted so that $\zeta \le 10^{-4}$ ($|\xi|, |\eta| \le 10$ mrad). It was ascertained by the numerical analysis of Eq. (6) that ζ in this case can be taken as zero without changing the result of $\tilde{a}$ by more than $10^{-8}\tilde{a}$. Thus for the determination of $\tilde{a}$ and x, $\Delta\psi_{hkl}$ has to be measured for two independent reflections (hkl).

The experiments were performed at two synchrotron radiation facilities.

At the wiggler beam line BW4 at HASYLAB (DESY, Hamburg) the vertical source size was $\simeq 0.8$ mm. A compromise between high count rate and small beam divergence allowed us to use only relatively large vertical apertures $\simeq 100$ μm of the slits $S1$ and $S2$, separated by a distance of 19.9 m. The vertical divergence of the beam was 12 ± 2 μrad, derived from the width of the angular dependence of transmission of Mössbauer photons through the λ-meter. The count rate of Mössbauer photons was 5 Hz. The $\Delta\psi_{hkl}$ values were measured for the backreflections (12 4 0) and (9 9 1) at different temperatures of the reference crystal. Results of the evaluation of $\tilde{a}$ are given in Fig. 3 (full circles). They fit well

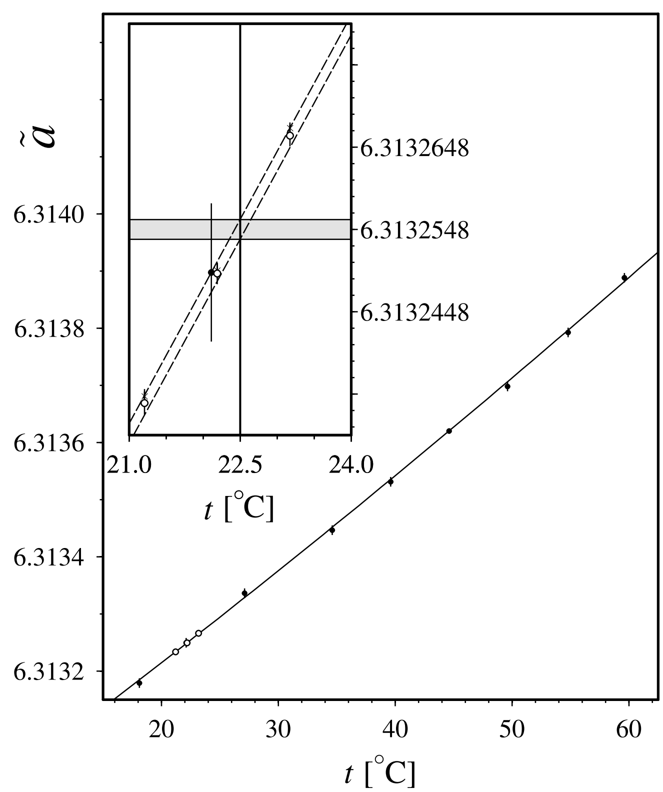

FIG. 3. The ratio $\tilde{a}$ of the lattice constant a of the reference silicon crystal to the wavelength λ_M of the ^{57}Fe Mössbauer radiation in silicon measured at different temperatures of the crystal. Full (●) and open (○) circles represent the results of evaluations of the HASYLAB and APS measurements, respectively, by using the pair of (12 4 0) and (9 9 1) reflections. The crosses (×) show the results of evaluations of these measurements by using three reflections: (11 5 3), (12 4 0), and (9 9 1). The solid line is the temperature dependence of a [16]. The dashed lines in the inset limit the significant error corridor for the APS data.

to the solid line, given by the thermal expansion of silicon [16]. The value of $\tilde{a}$ at $t = 22.500\,^\circ\mathrm{C}$ is determined as $\tilde{a} = 6.313\,254\,5(85)$ (an average of four measurements). The 1.3 ppm uncertainty is owing to the relatively large divergence of the beam and low count rate.

To improve this result, another experiment was carried out at the undulator beam line 3-ID at the Advanced Photon Source (APS) (Argonne). The source size was 120 μm (FWHM). A system of two roller-blade slits [17] $S1$ and $S2$ was used at a distance of $\simeq$26.6 m to collimate the beam. The smallest beam divergence of 4.5 μrad was achieved with vertical apertures of the slits of $S1 = 40$ μm and $S2 = 75$ μm, respectively. The smaller divergence and the higher count rate ($\simeq$100 Hz for Mössbauer photons) in this experiment allowed us to achieve a higher accuracy for $\tilde{a}$. Open circles in Fig. 3 represent results of evaluations of the APS measurements by using the pair of (12 4 0) and (9 9 1) backreflections. The crosses show the average for three independent pairs of backreflections: (11 5 3), (12 4 0), and (9 9 1). The dashed lines in the inset limit the significant error corridor for the APS data. By interpolation we obtain at $t = 22.500(5)\,^\circ\mathrm{C}$:

$$\tilde{a} = 6.313\,254\,8 \pm 0.000\,001\,2\,. \qquad (7)$$

This value agrees with the value determined from the HASYLAB measurements and is 7 times more accurate. By using the value of a for the reference crystal, we obtain

$$\lambda_{\mathrm{M}} = (86.025\,474 \pm 0.000\,016) \times 10^{-12}\ \mathrm{m}\,, \qquad (8)$$

for the wavelength of the ^{57}Fe Mössbauer radiation, and

$$E_{\mathrm{M}} = (14\,412.497 \pm 0.003)\ \mathrm{eV}\,, \qquad (9)$$

for its energy $E_{\mathrm{M}} = hc/\lambda_{\mathrm{M}}$ with Planck's constant $h = 4.135\,667\,27(16) \times 10^{-15}$ eV s [6]. It is noteworthy that our value $E_{\mathrm{M}} = 14\,412.497(3)$ eV agrees much better with the value 14 412.47(29) eV used in the previous seventh edition of *Table of Isotopes* [18] derived from the 35 year old measurements of Bearden [2] than with 14 413.00(15) eV of the newest edition [1].

The accuracy can be improved by at least an order of magnitude if the beam divergence is reduced to a sub-μrad level. The future x-ray free-electron laser sources with much better emittance will certainly allow this.

In conclusion, we have measured the ratio of the silicon lattice constant a to the wavelength λ_{M} of the ^{57}Fe Mössbauer radiation with a relative accuracy of 1.9×10^{-7}. This allows us to determine the absolute value λ_{M} with the same relative uncertainty. The wavelength of the ^{57}Fe Mössbauer radiation is easily reproducible with an accuracy of at least 10^{-11} and could be used as an alternative atomic scale length standard. These measurements are extended in another experiment [19] to γ radiation of ^{151}Eu (21.5 keV), ^{119}Sn (23.9 keV), and ^{161}Dy (25.6 keV). The backscattering technique introduced here can be alternatively used to measure unknown crystal lattice parameters in units of a known x- or γ-ray wavelength, e.g., in units of λ_{M}, with an uncertainty of $\simeq$0.1 ppm. High collimation of the synchrotron x rays combined with well-known fixed wavelength of the radiation generated at the nuclear resonance may enable more accurate determination of thermal expansion coefficients, as well as pressure dependence of lattice constants.

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