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Institut de Physique de Montpellier, University of Montpellier 2, Montpellier, Cedex, France ISSN 0933-033X e-ISSN 2196-2812 ISBN 978-3-319-06804-6 e-ISSN 978-3-319-06805-3 DOI 10.1007/978-3-319-06805-3 Springer Cham Heidelberg New York Dordrecht London Library of Congress Control Number: 2014940318 © Springer International Publishing Switzerland 2014 This work is subject to copyright. All rights are reserved by the Publisher, whether the whole or part of the material is concerned, specifically the rights of translation, reprinting, reuse of illustrations, recitation, broadcasting, reproduction on microfilms or in any other physical way, and transmission or information storage and retrieval, electronic adaptation, computer software, or by similar or dissimilar methodology now known or hereafter developed. Exempted from this legal reservation are brief excerpts in connection with reviews or scholarly analysis or material supplied specifically for the purpose of being entered and executed on a computer system, for exclusive use by the purchaser of the work. Duplication of this publication or parts thereof is permitted only under the provisions of the Copyright Law of the Publisher's location, in its current version, and permission for use must always be obtained from Springer. Permissions for use may be obtained through RightsLink at the Copyright Clearance Center. Violations are liable to prosecution under the respective Copyright Law. The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use. While the advice and information in this book are believed to be true and accurate at the date of publication, neither the authors nor the editors nor the publisher can accept any legal responsibility for any errors or omissions that may be made. The publisher makes no warranty, express or implied, with respect to the material contained herein. Printed on acid-free paper Springer is part of Springer Science+Business Media (www.springer.com) Preface This book aims at offering the master students "and probably to other ones" who are studying the physics of wide bandgap

semi-conductors, the elements required to rapidly grasp the concept of solid state physics that are needed to start a formation or a research activity at the boarder between physics, chemistry, electrical engineering. This field has known tremendous developments during the past 20 years, and it will probably continue to be very exciting: so many applications are possible to wide band gap semi-conductors and so few have been satisfied to date. I have been routinely working on optical properties of wide band gap semi-conductors, of their heterostructures, of their nanostructures since 1991 and I could attend every year a lot of scientific international conferences. Each time was the opportunity for me to meet new faces, sometimes very young ones. Most of these newcomers had received a formation in chemistry, electronics, physics, mathematics, or another one, sometimes including astronomy. They are often launched into the international scientific arena just after having spent a few weeks or months in immersion into a research group, and they have to accommodate a lot of concepts in very different domains. This is not so easy for them. I had a lot of opportunities to chat with many of these newcomers. I got convinced as many of my colleagues are now, too, that there is a need for a general book in which they could find gathered most of the concepts that they need to know. When Claus Ascheron from Springer asked me to write such book, I accepted it without having in mind the challenging proposal he had made to me. You are holding such a book in your hands. The field is wide and necessarily unexhaustively addressed, but I hope this monograph contains a strong enough message for being of value for a lot of my young colleagues. This book is declined along five chapters: basic symmetry and physical properties linked to it, basics of growth and structural characterization methods, band structure effects and lattice vibrations, optical properties of bulk materials, and finally physics and optical properties of low-dimensional systems. Photonics, quantum optics, plasmonics, and transport properties are not treated; they are from very much specialized areas and are addressed or in the way of being addressed in specialized research books. This book is the fruit of many collaborations. I would first like to thank the many students that I contributed to train. Many of them now being colleagues in some universities or at the National Centre of Scientific Research, my employing institution. Let me thank Philippe Boring, Pierre Bigenwald, Pierre Lefebvre, Matthieu Moret, Lionel Aigouy, Claude Boemare, Sandra Ruffenach, Andenet Alemu, Magloire Tchounkeu, Francois Demangeot, Cyril Pernot, Abdelhadi Abounadi, Amal Rajira, Tomasz Ochalski, Yves-Mathieu Le-Vaillant, Marian Zamfirescu, Mathieu Gallart, Aurelien Morel, Xue Bing Zhang, Sokratis Kalliakos, Romuald Intartaglia, Benedicte Maleyre, Richard Bardoux, Stephane Faure, Luc Beaur, Daniel Rosales, Julien Selles, Huong Thi Ngo, and Rereao Hahe. I had a chance to work with a lot of handsome colleagues among which are Olivier Briot, Mathieu Leroux, Jean-Yves Duboz, Nicolas Grandjean, Eric Tournie, Benjamin Damilano, Julien Brault, Jean-Michel Chauveau, Christian Morhain, Thierry Bretagnon, Amelie Dussaigne, Jean Massies, Bernard Beaumont, Pierre Gibart, Philippe Vennegues, Pierre Ruterana, Thierry Guillet, Christelle Brimont, Pierre Valvin, Christian L  henoret, Thierry Taliercio, Didier Felbacq, Brahim Guizal, Guillaume Cassabois, Emmanuel Rousseau, Christelle Eve, Jacques Leyzat, Regine Puzat, Bruno Daudin,   ! At the international scale, Izabella Grzegory, Hadis Morko  , Isamu Akasaki and Shuji Nakamura have been my mentors in the early days of the nitrides. I took advantages of fruitful exchanges with Yabusiko Arakawa, Bo Monemar, Alex Zunger, Hiroshi Amano, Akihiko Yoshikawa, Yasuhi Nanishi, Yoichi Kawakami, Kazamasu Hiramatsu, Katsumi Kishino, Shigefusa Chichibu, Hideto Miyake, Alexey Kavokin, Axel Homann, Martin Stutzmann, Bruno Meyer, Juergen Christen, Jorg Neugebauer, Friedhelm Beschedt, Alois Krost, Andreas Hangleiter, Martin Feneberg, Ruediger Goldhahn, Charles Foxon, Kevin O  Donnell, Rob Martin, Peter Parbrook, Galia Pozina, Tatiana Shubina, Serguey Ivanov, Valery Yu Davidov, Tanya Paskova, Plamen Paskov, Fernando Ponce, Russell Dupuis, Ted Moutsakas, Steven den Baars, Umesh Misra, James Speck, Zlatko Sitar, Eva Monroy, Xinqiang Wang, Piotr Perlin, Tadeusz Suski, Abderrahmane Kadri, Karima Zitouni, and so many others. Pierre Bigenwald who carefully

browsed the five chapters to expurgate them from typos deserves receiving specific acknowledgments. Bernard Gil Montpelliér Contents

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Bernard Gil Physics of Wurtzite Nitrides and Oxides Springer Series in Materials Science 19710.1007/978-3-319-06805-3_11. Basic Crystallography and Other Properties Linked with Symmetry Bernard Gil 1 (1)

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Bernard Gil Email: bernard.gil@univ-montp2.fr This chapter treats of crystallography, basic physical properties linked to the crystal structure. It also gives a few elements of basic group theory. **1.1 The Hexagonal Point Symmetry Deduced from the Shape of Natural Wurtzitic Crystals** Atomic organisation of wurtzitic semi-conductors follows the crystallographic structure named "zincite" Zincite, that was, to the author's knowledge, first described as red oxide of zinc by American mineralogist Archibald [1] at the dawn on the nineteenth century. Some sulfide minerals also crystallize according to this atomic organisation, among which are Greenockite greenockite (CdS), first observed in 1840 in Bishopton, Scotland, and named after the landowner Lord Greenock. The term "wurtzite" • Wurtzite is correlated with the description of the hexagonal form of zinc

sulfide crystals in 1861 and named after French chemist Charles-Adolphe Wurtz. The wurtzite group also includes, rambergite (MnS) discovered in sediments and named at the end of the twentieth century after the Norwegian-Swedish mineralogist, Hans Ramberg. We also wish to indicate that cadmoselite CdSe (artificially synthesized before it was identified as a mineral stone) was discovered in 1957. In the language of semi-conductorists, these materials belong to the group of II-VI compounds: the cation (resp. anion) is a group II (resp. VI) element of Mendeleev's table. Apart from the II-VI family, III-nitrides (III-Ns) are also important semi-conductors the cation is a group-III element generally boron, aluminium, gallium or indium. The most energetically favourable atomic organisation for boron nitride is not wurtzite; BN is a layered compound like graphite. The most stable crystalline structure of AlN, GaN and InN semi-conductors is wurtzitic. These materials are not extracted from mines as binary compounds probably due to the high stability of the nitrogen molecule (9.3 eV per bond) or to the chemical reactivity of ammonia. NH₃ is a very stable molecule: nitrogen single atoms are highly reactive and do not live long without binding to the most abundant elements in nature: H, O, etc. Therefore, the necessary conditions to associate nitrogen and group III element in order to form pure III-Ns are never met in nature, to the best of our knowledge. The existence of AlN was demonstrated in 1862 by Briegleb and Geuther [2] and Mallet synthesized it in 1876 [3] with metallic aluminum and sodium carbonate reactant at high temperatures. The growth of GaN was achieved in 1932 by Johnson et al. [4] later achieved, in 1938, the growth of indium nitride was later achieved, in 1938 [5].

The hexagonal symmetry of wurtzite crystals was quantitatively calibrated during the nineteenth century from the orientation of the directions orthogonal to the facets of crystals found in mines. Crystals are most generally found as small needles, exhibiting specific shapes as shown in Fig. 1.1. *Fig. 1.1* Photograph of a natural wurtzitic crystal. Note the needle-like shape and the existence of many facets with different orientations illustrating the hexagonal symmetry. *Fig. 1.2* *Left* Photograph of an artificial crystal of bulk gallium nitride (courtesy Dr. I. Grzegory, Polish Academy of Sciences). *Right* Photograph of an aluminum nitride bulk artificial single crystal (courtesy Prof. Zlatko Sitar, North Carolina State University) Artificially grown bulk semi-conductors like gallium nitride (Fig. 1.2-left) or aluminum nitride (Fig. 1.2-right) are also obtained under faceted crystalline shapes. Although the shape of the crystals presented in Fig. 1.2 (left) and 1.2 (right) are fairly different, they share a common property. The shape of the bunch of directions orthogonal to the crystal facets constitutes some kind of figure of merit. The figure obtained after the stereographic projection of the ensemble of the directions that are orthogonal to the facets of these crystals is compatible with the symmetry of a regular hexagonal polygon. This figure is a constant quantity typical of the crystalline structure according to the first law of crystallography. It is known as "the law of constant angles between crystal facets", early initiated by Danish Nicolas Steno's seminal work in 1669 and later formulated by French mineralogist Jean-Baptiste Romé de l'Isle in 1772. For wurtzitic semi-conductors, this figure permits to reveal a six-fold symmetry along a given direction, the existence of a three-fold and two-fold symmetries collinear to the six fold one and six symmetry planes parallel to the six-fold symmetry axis. These symmetry elements are generic of the crystalline form named di-hexagonal pyramidal by mineralogists. In terms of modern group theory language, from the shape of these needles, one establishes that the orientation (or point symmetry) of wurtzite crystals is a subgroup (labelled C in Schoenflies notation or P in the Hermann-Mauguin one) of the full hexagonal symmetry (labelled D in Schoenflies notations or P in the Hermann-Mauguin one). French abbot René-Just Haüy in 1774 carefully observed facets of minerals and proposed the second law of crystallography: crystals consist of three-dimensional stacking at a macroscopic scale of a given microscopic basic building block as tentatively illustrated in Fig. 1.3. This kind of wooden crystal were still used as tutorial examples to teach students

in the middle of the twentieth century. *Fig. 1.3* Photograph of wooden tutorial object designed such as to show a typical repeat of basic building blocks which at the end to form the whole crystal shape. In 1849, French scientist Auguste Bravais postulated the principle of specific three-dimensional translational invariance (existence of translational symmetry operations on the basic building block imagined by $h\mathbf{a} + \frac{1}{2}y$). From this postulate results the notion of three-dimensional periodic crystalline lattice which is the basic principle used to determine the crystallographic structure of materials using radio-crystallography. Before discussing this very recent technique since X-rays were discovered by German physicist Wilhelm Conrad Röntgen in 1895, it is worthwhile allocating some time to simple mathematical analysis of crystal properties in line with their symmetries. We will restrict ourselves to wurtzite and, in particular, catch the opportunity to show how powerful the first laws of crystallography are. From the bunch of angles made by the directions normal to the crystal's facet, we can determine the ratio of the basic hexagonal building block parameter a along the six-fold symmetry axis to the one orthogonal to it.

1.2 The Hexagonal Lattice, Its Reticular Planes and Their Description Using Simple Euclidian Geometry

The hexagonal crystallographic system is based on two vectors of identical length at from each other, and a third one, of length orthogonal to the plane generated by the preceding two, as described by French scientist Auguste Bravais in 1849 and as illustrated in *Fig. 1.4*. *Fig. 1.4* Representation of the crystallographic axes and that generate the unit hexagonal cell (blue lines). The full hexagonal cell is shown for the completeness. Lengths and being the dimensions of the lattice vectors of the hexagonal cell, we associate them to unit vectors, \mathbf{a}_1 and \mathbf{a}_2 . This gives: and We now consider the (hk) reticular plane in terms of its so-called Miller indices. This notation was proposed by British Mineralogist William Hallows Miller in 1839. By definition, the (hk) plane intersects the reticular axes at lengths $\frac{1}{h}$ and $\frac{1}{k}$ from the origin as shown in *Fig. 1.5*. Letters h , k and are algebraic integer numbers. There are some specific simple orientations for such planes: Miller indices (100) represent a plane parallel to the (010) plane, Miller indices (010) represent a plane parallel to the (100) plane, Miller indices (001) represent a plane parallel to the $(00\bar{1})$ plane. The notation $\{hk\}$ is traditionally used to identify all the planes equivalent to (hk) by the symmetry of the lattice. As an example, (100) and (010) planes belong to the $\{100\}$ family. Thanks to hexagonal symmetry, and in contrast to the cubic case, $[hk]$ with square brackets represent a direction in the basis of the direct lattice vectors: $[hk]$ is equivalent to $h\mathbf{a}_1 + k\mathbf{a}_2$. The notation is used for all the directions that are equivalent to $[hk]$ by symmetry operations of the crystal. In the most general case, and for non-cubic crystals, direction $[hk]$ is NOT orthogonal to plane (hk) . Given an (hk) plane, we define three unit vectors $(\mathbf{u}, \mathbf{v}, \mathbf{w})$ orthogonal to each other, two of them (\mathbf{u} and \mathbf{v}) being vectors of the (hk) plane, whilst the third one (\mathbf{w}) is orthogonal to it. *Fig. 1.5* Orientation of the reticular plane (hk) with respect to the crystallographic axes of the hexagonal lattice. The direction (\mathbf{u}) is parallel to vector \mathbf{a}_1 in *Fig. 1.5* is chosen as $[-k\mathbf{a}_1]$, and thus lies in the (\mathbf{u}, \mathbf{v}) plane. \mathbf{u} satisfies the sonal equation and corresponds to a crystal direction. The $[-0h]$ direction (vector \mathbf{v} in *Fig. 1.5*) connects intersections of the (hk) plane with crystallographic directions \mathbf{a}_1 and \mathbf{a}_2 at lengths $\frac{1}{h}$ and $\frac{1}{k}$ from the origin. It forms with \mathbf{u} a basis of the (hk) plane. This direction is not orthogonal to \mathbf{w} , which is not the best choice for mathematical calculations. Thus, it is more appropriate to determine, using an orthonormalization procedure a vector \mathbf{v} lying in the (hk) plane and perpendicular to \mathbf{u} . This vector is parallel to vector \mathbf{a}_2 with \mathbf{u} as indicated in *Fig. 1.5*. It also satisfies the zone equation for the (hk) plane, demonstrating that it corresponds to a crystal direction. To this couple of vectors (\mathbf{u}, \mathbf{v}) , we add \mathbf{w} , also obtained after an orthonormalization procedure. In the hexagonal $(\mathbf{u}, \mathbf{v}, \mathbf{w})$ basis, the coordinates of these unit vectors are: $(1, 0, 0)$ where \mathbf{u} and \mathbf{v} are in the (hk) plane and \mathbf{w} is perpendicular to it. The angle between the $[001]$ direction and \mathbf{w} is given as: that depends on the value of the three Miller indices and on the ratio c/a . At this stage, elementary Euclidian algebraic calculations demonstrate the possibility to determine the ratio in wurtzitic

crystals from the orientation of the directions of the crystal facets. The only issue is to have crystals with a large enough number of facets. Thus, we gain access to a well documented set of experimental values of α , so that their associated h , k and l numbers can be determined simultaneously with the value of α . Table 1.1 summarizes the results of c/a ratio typical of the most common wurtzitic semi-conductors. Table 1.1 Values of the ratio c/a for various wurtzitic semi-conductors

Material BN GaN InN AlN ZnS ZnSe CdS CdSe ZnO

1.641.621.611.61.641.631.621.631.60

The number of digits is compatible with the determination method 1.3 The Four-index Four-Index Bravais-Miller Representation of the Orientation of Reticular Planes in Hexagonal Crystals In the case of the hexagonal (and rhomboidal) Rhomboidal lattice systems, one alternative convention is to use a 4-numbers representation $(h\ k\ i\ l)$ for reticular planes, where l is the Miller index. In this case, h , k and i are identical to the Miller indices, and l is redundant: four independent vectors cannot generate a three-dimensional space. Figure 1.6 illustrates some typical orientations of (hk) planes that are found in natural crystals. Fig. 1.6 Sketches of typical orientations of reticular planes of interest for wurtzitic semi-conductors. The four-indices Miller-Bravais indexing system is used to represent the orientations This four-indices scheme for labelling Planes planes in a hexagonal lattice is very convenient to illustrate the identical nature of reticular planes by permutation of the indices. For example, we better spot the similarity between planes (110) and $(1\bar{1}0)$ than when they are written as (110) and (10) .

1.4 Representation of Hexagonal Crystal Directions Using Four Indices In some books of Mineralogy mineralogy, we find another four-indices system for the directions in hexagonal crystals. Let: This direction may be also noted as $u\ v\ w$ and is fully determined with respect to the hexagonal cell. The corresponding Weber Weber four indices are defined as: $u\ v\ w\ t$ and the relationships are: where t is a factor used to transform the new indices into smaller integers. To avoid confusion, we will not use this notation.

1.5 The Reciprocal Lattice The Reciprocal lattice reciprocal lattice is not necessary for geometric crystallography Geometric crystallography. However, it facilitates some calculations, and its utilization is mandatory when studying X-ray diffraction by periodical structures or, out of the context of crystallography, to treat band structure phenomena, or light propagation. The reciprocal lattice is the Fourier transform of the direct lattice.

This book gives a survey of the current state of the art of a special class of nitrides semiconductors, Wurtzite Nitride and Oxide Semiconductors. It includes properties, growth and applications. Research in the area of nitrides semiconductors is still booming although some basic materials sciences issues were solved already about 20 years ago. With the advent of modern technologies and the successful growth of nitride substrates, these materials currently experience a second birth. Advanced new applications like light-emitters, including UV operating LEDs, normally on and normally off high frequency operating transistors are expected. With progress in clean room technology, advanced photonic and quantum optic applications are envisioned in a close future. This area of research is fascinating for researchers and students in materials science, electrical engineering, chemistry, electronics, physics and biophysics. This book aims to be the ad-hoc instrument to this active field of research.

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