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Accurate determination of the voltage of a transmission electron microscope (TEM) by ⟨0 1 2⟩ CBED–HOLZ analyses using GaAs crystal

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Abstract. Convergent beam electron diffraction (CBED) is a powerful technique to estimate lattice distortion and lattice strain in crystals. The positions of the higher-order Laue-zone (HOLZ) lines in the transmitted disc of CBED patterns are very sensitive to the lattice parameter, and can therefore be used to estimate changes in the lattice parameter. This offers the possibility to calculate lattice misfit and lattice strain. The positions of the HOLZ lines depend not only on the lattice parameter, but also on the operating voltage of the microscope. It is essential to know the actual voltage of the microscope. In the present work, (1 0 0) GaAs crystal has been used as a standard. Cross-sectional TEM specimens were prepared by argon ion beam thinning technique using a liquid nitrogen cold stage. ⟨0 1 2⟩ on-zone CBED technique has been used to estimate the actual voltage of the transmission electron microscope (Philips EM430T TEM), when the voltage was set at 250 kV. CBED–HOLZ simulation and analyses have been done, using JEMS software, to correlate with the experimental data. The methodologies adopted for estimating the actual voltage of TEM are discussed in this paper. The studies have also been cross-checked using ⟨0 1 2⟩ and ⟨2 3 3⟩ zone axes using (1 0 0) silicon standard. The techniques established are found to be suitable for TEMs operating at a setting voltage of about 250 kV. For the TEM studies, a regular double-tilt specimen holder is required in order to be able to get to the desired zone axes. When the experiments were repeated using a cryogenic double-tilt holder, an improvement in the sharpness of HOLZ lines was observed. Wherever possible, the use of the cryogenic double-tilt holder is recommended. Care must, however, be taken to ensure that effects such as lattice parameter changes (due to temperature changes), phase transformations etc can be properly accounted for.

Keywords. TEM; ⟨0 1 2⟩ CBED–HOLZ analysis of GaAs; estimation of voltage.

1. Introduction

Convergent beam electron diffraction (CBED) is one of the well known techniques for estimation of lattice strain and lattice distortion in semiconductor thin films (Maher *et al* 1987; LeGoues *et al* 1992). The positions of higher-order Laue-zone (HOLZ) lines in the (000) disc of the CBED patterns are sensitive to lattice parameters as well as the operating voltage of the microscope. Lattice distortions involving a change of crystal symmetry are also reflected in the symmetry of the CBED–HOLZ patterns (Maher *et al* 1987). Due to good spatial resolution in the TEM, where probing volumes could be reduced to nanoscale, this technique has an advantage in estimating local lattice parameters, lattice misfit and lattice strain (Toda *et al* 2000). However, unlike normal imaging and diffraction which are somewhat insensitive to small changes (~ few kV) in the operating voltage of the microscope, positions of HOLZ lines are strongly dependent on the voltage (Spence and Zuo 1992) and change significantly even for changes as small as 100 V. Therefore, knowledge of the actual voltage of TEM is an *a priori* requirement for lattice parameter measurements using CBED technique. In order to determine the actual voltage, it is, therefore, essential to use samples with known lattice parameter(s). Conventionally, silicon has been used for this purpose (Zuo 1992). However, for III–V films having GaAs as substrate, it is convenient to use GaAs crystal to calibrate the microscope voltage because specimen need not be changed with silicon during the experiment, and changes in lattice parameter between the epi-layers and the substrate can be easily monitored. This is possible because the lattice parameter of substrate GaAs is known to great accuracy and is reported (Jain *et al* 1997) to be 0·565325 nm. In the present work, ⟨0 1 2⟩ CBED–HOLZ analysis techniques have been used on (1 0 0) GaAs crystal for estimation of the actual voltage of TEM. Further, the results have been verified using (1 0 0) silicon crystal, in ⟨0 1 2⟩ and ⟨2 3 3⟩ zone axes orientation. The methodologies developed for the estimation of the voltage of TEM are presented in this paper.

The CBED experiments were carried out using a cryogenic double-tilt holder. A marked improvement in the sharpness of HOLZ lines was observed. However, the lattice had undergone contraction due to cooling. Hence,

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for the analysis of HOLZ lines, it is essential to consider the thermal properties such as thermal expansion coefficient (a) of the sample.

2. Experimental

For the present studies, single crystal wafers of (1 0 0) polished and epi-ready semi-insulating GaAs (from American Crystal) and (1 0 0) *n*-type silicon (from PB-Technik AG, Switzerland) were used. Semiconductor crystals such as GaAs and Si, usually cleave along ⟨1 1 0⟩ directions. If the wafers are assumed to have (1 0 0) orientation (figure 1a), the allowed in-plane ⟨1 1 0⟩ directions are [0 1 1] and [0 1 – 1], and their counterparts, viz. [0 – 1 – 1] and [0 – 1 1]. In this paper, distinction is not made between [u v w] and [– u – v – w] directions, as it is not relevant. Cross-sectional TEM specimens of the samples were prepared from the cleaved samples (figure 1b) and subsequent argon ion beam thinning using Gatan-make Duo Mill (Model 600) with liquid nitrogen cold stage (Sridhara Rao *et al* 1999). The specimens were loaded in a regular double-tilt specimen holder and observed in a Philips EM430T TEM operated in the vicinity of 250 kV. The actual settings were dependent on the sample and the techniques (§ 3·1 – § 3·4) used. Some of the experiments were also performed using a cryogenic double-tilt holder (Gatan-make, model 613–050C) for examining the nature of HOLZ lines at cryogenic temperatures.

As the cross-sectional TEM specimens were prepared from the cleaved samples, the beam direction near zero tilt is always along either [0 1 1] or [0 1 – 1] depending on the actual nature of the longer edge (figure 1b), i.e. the sample is in a [0 1 1] or [0 1 – 1] zone axis orientation. The samples need to be tilted to a ⟨0 1 2⟩ zone axis for the CBED studies. As the HOLZ patterns from any of the ⟨0 1 2⟩ are equivalent, the notation ⟨0 1 2⟩ will be used in this paper henceforth. However, it must be noted that sample geometry and microscope tilt conditions permit only two of the different ⟨0 1 2⟩ orientations (and their [– u – v – w] counterparts) for a given sample. These two zone axes are at about ± 18° away from the zero tilt condition. For instance, if we take the beam direction at zero tilt to be parallel to [0 1 1], the accessible ⟨0 1 2⟩ zone axes are [0 1 2] and [0 2 1].

The voltage of the TEM was varied in steps of 25 V, using the auxiliary continuous H.T. control, for observing changes with accelerating voltage in the positions of HOLZ lines. The manufacturer of the TEM guarantees (Service Manual 1987) a voltage stability of better than 10 ppm, which amounts to 2·5 V at an operating voltage of 250 kV. As the smallest step considered (25 V) is much larger than this value, issues relating to the stability of the voltage have not been considered. The second condenser (C2) was operated at a spot size 3 to obtain optimal convergence of the electron beam at the sample. The CBED patterns were recorded at a camera length of 2900 mm. For observing the HOLZ-lines at a higher magni-

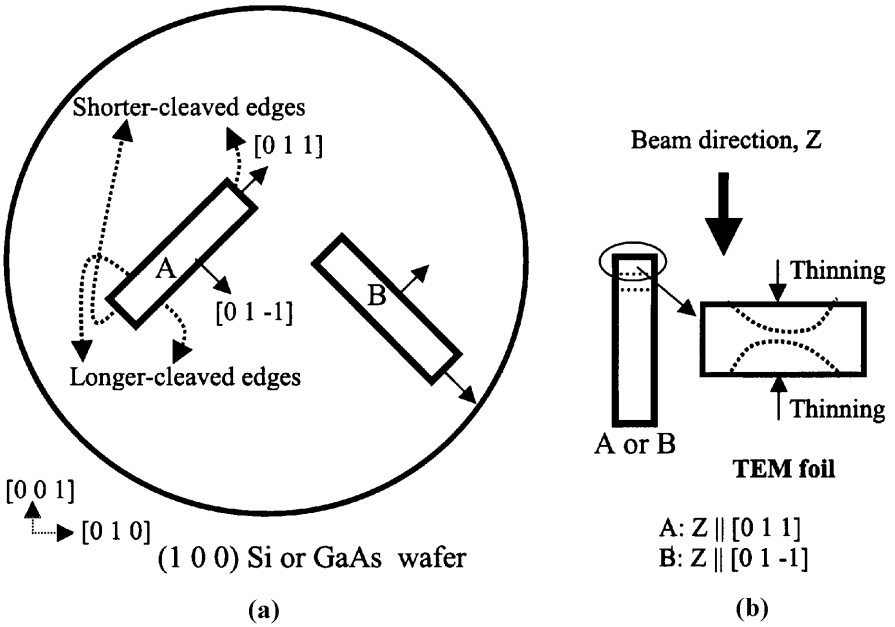

Figure 1. (**a**) Schematic representation of the cleaved samples of (1 0 0) Si or GaAs. The cleavage happens along ⟨1 1 0⟩ directions. The only two possible orientations, A and B, of the cleaved specimens are shown and (**b**) the possible beam directions are shown for A and B.

fication, an on-line CCD TV rate camera system attached to a TV monitor (Gatan-make, Model 622SC) was used.

The CBED–HOLZ patterns of GaAs (a = 0·5653 nm, ZnS structure) and Si (a = 0·5431 nm, diamond cubic structure) were simulated using JEMS software (Stadelmann 2001). The version of the software used permits the value of the lattice parameter to be chosen up to the fourth decimal place in nm. Hence, the lattice parameters of the crystals were rounded-off to the fourth decimal for the simulations. The parameters used for the simulation are listed in table 1. Simulations were performed under dynamical conditions, which are emulated in the software through the use of a 'HOLZ shift'.

3. Methodologies and discussion

A cursory observation of figures 2 and 3 shows that a good match exists between simulation and experiment, in positions of HOLZ lines in the transmitted discs of GaAs and Si, respectively. The suitability of JEMS software and the conditions employed have further been examined thoroughly for GaAs crystal. Only qualitative assessment has been performed for silicon crystal.

For the determination of voltage, a number of techniques involving different aspects of HOLZ line intersections have been developed. The features/aspects considered are: (a) multiple-intersections (§ 3·1), (b) all-HOLZ line collapse (§ 3·2), (c) size of triangle CIK (§ 3·3), and (d) collapse of triangle CIK to a point (§ 3·4). However, all these techniques have an inherent limitation caused by the finite width of HOLZ lines. Examination of CBED patterns obtained from silicon suggests that it is possible to discern differences in the HOLZ pattern when the difference in kV is at least 0·1 kV (figures 4a–d). As a result, an error bar of ± 0·1 kV has been used for the voltages determined from the experiments (§ 3·1 – § 3·4). The

Table 1. Parameters used for the JEMS simulation of CBED–HOLZ patterns for GaAs and silicon.

	Crystal	
Parameter	GaAs	Silicon
Zone-axis	[0 1 2]	[0 1 2], [2 3 3]
Laue zone no.	1	1
Acceptance angle	~ 50 mrad	~ 50 mrad
Camera length	~ 3500 mm	~ 3500 mrad
Half-convergence angle	~ 4 mrad	~ 4 mrad
Voltage	249–250 kV in steps of 0·1 kV	250 kV
Lattice parameter†	0·5650–0·5660 nm	0·5431 nm
HOLZ threshold††	~ 100 mV	~ 100 mrad
Deviation††	~ 0·15 nm^{-1}	~ 0·15 nm^{-1}

†Rounded-off to the fourth decimal in nm; ††different parameters such as HOLZ threshold, deviation etc were chosen so as to obtain a qualitative match with the experimental patterns.
Note: Dynamical effects were incorporated by enabling 'HOLZ-shift'

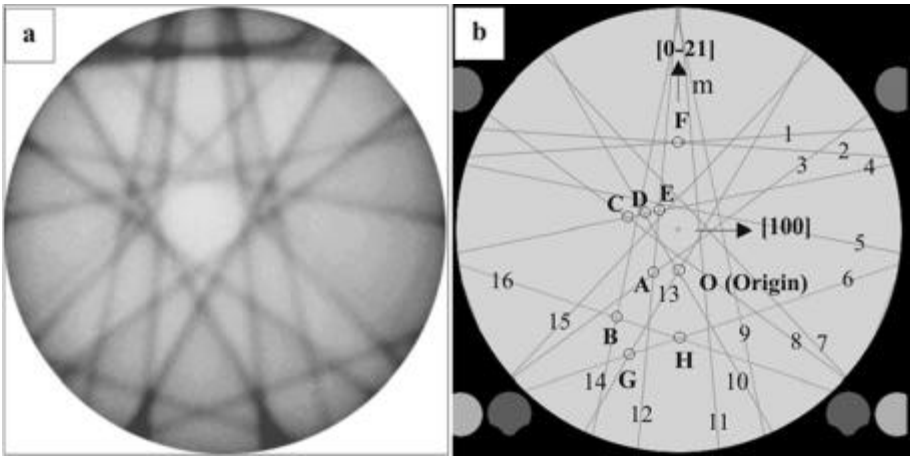

Figure 2. HOLZ lines in the transmitted disc for on zone [0 1 2] CBED pattern in GaAs: **a**. experimental and **b**. simulation at 250 kV for a = 0·5653 nm. All the sixteen HOLZ lines have been identified. The indices of HOLZ lines are shown in table 2.

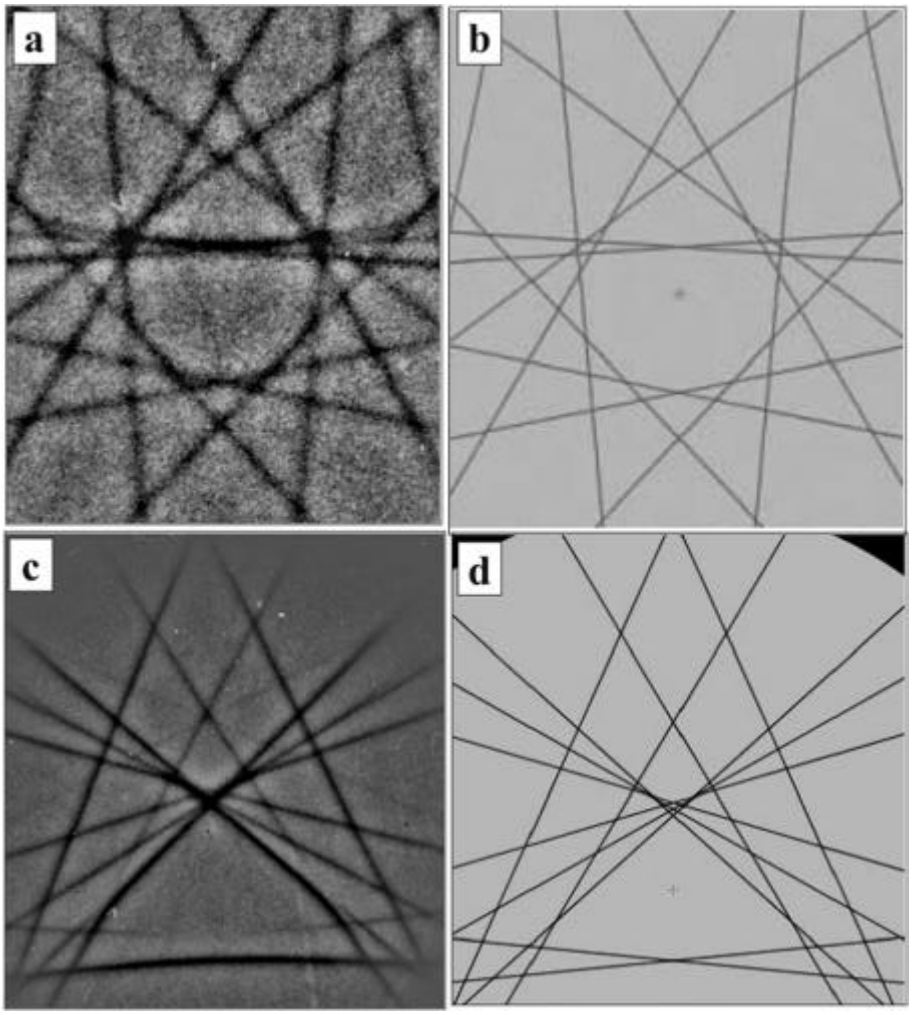

Figure 3. Experimental (**a, c**) and simulated (**b, d**) transmitted discs of silicon in [0 1 2] and [2 3 3] zone axes, respectively at 250 kV. Good agreement between the experimental and simulated patterns can be seen.

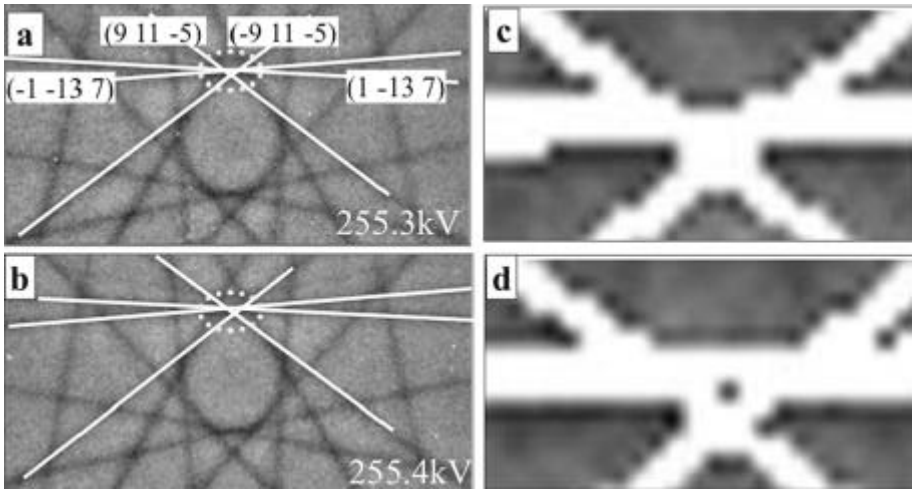

Figure 4. Experimental transmitted discs of silicon in [0 1 2] zone-axis taken at **a**. 255·3 kV and **b**. 255·4 kV, showing the intersections between four HOLZ lines: (– 1 –13 7), (1 – 13 7), (9 11 – 5) and (– 9 11 – 5). **c**. and **d**. show the details (digital enlargement) near the region of the three intersections (marked with a dotted circle) of **a**. and **c**. respectively.

sensitivity can, however, be improved by performing CBED experiments at cryogenic temperatures since the reduced thermal diffuse scattering of the atoms results in much sharper HOLZ lines (§ 3·7), leading to discernible changes in HOLZ patterns even for voltage changes as small as 25 V.

3.1 *Positions of intersections of HOLZ lines in (000) disc*

In the simulated [0 1 2] pattern of GaAs (figure 2b), sixteen HOLZ lines have been identified (table 2) and the intersections of the HOLZ lines (3, 12), (14, 16), (4, 8), (10, 4), (4, 12), (1, 2), (6, 13) and (6, 16) have been selected for analyses and named as A–H. The relative positions of these eight intersections with respect to an arbitrarily selected origin (O) were analysed for different lattice parameters at a given voltage and at different voltages for a given lattice parameter. It was determined that the distance between the points F and H (FH) is invariant for lattice parameters in the range 0·5651–0·5656 nm and for voltages in the range 249–250 kV. Therefore, all the distances have been normalized with FH. The normalized distances, A–G, from O, are plotted as a function of lattice parameter, at a constant voltage of 250 kV (figure 5). A least square fit was used to obtain a linear function describing the changes with lattice parameter. The least square fits are given in table 3. It can be seen that the fits are good with the value of the coefficient of determination (R^2) above 0·96 in all the cases.

The lengths of intersections OA–OG obtained experimentally (voltage setting of microscope: 250 kV, regular double-tilt holder) from GaAs (figure 2a) were normalized with the length FH. The lattice parameter of GaAs was calculated by fitting this data to the linear equations for the normalized lengths obtained from simulation (table 3), and found to be 0·5653 nm. This is in good agreement with the value of 0·565325 nm reported in the literature (Jain *et al* 1997). This agreement between simulation performed at 250 kV and experiment when the microscope voltage was set at 250 kV, suggests that the actual voltage of the microscope was indeed 250 ± 0·1 kV. This clearly demonstrates that this technique can be used for estimation of voltage of the microscope.

Table 2. HOLZ lines observed in the (0 0 0) disc of ⟨0 1 2⟩ CBED of GaAs. The corresponding disc is shown in figure **2b**.

HOLZ-line	Index	HOLZ-line	Index
1	(1, – 13, 7)	9	(15, 3, – 1)
2	(– 1, – 13, 7)	10	(13, 7, – 3)
3	(– 9, 11, – 5)	11	(– 15, – 1, 1)
4	(3, – 13, 7)	12	(15, – 1, 1)
5	(– 3, – 13, 7)	13	(– 13, 7, – 3)
6	(5, – 13, 7)	14	(– 15, 3, – 1)
7	(– 11, – 9, 5)	15	(11, – 9, 5)
8	(9, 11, – 5)	16	(– 5, – 13, 7)

3.2 *All-HOLZ line collapse*

It has been determined from simulations that a majority of HOLZ lines (10 out of 16) present in figure 2a collapse to a single point at the centre of the transmitted disc at a specific voltage for a given lattice parameter. The six HOLZ lines: 14, 9, 2, 1, 16 and 6 do not collapse to a point for this condition. For experimental convenience, this voltage has been denoted as all-line collapse voltage (ACV). Hence, the determination of ACV, for a crystal with known lattice parameter such as GaAs, can be used to determine the voltage of TEM.

The simulation and experiments have been conducted on GaAs. The simulation showed that the ACV decreases with increase of lattice parameter. The simulated transmitted disc depicting the all-HOLZ line collapse for a = 0·5653 nm, which represents the experimental lattice parameter of GaAs, is shown in figure 6a. This corresponds to an ACV of 245·5 kV. The ACV values for a =

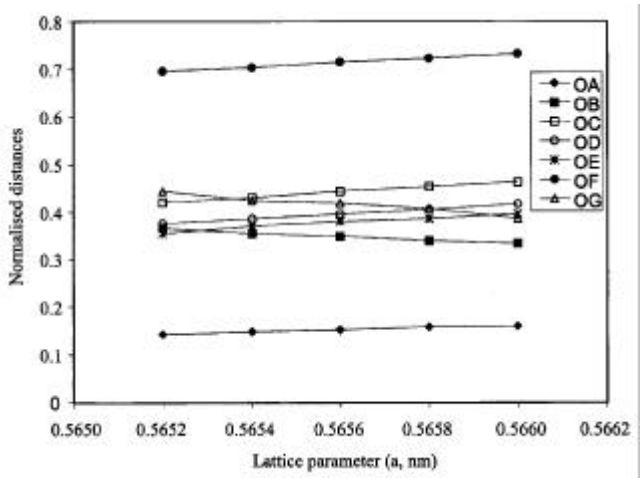

Figure 5. The variation of lengths OA–OG, normalized by the length FH, as a function of lattice parameter at 250 kV. The corresponding linear least squares fit equations are given in table 3.

Table 3. Linear fitting functions, obtained by least squares method, for the distances OA–OG, normalized with the distance FH, as a function of lattice constant for GaAs. The coefficient of determination (R^2) is also given.

Normalized distances*	Linear fits to the normalized distances	Coefficient of determination (R^2)†
OA	22·225x – 12·418	0·9907
OB	– 39·516x + 22·698	0·9870
OC	53·803x – 29·989	0·9926
OD	49·967x – 27·866	0·9994
OE	47·620x – 26·557	0·9635
OF	46·220x – 25·428	0·9963
OG	– 66·682x + 38·131	0·9693

*The distances have been normalized by length FH; †rounded-off to the fourth decimal.

0·5652, 0·5654 and 0·5655 nm were found to be 245·6, 245·4 and 245·3 kV, respectively. This can be represented in the form of a linear relationship, y (y is ACV, in kV) = – 1000x (x is lattice parameter, in nm) + 810·8 with $R^2 = 1$. From this relationship, the ACV for GaAs crystal with a lattice parameter of 0·565325 nm (Jain *et al* 1997) has been found to be 245·475 kV. ACV, as determined from experiment (figure 6b), was found to be 245·5 ± 0·1 kV. This is in excellent agreement with simulation.

3.3 *Size of the triangle CIK (denoted as ΔCIK)*

A close observation of the intersections formed by the HOLZ lines—(8, 4), (8, 14) and (4, 14), (experimental and simulation studies, figure 7), shows that it is not a point but a triangle (ΔCIK). Careful examination of the simulated patterns shows that the nature of ΔCIK is voltage-dependent for a given lattice parameter. As the voltage is changed, the triangle collapses to a point and again starts expanding into a triangle but now with a different relative orientation of the intersections forming the vertices C, I and K. Figure 8 shows this voltage dependency in the 249–250 kV range, for a = 0·5653 nm. It is evident that the ΔCIK collapses to a point between 249·5 and 249·6 kV, and the relative orientations of the vertices are different on either side of this collapse. This suggests that, if the lattice parameter 'a' is known, the size of ΔCIK in conjunction with the relative orientation of the three vertices could be used to estimate the voltage of microscope.

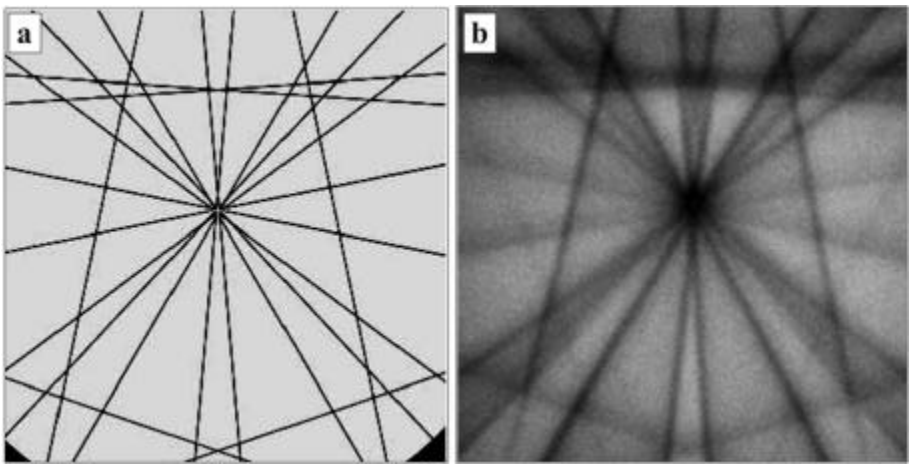

Figure 6. Collapse of all HOLZ lines in the transmitted disc in [0 1 2] zone axis for GaAs. **a**. simulation, 245·5 kV and **b**. experiment, 245·5 ± 0·1 kV.

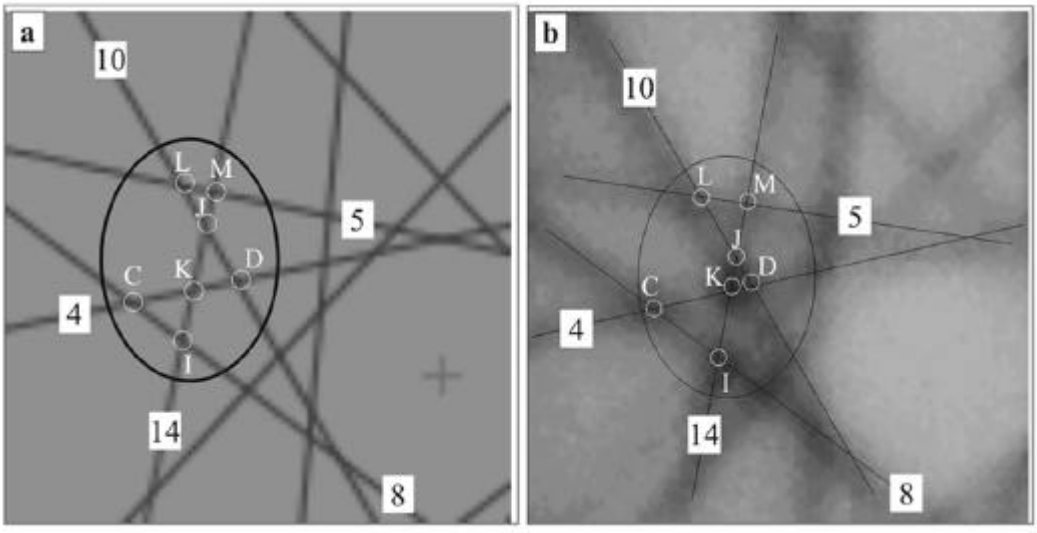

Figure 7. The triangle formed by the intersections of HOLZ lines 4, 14 and 8 (table 2) in GaAs crystal: **a**. simulation, 250 kV and **b**. experiment, 250 ± 0·1 kV.

However, it should be noted that a similar effect would be seen if the lattice parameter is varied at a constant voltage. An example is shown in figure 9, for four different lattice parameters (a = 0·5645, 0·5650, 0·5655 and 0·5660 nm) at the same voltage of 250 kV. Thus, it is emphasized that these techniques can be used to estimate voltage only if the lattice parameter is known with certainty.

The lengths of the sides of ΔCIK were measured on the simulated patterns. To minimize the errors, measurements have been carried out from O to the points C, I and K. Then CI (OI–OC), IK (OK–OI) and CK (OK–OC) have been calculated for different lattice parameters (0·5652 nm, 0·5653 nm and 0·5654 nm) and voltages (249–250 kV, in steps of 0·1 kV). The data was plotted assuming a linear dependency (table 4) for each lattice parameter. The representative plots for a = 0·5653 nm are shown in figures 10a and b (only magnitudes are considered). As can be seen, the lines have both negative and positive slopes, for the collapsing and expanding regimes, respectively. For a given experimental pattern, once the lengths of the three sides of the triangle are measured, the correct regime needs to be chosen based on the relative orientation of the three vertices. For example, the nature of ΔCIK, from the experimental CBED pattern (figure 7) does not match with the simulated data at 249 kV (figure 8) because the relative orientation of the three vertices of the triangle is different. For this experimental pattern, the expanding regime (voltage > 249·6 kV) has to be chosen. Once the regime is chosen correctly, the voltage is calculated from the linear equations for the respective lengths. The average of the three voltages (corresponding to CI, IK and CK) obtained can be taken to be the actual operating voltage of the microscope.

The analysis of the data suggests that the actual voltage of microscope will be 249·475 kV for 0·5653 nm when it is operated at 250 kV. This value has been found to be 249·59 kV and 249·415 kV for a = 0·5652 and 0·5654 nm, respectively. In linear form, the linear relationship bet-

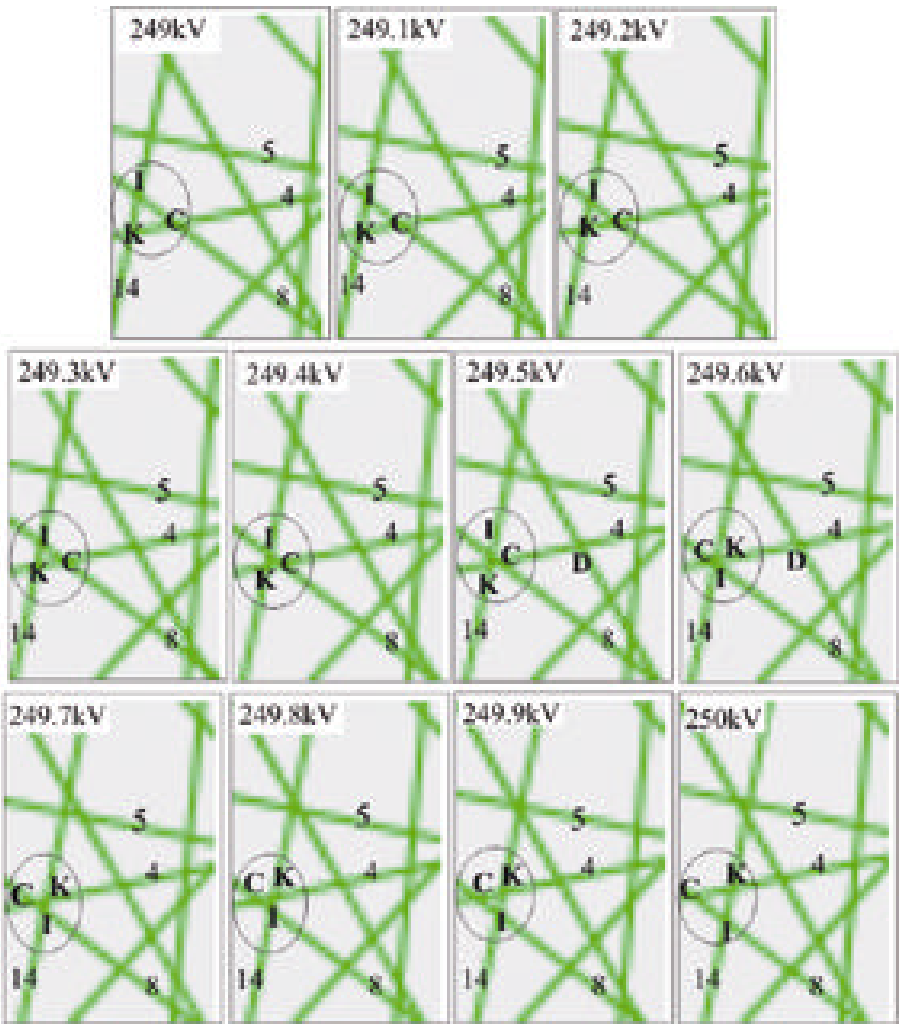

Figure 8. Effect of voltage on the positions of intersections of HOLZ lines in the transmitted disc of [0 1 2] zone axis for GaAs with a = 0·5653 nm. For easy visualization, the triangle CIK is encircled.

ween voltage and lattice parameter is y (voltage, in kV) = – 883·66x (x is lattice parameter, in nm) + 749·03 with R^2 = 0·965. Using this linear relationship, for GaAs crystal (a = 0·565325 nm, Jain *et al* 1997) the voltage of the microscope when it was set at 250 kV has been estimated to be 249·47 kV. This estimated value does not agree with the value estimated by the two previous methods (§ 3·1 and § 3·2), and differs by about 400 V.

This disagreement is likely caused mainly by two factors: (a) errors in length measurement, and (b) the assumption of linear dependency of the lengths with voltage. In spite of obtaining R^2 values in excess of 0·95, it can be seen that significant deviations are present between the actual data points and the linear fit (figures 10a and b). It might be possible to more accurately estimate the voltage of microscope by considering non-linear (such as quadratic or cubic) fits to the actual data. However, this technique, being more involved and less accurate than the method of all-line collapse, was not explored further.

3.4 *Triangle collapsing voltage*

As mentioned in § 3·3, simulation shows that the triangle CIK (figure 7) collapses to a point at a particular voltage for a given lattice parameter. For example, simulation suggests that the collapse occurs somewhere between 249·5 kV and 249·6 kV for a = 0·5653 nm (figure 8). The experiments (figure 11) on GaAs show the triangle collapsing voltage to be 249·7 ± 0·1 kV.

3.5 *Comparison of the techniques*

The multiple-intersection technique (§ 3·1) has the unique advantage of not requiring the continuous H.T. control, which is only an auxiliary attachment on many microscopes. However, this technique has a disadvantage of requiring identification of various intersections and various length measurements followed by normalization, involving considerable off-line analysis methods and statistical calculations.

The techniques involving all-HOLZ line collapse (§ 3·2) and triangle collapse (§ 3·4) offer the advantage of only on-line observations. Both of these require the continuous H.T. control. However, the triangle collapsing method, besides requiring the identification of the intersections C, K, and I, also tends to be less accurate because it involves only three intersections as compared to ten for the all-line intersection technique. Thus, it appears that the all-HOLZ line technique is the most efficient and accurate for determination of the microscope voltage.

3.6 *CBED studies of silicon*

For silicon crystal, similar detailed studies, as discussed in § 3·1–§ 3·4, have not been performed. However, the positions of the HOLZ lines were qualitatively checked in the simulation and experimental patterns, using a = 0·5431 nm (Basile *et al* 1994) in ⟨0 1 2⟩ and ⟨2 3 3⟩ zone axes (figure 3). As explained in § 2, ⟨2 3 3⟩ has been used to denote two of the different ⟨2 3 3⟩ orientations (and their [– u – v – w] counterparts) which are at about ~ 25° to the ⟨0 1 1⟩ zone axis. It appears that a good match exists in the positions of HOLZ lines.

As the all-HOLZ line collapsing technique is more convenient among other methods, it has been used for the studies. From simulation and experimental study, the ACV has been found to be 261·7 kV (figure 12a) and 261·7 ± 0·1 kV (figure 12b), respectively.

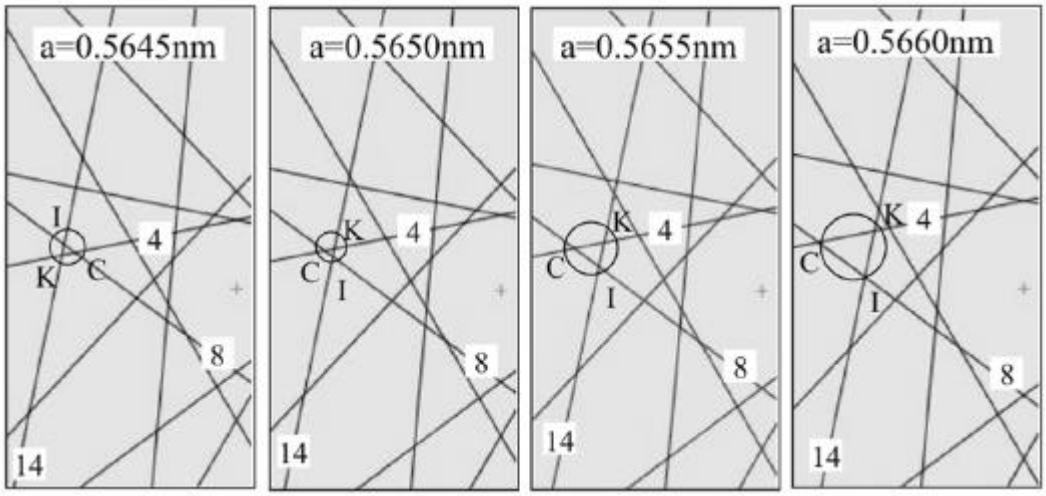

Figure 9. Effect of lattice parameter on the positions of intersections of HOLZ lines in the transmitted disc of [0 1 2] zone axis for GaAs at 250 kV. For easy visualization, the triangle CIK is shown encircled.

Table 4. Length of the sides (CI, IK and CK) of the triangle CIK as a function of operating voltage for three different lattice parameters in GaAs, obtained from simulation. The data set for each lattice parameter has been obtained by varying the operating voltage in steps of 0·1 kV in the range 249–250 kV. Linear fitting functions, obtained by least squares method, for the lengths of the sides are also given.

Lattice parameter, a (nm)	Length of the sides of ΔCIK, y (mm)*	$y = mx + c$, x is voltage in kV	
		m (slope, mm/kV)	c (mm)
		249–249·7 kV	
	IK	– 24·556	6131·7
	CI	– 19·641	4904·4
0·5652 nm	CK	– 19·328	4826·2
		249·7–250 kV	
	IK	31·622	– 7896·2
	CI	24·992	– 6240·6
	CK	24·742	– 6178·1
		249–249·6 kV	
	IK	– 22·987	5738·4
	CI	– 18·570	4635·7
0·5653 nm	CK	– 18·225	4549·7
		249·6–250 kV	
	IK	26·705	– 6664·9
	CI	21·444	– 5351·9
	CK	21·350	– 5328·4
		249–249·7 kV	
	IK	– 20·381	5085·3
	CI	– 20·505	5116·1
0·5654 nm	CK	– 25·776	6431·4
		249·7–250 kV	
	IK	25·192	– 6286·0
	CI	20·111	– 5018·1
	CK	19·890	– 4963·0

*All measurements made from simulated patterns obtained under identical conditions. Length units are consistent.

3.7 *CBED studies using cryogenic double-tilt holder*

In the experiments with a regular double-tilt holder (§ 3·1–§ 3·6), an error bar ± 0·1 kV has been used in the experimental findings. This is due to the broadness of the HOLZ-lines at room temperature. Cooling of the specimen can lower the thermal diffuse scattering of the atoms and therefore, improve the sharpness of HOLZ lines. Hence, the changes in the positions of intersections of HOLZ lines may be detectable even if the changes in voltage are smaller than 0·1 kV, which is an error bar.

The ⟨0 1 2⟩ CBED experiments were performed on GaAs and silicon crystals, using cryogenic double-tilt holder. The studies showed that the cooling of specimens improved the sharpness of HOLZ lines, as shown comparatively at 21°C and at – 165°C for GaAs (figure 13), near all-HOLZ line collapsing condition. As a result, the changes in the positions of HOLZ lines were detectable for much smaller changes in voltage (~ 0·025 kV, as shown in figure 14 for silicon), as compared to the detectable limit of 0·1 kV at room temperature (figure 4).

However, the cooling of specimen has changed the lattice parameter, as dictated by the coefficient of thermal

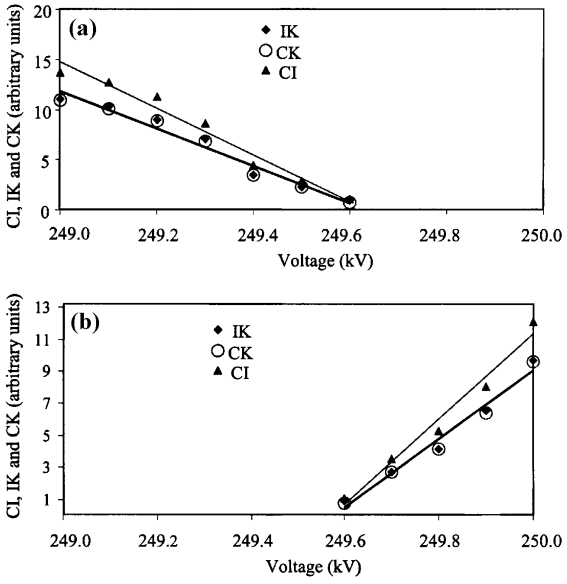

Figure 10. Simulation studies showing the size of CI, IK and KC as a function of voltage for GaAs crystal with a = 0·5653 nm, (**a**) 249·0–249·6 kV and (**b**) 249·6–250·0 kV. A least squares fit has been used to join the points (table 4).

expansion. For example, for GaAs crystal, the ΔCIK becomes smaller in size (expansion regime, voltage > 249·6 kV, figure 8, § 3·3) when the specimen temperature was lowered from room temperature (21°C) (figure 15a) to –79°C (figure 15b), and its nature was flipped (contraction regime, voltage < 249·6 kV, figure 8, § 3·3) when the specimen was further cooled to –159°C (figure 15c), indicating a lowering of lattice parameter with the cooling. This observation is in agreement with the literature (Harper 1970). By assuming a cubic contraction of the lattice due to cooling and using the relationship

$$\alpha = (d\,a/a) \times (1/d\,T),$$

where α = thermal expansion coefficient (/°C), a = lattice parameter, $d\,a$ = change in lattice parameter and $d\,T$ = change in temperature, $d\,a$ at –79°C was calculated by using $\alpha_{-79°C} \sim 4{\cdot}5 \times 10^{-6}$/°C (Harper 1970). As a result, the lattice parameter of GaAs at – 79°C would become 0·56507 nm, if a = 0·565325 nm, at room temperature (21°C). This value

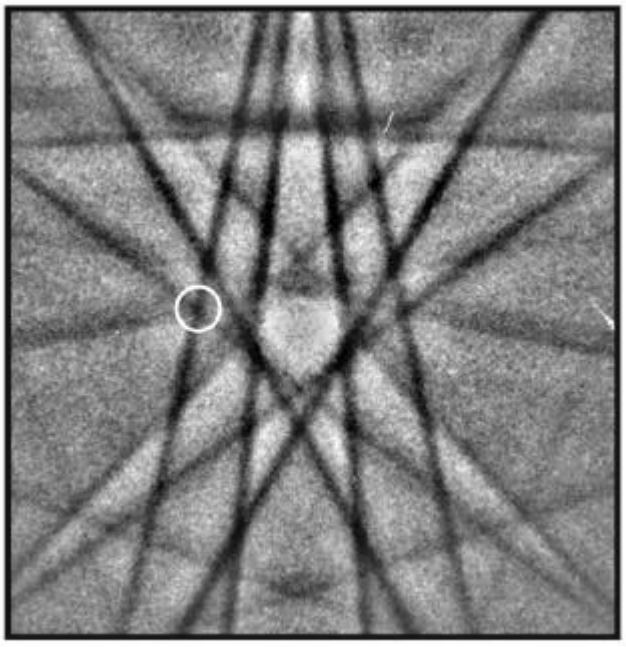

Figure 11. Experimental HOLZ pattern in the transmitted disc of [0 1 2] zone axis for GaAs crystal (a = 0·5653 nm) showing the collapse of triangle CIK to a point (encircled) at 249·7 ± 0·1 kV.

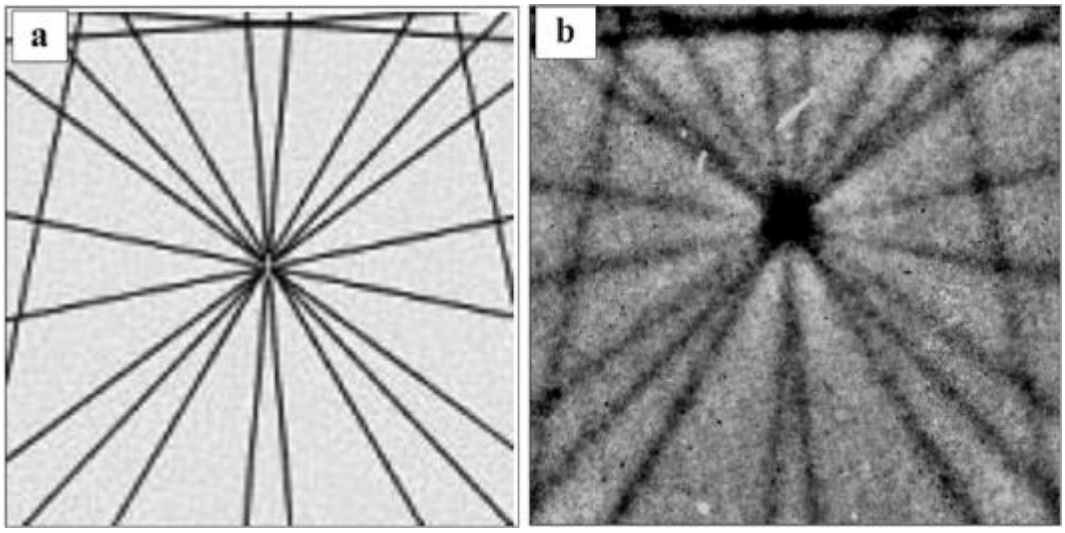

Figure 12. Collapse of all HOLZ lines in the transmitted disc in [0 1 2] zone axis for silicon. (**a**) simulation, 261·7 kV and (**b**) experiment, 261·7 ± 0·1 kV.

was qualitatively in good agreement with the findings from the CBED experimental and simulated data, as shown in the insets of figures 15a and b.

At –159°C, the observations were found to be interesting. Both experimental and simulation studies (figure 15c) suggest that the lattice parameter would become ~ 0·5648 nm. These findings will be valid if a remains invariant ($4{\cdot}5 \times 10^{-6}$/°C). In such a case, the lattice parameter would become ~ 0·5648 nm. However, $a_{-159°C} \sim 3 \times 10^{-6}$/°C (Harper 1970). Using this value, the change in the lattice parameter ~ – 0·00030 nm. As a result, the new lattice parameter should become 0·56502 nm, a contradiction with our observations. Hence, based on the CBED observations, it is suggested that the value of a needs to be verified at such low temperatures.

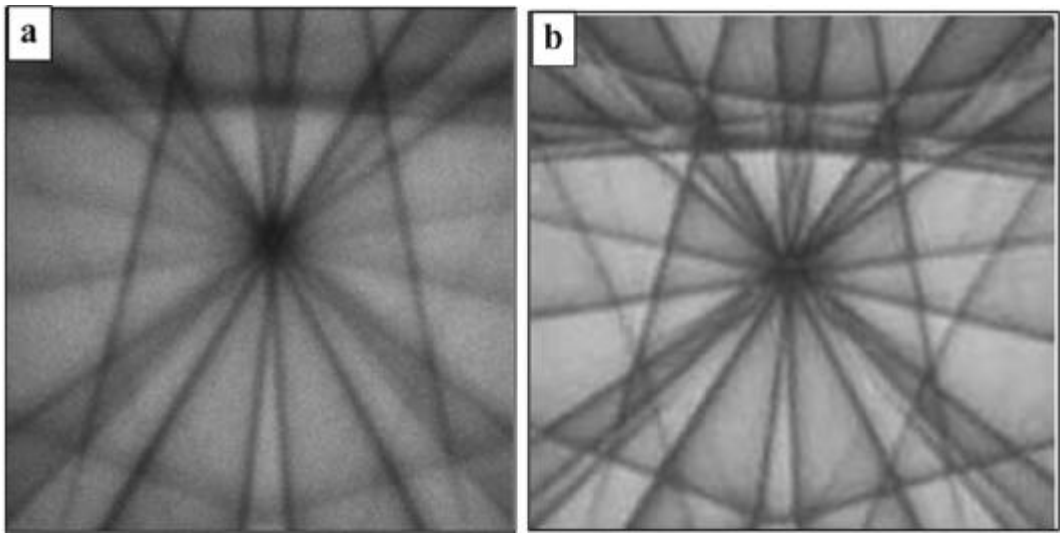

Figure 13. Improvement in the sharpness of HOLZ lines due to specimen cooling. The observations were made at **a**. 21°C and **b**. –165°C.

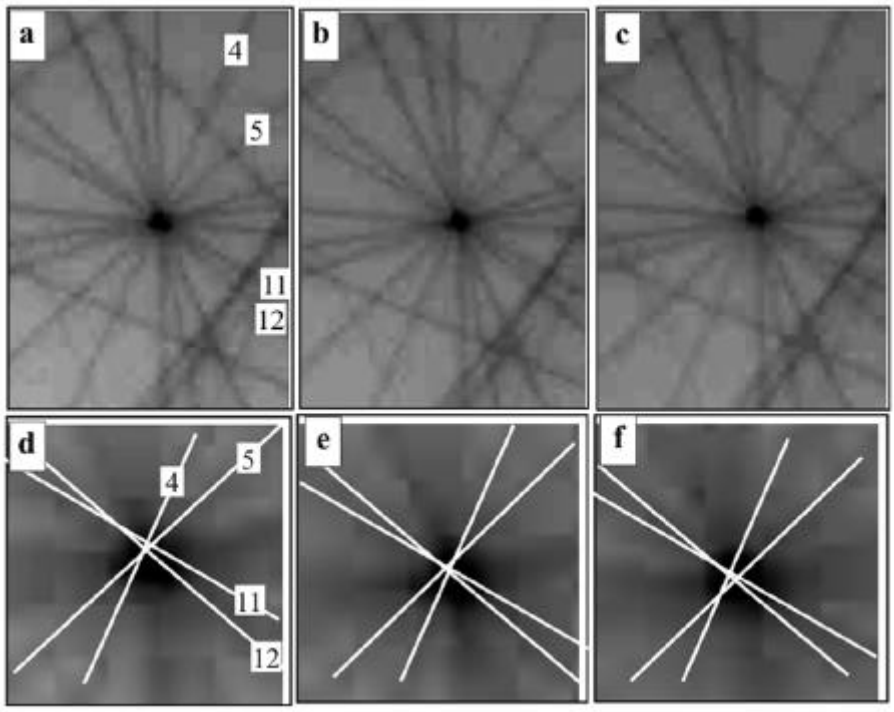

Figure 14. (**a**–**c**) Positions of HOLZ lines in the transmitted disc in [0 1 2] zone axis for silicon, at – 165°C, at 261·950 kV, 261·975 kV and 262·000 kV, respectively. (**d**–**f**) show the positions of the intersections formed by the HOLZ lines 4, 5, 11 and 12 (indices are given in table 2) after about 25 × digital enlargement of (**a**–**c**), respectively.

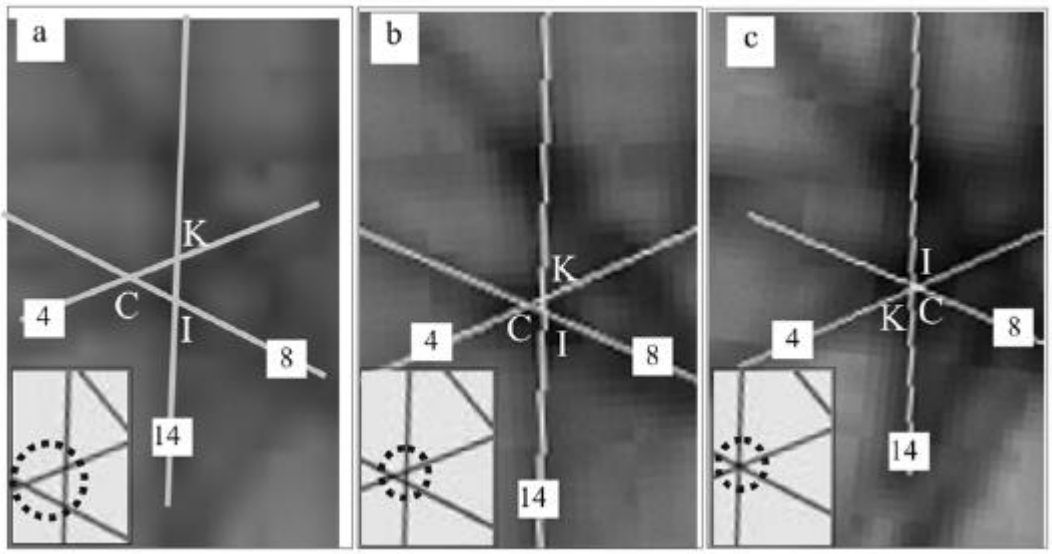

Figure 15. Positions of HOLZ lines in the transmitted disc in [0 1 2] zone axis for GaAs, showing size of triangle CIK at **a**. 21°C (room temperature), **b**. – 79°C and **c**. – 159°C, respectively. The simulation studies performed on GaAs crystal with a = 0·5653 nm, 0·5650 nm and 0·5648 nm (shown in the inset of **a**–**c**) matches qualitatively with the experimental observations, suggesting lattice contraction.

The experiments with the cooling holder suggests that although the sharpness of HOLZ lines improves with specimen cooling, it is essential to consider the changes in the lattice parameters due to thermal expansion coefficient and therefore, their effects on the positions of HOLZ lines.

4. Conclusions

The accelerating voltage of a TEM has been estimated using on-zone ⟨0 1 2⟩ CBED-HOLZ techniques and a high purity GaAs sample with known lattice parameter. Of the four different techniques utilized, the all-HOLZ line collapse method appears to be the most promising for voltage determination, as it is both quick and accurate. The results from the GaAs sample have been further verified using a silicon sample.

The studies have shown that the microscope voltage can be estimated to an accuracy of ± 100 V in studies carried out at room temperature. The accuracy can be improved to ± 25 V by cooling the specimen to cryogenic temperatures (~ – 150°C). However, the change in lattice parameters of the sample due to thermal contraction/ expansion effects needs to be accurately accounted for in such cases. It should also be ensured that there are no other effects, such as phase transformations, due to the reduction in sample temperature.

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