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Hexagonal Close-Packed (HCP) Crystallography, Diffraction, and Indexing Conventions

by Wendy L Sarney

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Hexagonal Close-Packed (HCP) Crystallography, Diffraction, and Indexing Conventions

Wendy L Sarney

Sensors and Electron Devices Directorate, DEVCOM Army Research Laboratory

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14. ABSTRACT This report details the crystallography and diffraction pattern indexing of hexagonal close-packed (HCP) crystals compared with the more familiar cubic close-packed crystals. HCP films are frequently grown and characterized at the US Army Combat Capabilities Development Command Army Research Laboratory, in particular alloys of gallium nitride, indium nitride, aluminum nitride, and silicon carbide. We discuss the Miller–Bravais indexing system, which relies on four indices (HKIL), as opposed to the more familiar three-index (hkl) Miller system. Both systems appear in the literature. Visualizing the HCP system and understanding its indexed features are much less intuitive than it is for the cubic system. The purpose of this note is to 1) assist researchers in their understanding of the literature related to HCP alloys and allow them to correctly describe these alloys' characteristics in their own work, and 2) serve as a reference for correctly interpreting transmission electron microscopy images and diffraction patterns.					
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1. Introduction

US Army Combat Capabilities Development Command (DEVCOM) Army Research Laboratory (ARL) researchers grow crystalline films in several molecular beam epitaxy chambers, as well as by metalorganic chemical vapor deposition, atomic layer deposition, and other evaporation techniques. There has been extensive work over the years in III-V semiconductor films, which crystallize in the face-centered cubic (FCC) lattice, usually with the zinc-blende structure. Figure 1a shows the conventional FCC unit cell. A prior technical report details the crystallography and indexing conventions for transmission electron microscopy (TEM) diffraction patterns and images of FCC materials.¹

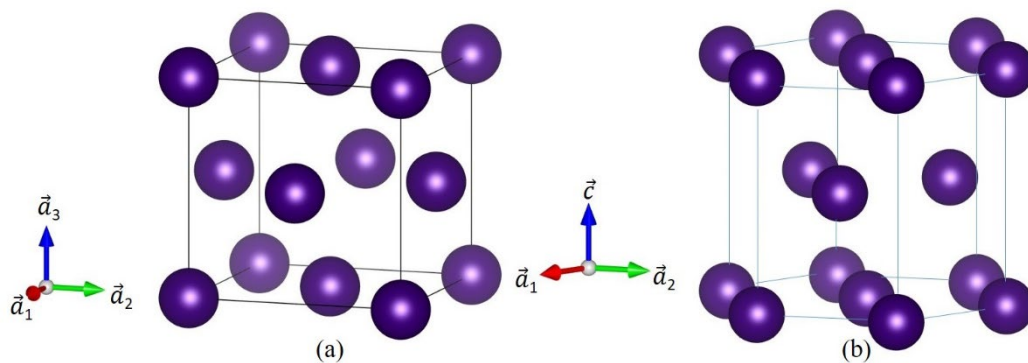


Fig. 1 Conventional unit cell and axes of an a) FCC crystal and b) hexagonal close-packed (HCP) crystal

The DEVCOM Army Research Laboratory also studies the III-Nitride (III-N) family of semiconductors, such as gallium nitride (GaN) and aluminum nitride, and their associated ternaries. Some of the alloys are polymorphic, but commonly crystallize in the wurtzite phase, which is a hexagonal close-packed (HCP) structure. Figure 1b shows the conventional HCP unit cell. We generated the crystal schematics for this report using the free VESTA web application.²

The crystallography and indexing conventions of the HCP system are less intuitive than for the FCC system, making the interpretation of TEM images and diffraction patterns more difficult. This note explains the HCP system and its nomenclature, to aid in the interpretation of TEM data collected at ARL and/or published in the literature.

1.1 Cubic System Basics

We begin by briefly reviewing the cubic system, so that we can clearly demonstrate the HCP system's differences. Figure 1a and Eq. 1 describe the three orthogonal vectors as the defining axes of the conventional cubic unit cell.

$$\vec{a}_1 = a\hat{x} \quad \vec{a}_2 = a\hat{y} \quad \vec{a}_3 = a\hat{z} . \quad (1)$$

Sometimes books and papers describe crystals by their primitive cell, which is the smallest repeat unit that leaves no “empty space” and contains only one lattice point. Figure 2 shows the primitive cell of the FCC lattice a) inscribed within the conventional unit cell and b) on its own. The primitive lattice vectors for the FCC lattice (Eq. 2) describe a rhombohedral primitive cell, where a is the lattice constant as

$$\vec{a}_1 = \frac{a}{2}(\hat{x} + \hat{y}) \quad \vec{a}_2 = \frac{a}{2}(\hat{y} + \hat{z}) \quad \vec{a}_3 = \frac{a}{2}(\hat{x} + \hat{z}) = \pi r^2 . \quad (2)$$

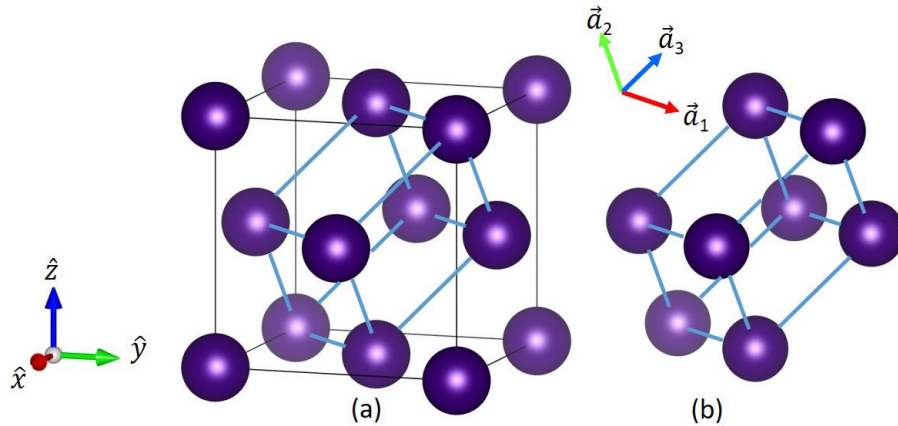


Fig. 2 Primitive cell for the FCC lattice a) inscribed inside its conventional unit cell and b) on its own

Most researchers prefer to use the conventional unit cell, because it is more descriptive and it is easier to visualize the symmetry.

We apply the conventions of the three-dimensional Cartesian coordinate system to index the atomic positions, crystal directions, and planes. It is simple to use basic geometry and trigonometry to find distances between atomic positions and crystal planes, and define directions for dislocations and interfaces. Planes and directions that are crystallographically related have common but differently permuted indices. As shown in Fig. 3, the (100), (010), and (001) planes are all symmetric and equivalent, and are referred to collectively as the $\{100\}$ family of planes. Likewise, $\langle 100 \rangle$ family of directions includes the symmetrically equivalent [100], [010], and [001] directions. Another benefit of this coordinate and indexing scheme

is that a crystal plane has the same indices as its normal. For example, the (001) plane, which is our usual substrate growth front, is perpendicular to the [001] direction. Similarly, the $(\bar{1}10)$ plane is perpendicular to the $[\bar{1}10]$ direction. The $\langle 110 \rangle$ directions are the most common TEM zone axes for cross-sectional FCC materials. Figure 4 shows the orthogonal $(\bar{1}10)$ and $(1\bar{1}0)$ planes and corresponding directions. As explained in a previous report,³ it is good practice to prepare TEM samples with two pieces from orthogonal planes.

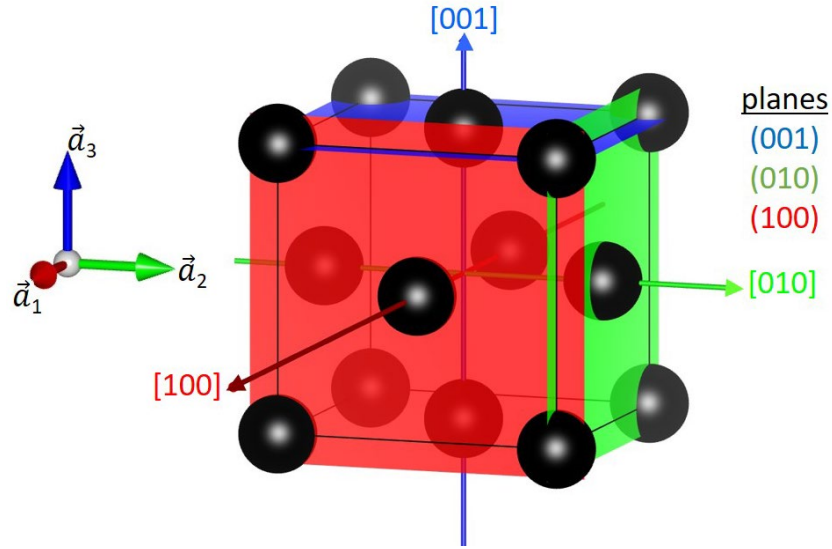


Fig. 3 $\{100\}$ family of planes and $\langle 100 \rangle$ family of directions

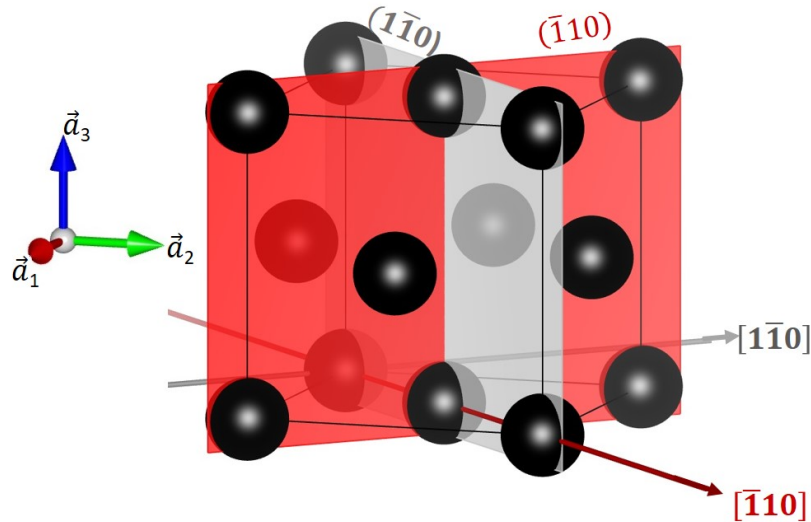


Fig. 4 Crystallographically equivalent and orthogonal $(1\bar{1}0)$ and $(\bar{1}10)$ planes and corresponding directions

1.2 HCP System Basics

Some elemental materials with the HCP structure are beryllium, magnesium, titanium, cobalt, zinc, and cadmium. HCP semiconductor alloys include the III-N family and also zinc sulfide, cadmium sulfide, and silicon carbide (SiC). Some compounds are polymorphic, meaning that they can crystallize in more than one phase depending on the growth conditions. Examples include GaN, which can crystallize in the 3C (cubic) structure or the 2H (HCP) structure, and SiC, which has over 150 known polytypes.

Figure 5 shows the primitive HCP cell inscribed within the conventional unit cell. The conventional cell includes more than the minimum number of atoms needed to represent a primitive cell, but it conveniently shows the six-fold symmetry of the lattice. The HCP structure's basis consists of two identical atoms, one at the origin and the other at $(\frac{2}{3}, \frac{1}{3}, \frac{1}{2})$ when using three-index notation. Unlike the cubic system, the axes used to define the hexagonal lattice are not orthogonal. The two axes defining the basal plane, $\vec{a}_1$ and $\vec{a}_2$, have a 120° orientation relationship and the third axis $\vec{a}_3$ lies perpendicular to both $\vec{a}_1$ and $\vec{a}_2$. The lattice constant along $\vec{a}_1$ and $\vec{a}_2$ is a , and along $\vec{a}_3$ is c . The $\vec{a}_3$ axis, elongated relative to the basal plane axes, has the relationship of $\frac{c}{a} = \sqrt{\frac{8}{3}}$ assuming a perfectly close-packed arrangement of spheres.

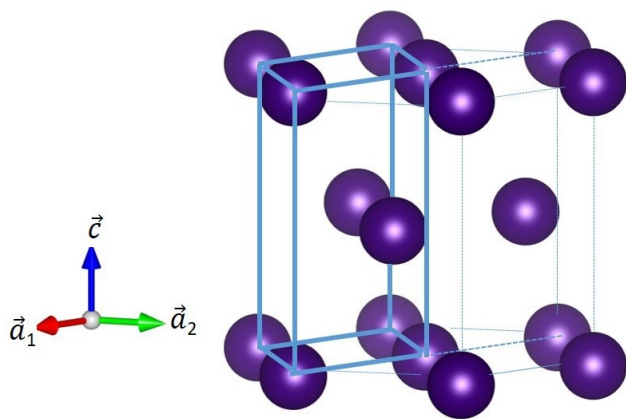


Fig. 5 Primitive HCP cell inscribed inside of a conventional unit cell

The (001) basal plane of the HCP structure and the (111) plane of the FCC structure are equivalent. The (111) FCC plane is shown in Fig. 6a. It is oriented slightly differently in Fig. 6b to show parallel $\{111\}$ planes. Figure 6c shows the (001) HCP planes.

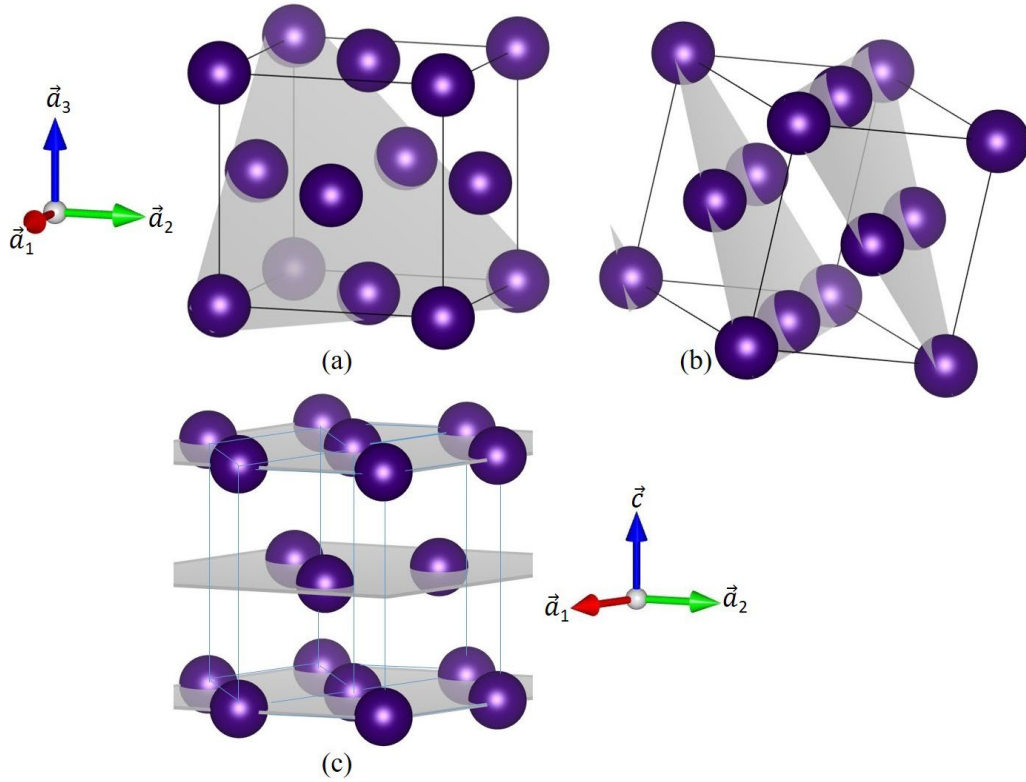


Fig. 6 a) Single (111) plane in FCC unit cell, tipped in b) so multiple (111) planes are seen. c) Crystallographically equivalent (001) HCP planes.

Figure 7 shows a) a (111) FCC plane and b) the (001) HCP plane with the spheres drawn to fill all possible empty space. We show the outline of the conventional unit cells as a guide to the eye. Both planes have the same symmetry with the spheres arranged so that each is in contact with six others. This arrangement maximizes the packing fraction, which is ≈ 0.74 for both unit cells.

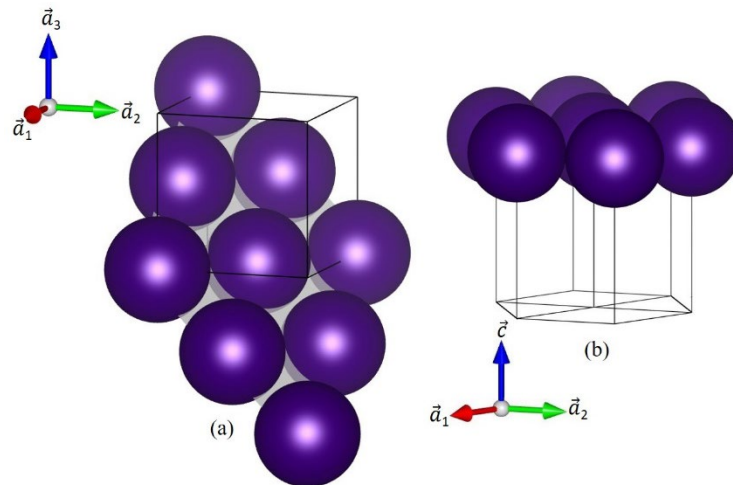


Fig. 7 Equivalent a) (111) FCC and b) (001) HCP planes

The physical difference in the HCP and FCC structure has to do with the stacking of these closest-packed planes. In the FCC structure, the planes are stacked with the a-b-c pattern (Fig. 8a), while the HCP planes are stacked with the a-b-a-b pattern (Fig 8b).

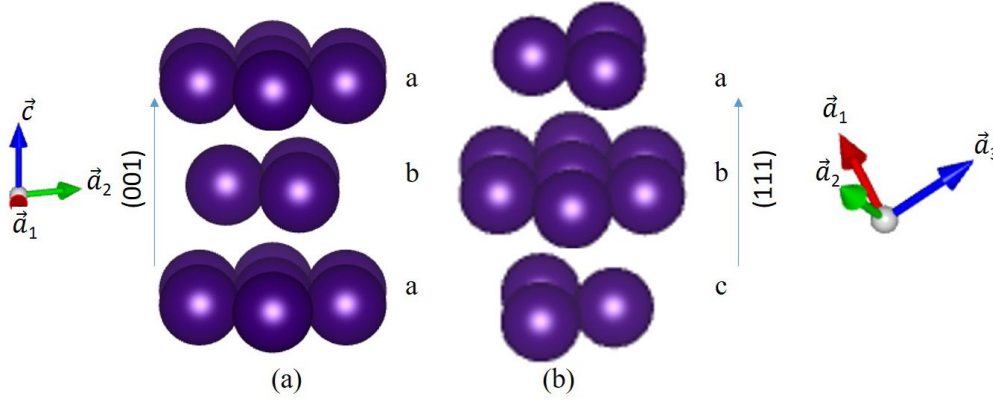


Fig. 8 a) a-b-a stacking in an HCP cell and b) a-b-c stacking in an FCC cell

2. Indexing Systems

We used three-index notation, known as *Miller indices*, to describe the simple cubic and HCP crystal structures discussed in the prior sections. In this section, we compare this system to the four-index *Miller–Bravais indices*.

2.1 Three-Index Miller System

For simple cubic crystals, Miller indices are very convenient due to the crystal's high symmetry. The three axes are orthogonal, as discussed in Section 1.1. For instance, (hkl) planes are perpendicular to $[hkl]$ directions. Simple dot and cross products of indices allow us to determine the orientation of the planes and lattice directions relative to one another and distances between planes.

For the HCP system, Miller indices are not as convenient. Because the basal plane axes are not perpendicular to one another (i.e., $\vec{a}_1$ and $\vec{a}_2$ have a 120° orientation relationship) as described in Section 1.2, we cannot use simple dot and cross products of the indices to find orientation relationships or distances between planes. One complication of the HCP structure is that planes do not necessarily have the same indices as their normal. Figure 9a is a conventional HCP unit cell with its close-packed planes and directions labeled. Figure 9b is a top-down view of the (001) plane. The normal to (010) plane lies along the $[120]$ direction, the normal to the (100) plane is $[210]$, and the normal to the $(\bar{1}10)$ plane is $[\bar{1}20]$. Only the (001) plane shares the same indices as its $[001]$ normal.

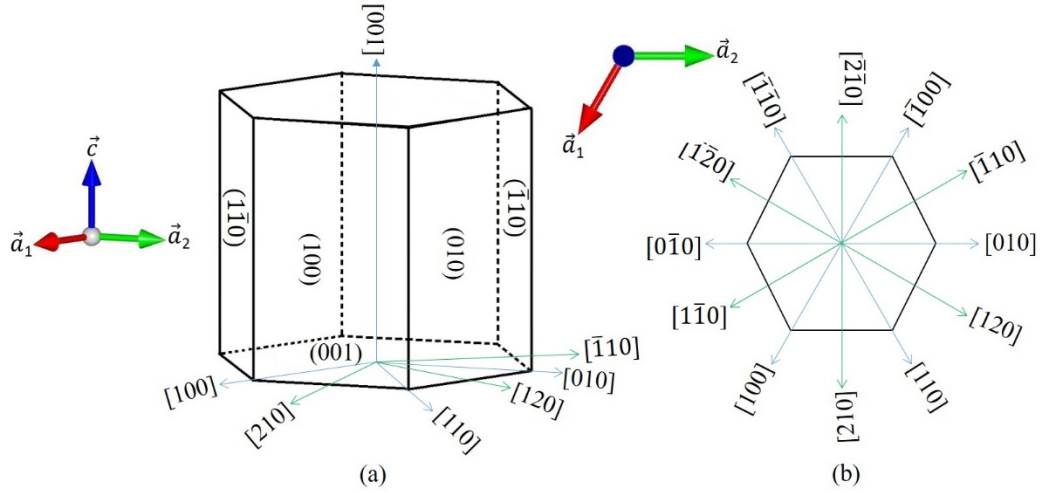


Fig. 9 a) Conventional HCP unit cell with its close-packed planes and directions labeled. b) A top-down view of the (001) plane.

Another challenge is that we cannot determine symmetry relationships by looking at the plane or direction indices. For instance, the (100) and (010) planes are equivalent, which seems obvious, but both are also equivalent to the $(1\bar{1}0)$ plane, which does not seem obvious. Meanwhile, the (001) and (100) planes are not equivalent at all. A close-packed schematic of the (100) plane is shown in Fig. 10a, which clearly is different from the arrangement of the (001) planes shown in Fig. 10b.

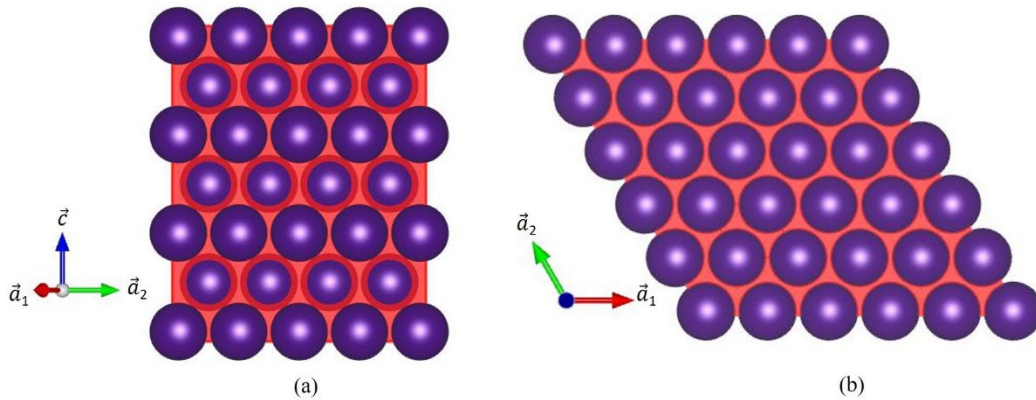


Fig. 10 a) $[210]$ projection of the (100) HCP plane and b) $[001]$ projection of the (001) HCP plane

2.2 Four-Index Miller–Bravais System

Using the four-index Miller–Bravais system relieves some of the shortcomings of the three-index system for HCP materials. Shown schematically in Fig. 11, the

“extra” axis lies in the basal plane, and all three basal axes are 120° from each other. The fourth axis is orthogonal to all three basal plane axes.

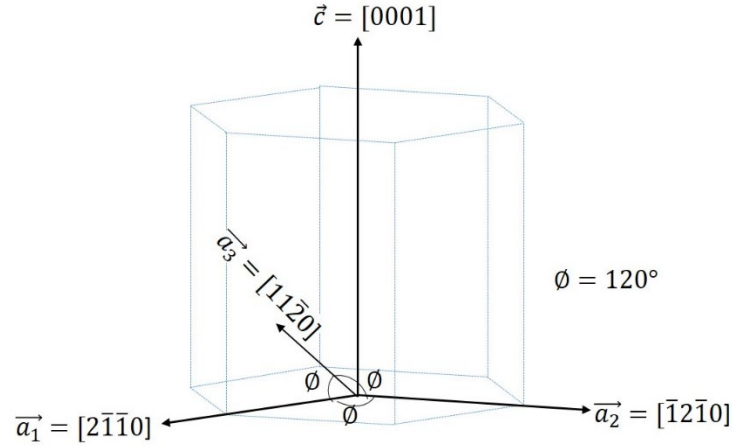


Fig. 11 Defining the axes of the HCP system using Miller–Bravais notation

Table 1 lists the conversion between the three-index (hkl) and four-index ($HKIL$) systems for common planes. The additional index I is dependent on H and K , as shown by the relationship in Eq. 3.

$$H + K + I = 0 \quad (3)$$

Table 1 Three- and four-index conversion for planes

(hkl)	$(HKIL)$
(001)	(0001)
(010)	(01 $\bar{1}$ 0)
(100)	(10 $\bar{1}$ 0)
(1 $\bar{1}$ 0)	(1 $\bar{1}$ 00)
(2 $\bar{1}$ 0)	(2 $\bar{1}$ $\bar{1}$ 0)
($\bar{1}$ 20)	($\bar{1}$ 2 $\bar{1}$ 0)
(110)	(11 $\bar{2}$ 0)

The extra index clarifies symmetry, because it allows families of planes and directions in the Miller–Bravais system have similar-looking indices, as shown in Fig. 12. For instance, the $\{1\bar{1}00\}$ family includes the symmetric (01 $\bar{1}$ 0), (0 $\bar{1}$ 10), (10 $\bar{1}$ 0), ($\bar{1}$ 010), (1 $\bar{1}$ 00), and ($\bar{1}$ 100) planes. Using the four-index notation, HCP planes now have the same indices as their normal, as it was for the cubic system.

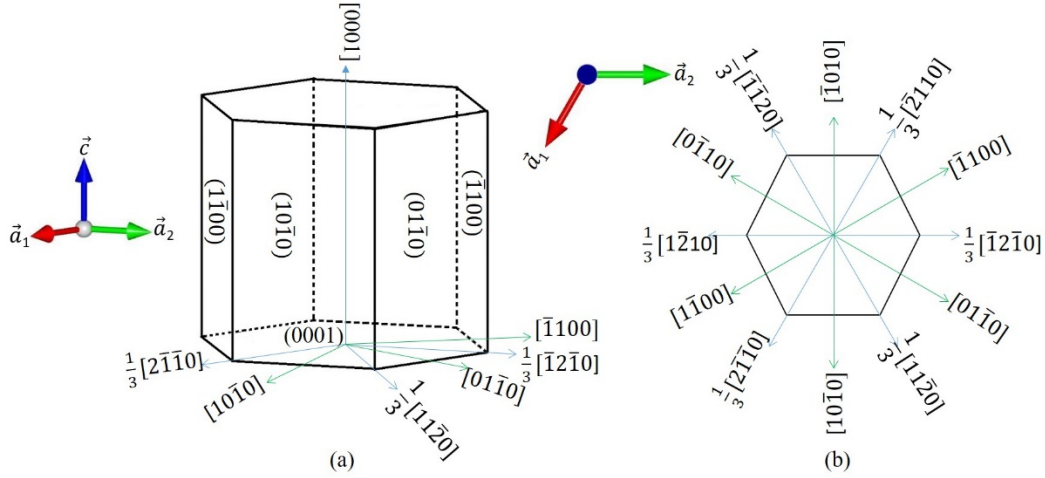


Fig. 12 Labeled Miller-Bravais planes and directions of the a) conventional HCP unit cell b) projected along $[0001]$

It is not as straightforward to convert directions between the Miller $[hkl]$ and Miller-Bravais $[HKIL]$ systems. In the Miller system for HCP crystals, any given vector can be written as

$$\vec{v} = h\vec{a}_1 + k\vec{a}_2 + l\vec{c}. \quad (4)$$

Similarly, with the Miller-Bravais system, the same vector can be written as

$$\vec{v} = H\vec{a}_1 + K\vec{a}_2 + I\vec{a}_3 + L\vec{c}. \quad (5)$$

We can substitute the relationship

$$\vec{a}_1 + \vec{a}_2 = \vec{a}_3 \quad (6)$$

into Eq. 5 and obtain

$$\vec{v} = (H - I)\vec{a}_1 + (K - I)\vec{a}_2 + L\vec{c}. \quad (7)$$

Since

$$I = -(H + K), \quad (8)$$

we can rewrite Eq. 7 as

$$\vec{v} = (2H + K)\vec{a}_1 + (2K + H)\vec{a}_2 + L\vec{c}. \quad (9)$$

Now we equate the coefficients in Eqs. 4 and 9 and obtain

$$h=2H+K \quad k=2K+H \quad l=L \quad (10)$$

Solving in terms of $HKIL$,

$$H = \frac{1}{3}(2h - k) \quad K = \frac{1}{3}(2k - h) \quad I = -(H + K) \quad L = l \quad (11)$$

Table 2 lists common HCP crystal directions in the three- and four-index notation.

Table 2 Three- and four-index conversion for directions

$[hkl]$	$[HKIL]$
[001]	[0001]
[010]	$\frac{1}{3}[\bar{1}2\bar{1}0]$
[100]	$\frac{1}{3}[2\bar{1}\bar{1}0]$
[110]	$\frac{1}{3}[11\bar{2}0]$
[210]	[10 $\bar{1}$ 0]
[120]	[01 $\bar{1}$ 0]
$[\bar{1}10]$	$[\bar{1}100]$

The $\frac{1}{3} < 2\bar{1}\bar{1}0 >$ family of directions is often represented by removing the common denominator, becoming $< 2\bar{1}\bar{1}0 >$. While not obvious for permutations of h, k , and l indices, we can easily identify symmetrically equivalent directions by permutations of H, K, I , and L . The lowest-index planes are usually the most favorable for high-resolution TEM imaging. We typically prepare our samples with the [0001], $< 2\bar{1}\bar{1}0 >$, or $< 01\bar{1}0 >$ family of zone axes, as shown in Fig. 13. These zone axes are orthogonal, and imaging more than one is often necessary to identify dislocations. The (0001) basal planes are known as the c-planes, the $\{01\bar{1}0\}$ as the m-planes, and the $\{2\bar{1}\bar{1}0\}$ as the a-planes. For cross-sectional high-resolution TEM imaging, the closest-packed plane available is usually one of the three prismatic $\{01\bar{1}0\}$ planes. Figure 13a shows a projection of the [0001] axis with the $\{10\bar{1}0\}$ family of planes labeled. Figure 13b shows the projection along the $[10\bar{1}0]$ axis with orthogonal $(1\bar{2}10)$ and (0001) planes labeled to serve as an orientation marker. Figure 13c is a projection of the [0001] axis with the $\{2\bar{1}\bar{1}0\}$ planes labeled. Figure 13d is the projection along the $[2\bar{1}\bar{1}0]$ axis with the orthogonal $(01\bar{1}0)$ and (0001) planes labeled.

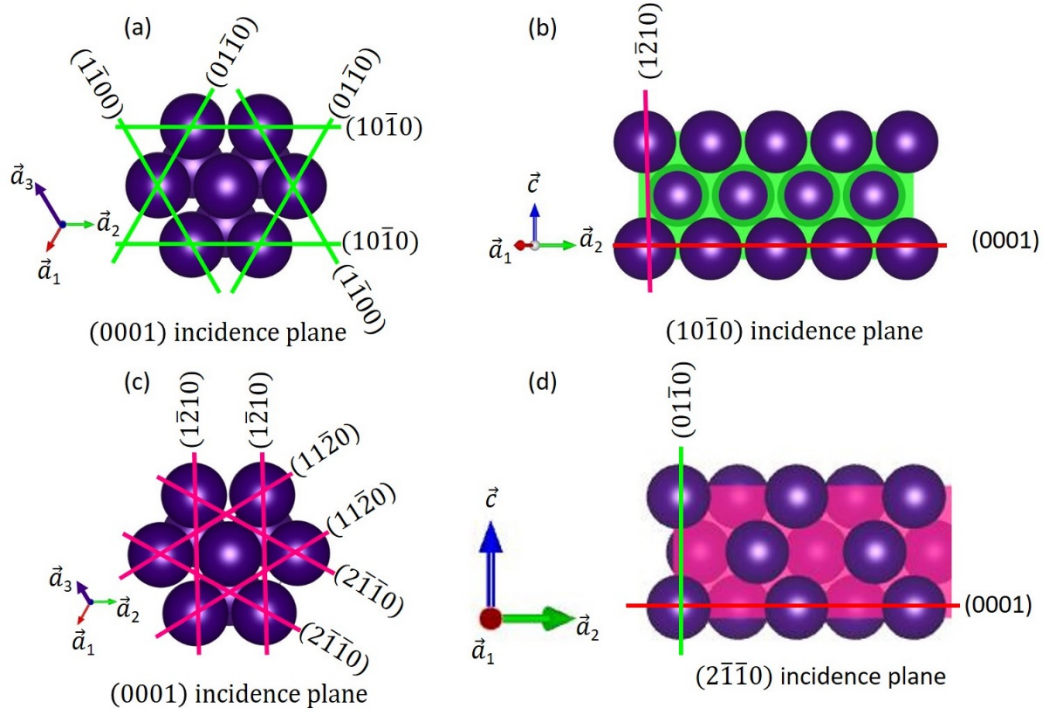


Fig. 13 Projections of a HCP crystal along a) the $[0001]$ direction with the $\{10\bar{1}0\}$ family of planes labeled, b) $[10\bar{1}0]$, c) $[0001]$ with the $\{2\bar{1}\bar{1}0\}$ planes labeled, and d) $[2\bar{1}\bar{1}0]$ with the (0001) and $(01\bar{1}0)$ planes labeled

3. Important Example: GaN

GaN is a good, illustrative HCP crystal grown at ARL. GaN is polymorphic, with the two most-prevalent crystals shown in Fig. 14: a) 3C (cubic) and b) 2H (hexagonal).

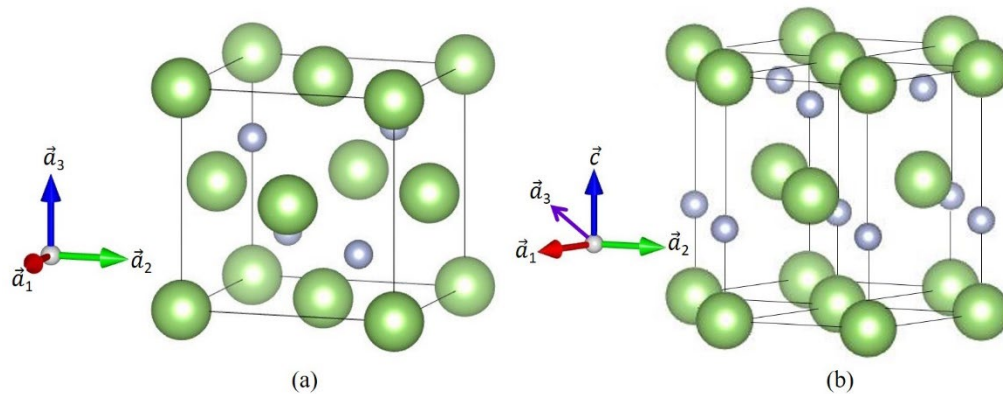


Fig. 14 a) 3C GaN and b) 2H GaN

The FCC 3C-GaN structure is zinc-blende, meaning that the larger gallium (Ga) atom is at the origin and the smaller nitrogen (N) atom is located $1/4$ of a body

diagonal from the origin. The 2H GaN base structure has a Ga atom at the origin $[0,0,0]$ and another at $[\frac{1}{3}, \frac{2}{3}, \frac{1}{2}]$. The N atoms at $[0,0,\frac{3}{8}]$ and $[\frac{1}{3}, \frac{2}{3}, \frac{7}{8}]$ are directly over the Ga atoms, displaced by $\frac{3}{8}$ of the c-dimension of the unit cell along the $\vec{c}$ axis.

4. Diffraction Patterns

The ARM-200F at ARL typically operates at an acceleration voltage of 200 keV. This corresponds to a relativistic beam wavelength of 0.0251 Å (2.5 pm). The periodic crystal being examined acts as a diffraction grating. TEM diffraction patterns complement images in allowing full characterization of a crystal's morphology.

Figure 15 shows HCP diffraction patterns for the zone axes corresponding to a) $[\bar{2}110]$, b) $[0\bar{1}10]$, and c) $[0001]$. The simulated patterns were generated with the CrystalMaker software.

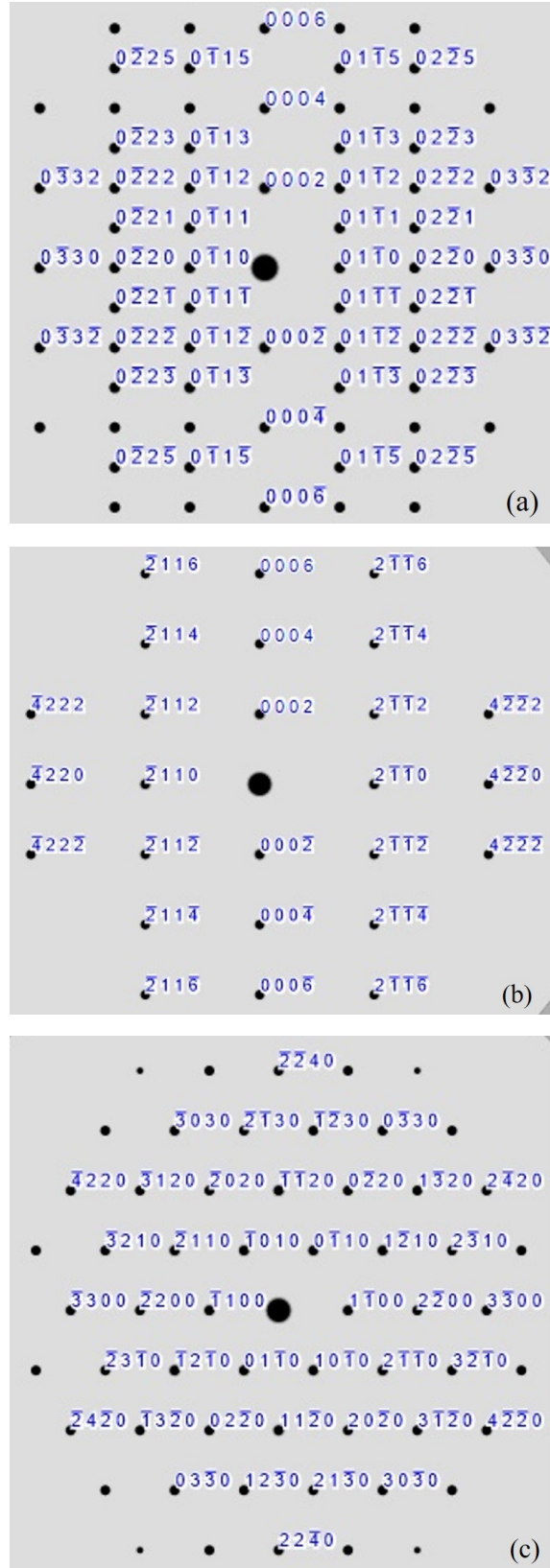


Fig. 15 HCP diffraction pattern for the (a) $[2\bar{1}10]$, (b) $[01\bar{1}0]$, and (c) $[0001]$ zone axes

5. Summary and Conclusions

This report thoroughly described the three- and four-index systems for HCP materials, with a comparison to the FCC material system.

It is important to avoid two frequent mistakes when using either the Miller or Miller–Bravais system. The first is using dot and cross product calculations on the Miller system for HCP materials, since the basal plane axes are not orthogonal. The other common error is using the same conversion relationship from three to four indices for both planes and directions. While one can easily drop the I index from a $(HKIL)$ plane and convert it to an (hkl) plane, one must use the formulas in Eqs. 10 and 11 to convert $[HKIL]$ directions to $[hkl]$ directions. These sorts of errors, in addition to misrepresenting the crystal's structure, cause major problems when determining dislocation visibilities.

While it may seem that one should just use the Miller–Bravais system for ease of describing symmetry, and so on, a basic familiarity with the Miller system for HCP materials is necessary. The Miller–Bravais system is not completely conventional internationally, and some journals exclusively use the Miller system. Software packages for crystal simulation sometimes only use the Miller system. CrystalMaker understands and interprets the Miller–Bravais system, but other software packages do not. Furthermore, there are numerous online calculators for determining angles between planes and directions, or lattice spacings, which rely on the Miller system. Therefore, it is critical that anyone studying the crystallography of HCP materials is literate in both the Miller and Miller–Bravais systems.

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List of Symbols, Abbreviations, and Acronyms

ARL	Army Research Laboratory
DEVCOM	US Army Combat Capabilities Development Command
FCC	face-centered cubic
Ga	gallium
GaN	gallium nitride
HCP	hexagonal close-packed
III-N	III-Nitride; alloy consisting of one or more group III elements and nitrogen
III-V	alloy consisting of one or more group III elements and one or more group V elements
N	nitrogen
SiC	silicon carbide
TEM	transmission electron microscopy

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