Simultaneous observation of Kikuchi pattern contrast and surface morphology contrast in secondary electron image using a cylindrically symmetric rotating crystal

(B. Da$^{1,2}$$^*$, L. Cheng$^{1,3}$, X. Liu$^{1,3}$, K. Shigeto$^4$, T. Kizu$^5$, K. Tsukagoshi$^{5#}$, T. Nabatame$^5$, Z. J. Ding$^{3†}$, Y. Sun$^6$, H. Jin$^7$, J. W. Liu$^8$, D.M. Tang$^5$, H. Zhang$^2$, Z. S. Gao$^9$, H. X. Guo$^{10}$, H. Yoshikawa$^{1,2}$, and S. Tanuma$^2$

$^1$Research and Services Division of Materials Data and Integrated Systems, National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

$^2$Research Center for Advanced Measurement and Characterization, National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

$^3$Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, China

$^4$Hitachi High-Tech Corporation, Hitachinaka, Ibaraki 312-8504, Japan

$^5$International Center for Materials Nanoarchitectonics (WPI-MANA), National Institute for Materials Science, Tsukuba, Ibaraki 305-0044, Japan

$^6$Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY, 10027, USA

$^7$Department of Physics and Institute for Nanoscience and Engineering, University of Arkansas, Fayetteville, Arkansas 72701, United States

$^8$Research Center for Functional Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

$^9$Institute of Electrical Engineering, Chinese Academy of Sciences, Beijing, 100190, P.R. China.

$^{10}$Key Laboratory of MEMS of Ministry of Education, School of Electronic Science and Engineering, Southeast University, Nanjing, 210096, People’s Republic of China.

*$DA.Bo@nims.go.jp$

#$zjding@ustc.edu.cn$
Abstract:
The Kikuchi lines arise from Bragg diffraction of incoherent electrons scattered within the specimen, which can be observed in both transmission and reflection modes of scanning electron microscopy (SEM). Technically, to obtain the Kikuchi pattern, converging beams, rocking beams or grazing incidence beams have to be used to generate the divergent electron sources. Here we present an extremely special rotating crystal with continuous rotation in the local crystal direction and satisfying cylindrical symmetry, named cylindrically symmetric rotating crystals. When using an SEM in the raster scanning mode, the backscattered electron images and secondary electron images show not only the surface morphological contrast and elemental composition contrast of the sample, but also diffraction contrast with a clear Kikuchi pattern. By retracing the Kikuchi line pattern, it is even possible to obtain the lattice structure, lattice constants, crystal orientation, stress information, defect concentration, and Brillouin zone information of the presented sample from the conventional SEM images. SEM images of these cylindrically symmetric rotating crystals contain the interactions between electrons and the presented sample in both real-space and momentum-space. It is a very interesting and representative new case for the study of the contrast mechanism in SEM.
1. Introduction

Scanning electron microscopy (SEM), one of the most common modern scientific instruments, produces images of the sample surface by scanning the surface with a focused electron beam [1]. The complex electron beam/sample surface interaction produces various signals at each pixel, which are detected and displayed on a scanning display unit that scans over the sample in parallel with the beam scan. When the focused beam hits a focal point on the specimen, the signal intensity is measured by a detector to integrate the dwell time and expressed as the brightness of the pixel in a digital image. Obviously, this gray-level image is related to some characteristics of the specimen and can be interpreted as various image contrast caused by different mechanisms. Therefore, it is very important to understand the concept of contrast and its numerical meaning in SEM. Any new contrast mechanism discovered in the SEM has the potential to expand the range of applications of SEM and give a boost to research and industrial production.

The most common mode of operation of SEM is the raster scan [2] on the surface of a specimen, i.e., energetic electrons are focused into a beam with a small spread, and the beam is incident on the sample in the same direction and scanned point by point. The electron beam is parallelly incident into the sample surface so that the resulting digital image is a real space image. Each pixel in the image corresponds to a different location in the real space of the sample. Throughout the historical development of SEM and its applications, various contrast mechanisms in raster scanning mode have been developed. Electrons interact with the sample surface to produce various signals containing information about the sample surface, in which the surface morphology and composition of the sample is essential [3,4]. Thus, the most common types of contrast are topographic contrast [5] and composition/elemental contrast [6,7], which are applicable and available for almost all specimens and provide the basis for forming SEM images. In addition,
some special contrast mechanisms, such as electric field contrast [8,9], and magnetic contrast [10,11], exist in certain types of materials and are present in certain types of materials and are closely related to their specific properties. All these contrasts can be explained by the particle nature of electrons.

Besides, SEM also produces diffraction contrast [12-16]. Diffraction contrast usually reflects the structural information of the specimen, such as grain orientation, local texture, phase identification and distribution. The mechanism for the formation of diffraction contrast can only be explained by the wave nature of electrons. If one wants to observe a relatively complete diffraction pattern of a crystal sample or a polycrystal sample using an SEM, one needs to change the mode of operation of the SEM.

There are two modes of SEM operation to obtain relatively complete diffraction patterns. The first one is to collect backscattered electrons from different directions exiting the sample surface when electrons are incident on the sample at a large angle, i.e., EBSD mode [17]. In this mode, when the incident electron beam glancing enters the sample, it is scattered by the atoms inside the sample, and a significant portion of these electrons escape from the sample surface due to the large scattering angle, which are called backscattered electrons [18]. In the process of leaving the sample, the backscattered electrons that meet the Bragg diffraction condition [19], \(2d\sin\theta = \lambda\), will diffract from a certain family of crystal planes of the sample, forming two conical surfaces with the central axis perpendicular to the family of crystal planes. The two conical surfaces will intersect with the receiving screen to form a bright band, i.e., the Kikuchi band. The centerline of each Kikuchi band corresponds to the intersection of the expanded crystal plane corresponding to this Bragg diffraction with the receiving screen. Because each Kikuchi line is associated with Bragg diffraction from one side of a set of lattice planes, it is an effective method for studying the internal
crystal structure of a substance [20], since it is observed by Kikuchi in transmission electron microscopy as early as 1928 [21].

The second mode to observe a complete diffraction contrast is to scan the single crystal sample surface by changing the incidence direction of the electron beam so that the electron beam forms a continuously changing angle with the crystal direction of the sample [22]. Or, by vibrating the electron beam, the electron beam diffracts with the lattice at a certain place on the sample surface in different directions to form a Kikuchi line pattern. That is, the electron channeling patterns (ECP) mode [23]. Both the EBSD mode and ECP mode are used to form a Kikuchi line pattern by continuously changing the angle between the emission or incident electrons and the crystal orientation of the sample, and subsequently analyzing these Kikuchi line patterns to obtain the corresponding crystal structure information [24,25]. Therefore, the pixel position change of the acquired Kikuchi patterns in both EBSD and ECP modes corresponds to the momentum change of the electrons in the plane normal to the sample surface.

It is common sense that the raster scan mode of SEM is used to acquire information in the real space of the sample, while the ECP and EBSD modes of SEM are used to acquire information about the sample in the momentum. It is a fantastic conception that a sample could be investigated in both real space and momentum space from a single SEM image. Here we achieved this conception by a so-called cylindrical symmetric rotating crystal. In this sample, the crystal direction at the local position is continuously rotating and satisfies the cylindrical symmetry with respect to the central position. By using this sample, it is possible to observe both the contrast of the surface morphology and a complete Kikuchi line pattern from one SEM image measured in the raster scan mode. The contrast produced by the overlap of real and momentum space in this unique SEM image reflects both the particle and the wave nature of the electrons which interacted
with the sample surface. Subsequently, by analyzing the Kikuchi line pattern, the lattice constant, crystal orientation, strain properties, and Brillouin zone of this crystalline material could be obtained from one SEM image. Finally, there is an interesting correlation between the SEM images and the EBSD Kikuchi map of this unique cylindrically symmetric rotating crystal. This correlation is a practical example of the map-related Banach fixed-point theorems in mathematics.

Fig. 1 (a) Schematic diagram of the crystal plane rotation of a rotating crystal and the corresponding Kikuchi pattern in the sample region. (b) A cubic unit cell of bixbyite type $\text{In}_2\text{O}_3$. 
The bixbyite type $\text{In}_2\text{O}_3$ is a centrosymmetric cubic structure with the $\text{Ia}$\text{3} space group. Within each cell, there are 32 indium atoms (violet) and 48 oxygen atoms (red). The lattice constant is 10.29985 Å. (c) The observed SEM images of InSiO film detected by backscattered electrons, during the crystallization process of amorphous film at 300 °C. (d) The observed EBSD-normal direction (ND) images in the same region. (e) The simulated SEM images in the same region based on the dynamical method. (f) The cross section of the rotating crystal island for [111] direction that marked by dashed blue lines in (c), in which the central region is the rotating crystal island, and the neighboring two sides are the amorphous region. Enlarged cross sections of the crystal region are shown below. The (110) plane in InSiO is marked by red lines while the (100) plane in the sapphire substrate is marked by blue lines. The angles between (110) plane in InSiO and (100) plane in the sapphire substrate are also labeled out. The lattice constants measured through the TEM images are, 0.714 nm for InSiO and 0.435 nm for the sapphire substrate.

2. Results

2.1 Mechanism of Kikuchi pattern imaging in SEM

Since the Kikuchi pattern is theoretically formed by the continuous variation of the angle between the electron and the crystalline surface of the material, it is possible to observe the Kikuchi pattern from a so-called cylindrically symmetric rotating crystal sample in the raster mode of SEM. Fig. 1a illustrates schematically the cylindrically symmetric rotating crystal sample, in which the local crystal orientation of the crystal at different positions changes slowly and continuously, and the relative local crystal orientation refers to the central of this sample satisfying the cylindrical symmetry.
As can be seen in Fig. 1a, the [001] crystal orientation deflection gradually increases from its normal position in the center of the crystal (perpendicular to the film plane) as the circular crystallographic front gradually moves outward, as illustrated by the sketch of the [001] direction outlined in the cross section of the crystal in the same figure. In other words, during crystal growth, it is observed not only that the unit cell rotates uniformly along the cross-section that passes through the center point of the circular crystal film, but also that the unit cell (if traced along any radial outward direction of crystal growth) rotates permanently around an axis located in the plane of the film.

Therefore, in the conventional SEM mode, the electron beam is scanned perpendicular to the surface of the sample. The incident electrons diffract with the atomic lattice having different orientations at different landing positions, and when the sample satisfies the special rotational structure described above, it is possible to observe the Kikuchi pattern in the image of the scan of the whole sample.

2.2 Structure of the bixbyite type In$_2$O$_3$

In this work, the rotating crystal is prepared based on the bixbyite type In$_2$O$_3$ as shown in Fig. 1b.

2.3 Thin film preparation

Amorphous films with a thickness of 30 nm were prepared by DC magnetron sputtering (Shibaura Mechatronics, CFS-4EP-LL i-miller) on a sapphire substrate at room temperature. Sputtering targets consisting of In$_2$O$_3$ and SiO$_2$ were used. The ratio of Si/In in the sputtering target was 2.3
at. %, which corresponds to 1 wt. % in terms of SiO$_2$/(In$_2$O$_3$ + SiO$_2$). The sputtering target and the substrate were separated by 160 mm in the sputtering system. InSiO films were generated by a plasma at 200 W under a mixed atmosphere of argon and oxygen with a gas flow ratio of 1:1 between argon and oxygen and a total pressure of 0.25 Pa. This gas flow ratio was determined to produce electrically stable InSiO films that were resistant to thermal stress. [26-28] In-situ SEM observations were performed to obtain images of dynamic crystallization along a precisely fixed observation area. The InSiO films were heated in-situ to 300 degrees in the SEM environment. Fig. 1c shows typical results of SEM and EBSD observations of InSiO films after crystallographic heating.

2.4 Observed SEM images

Fig. 1c shows the crystallization of InSiO during the annealing process. The films grown on these substrates are partially crystallized, with a significant volume of material remaining amorphous. When the films are annealed at 300 °C, it can be observed that the amorphous films gradually crystallize and grow into round crystal islands with diameters of about 1-2 μm and form distinct Kikuchi patterns. These rotating crystal islands are circular structures that grow quasi-isotopically and radially from a single nucleation site.

It is reasonable to assume that the growth of these islands of rotating crystals begins with randomly distributed nucleation sites in the amorphous film, followed by two-dimensional growth from the center. In some cases, the nucleating site is isolated while in some cases the growth stops due to the presence of another crystalline site from a neighboring nucleus. This is a typical island nucleation and growth process in the thin film as reported in Ref. [29-32].
These crystalline islands all exhibit distinct Kikuchi pattern-like diffraction patterns in the SEM images. All the observed Kikuchi patterns belong to the cubic (bixbyite type) In$_2$O$_3$ structure. It can be further found that these crystalline islands exhibit different Kikuchi patterns in the SEM images, which implies that the crystallographic orientation is different in these crystalline islands. The observed Kikuchi pattern has several characteristic patterns, which suggest several preferred nucleation directions in the crystallization process, similar to those observed in previous studies [33].

For example, crystallization usually starts with the formation of a crystalline nucleus with the In$_2$O$_3$ structure and with the [440] direction approximately normal to the film plane. The preferred lattice orientation in In$_2$O$_3$ with the [440] orientation is easily revealed from the observed Kikuchi patterns, whose characteristic pattern is shown in Fig. 1b. This orientation is actually deduced from the Kikuchi pattern of In$_2$O$_3$ crystal, which allowed (using Kikuchi pattern spherical projection of In$_2$O$_3$) to assign indices to the Kikuchi band. The nuclei always show a strong curvature of the lattice, as is indicated by the small inter-contour distances between the Kikuchi band (hkl) and (-h-k-l), appearing always in pairs. Six major Kikuchi bands visible arise from the two sets of (-22-6)-type planes, two sets of (-22-2)-type planes, one (4-40)-type planes and one (00-4)-type planes, which can be seen in the SEM image shown in Fig. 1c. Different sets of Kikuchi bands process different band widths, and they intersect in a "pole", forming a bright complex hexagon. Similarly, the Kikuchi patterns for [400], [222] zone axis with different characteristics are also shown in the SEM image. The indexes for these bands are also labeled in the SEM image.

These Kikuchi patterns can be assigned to the rotating crystal with zone axes of [211], [222], [400], [440], and [622], where crystal planes as marked in Fig. 1c. We note that there is no Kikuchi pattern originating from SiO$_2$ structure, representing that at the presented rotating crystal region,
those Si atoms are soluble in the In$_2$O$_3$ matrix. This result is consistent with the crystallization information of InSiO film acquired with XRD [26], in which there was no XRD peak originating from crystalline SiO$_2$.

### 2.5 Observed EBSD images

According to Fig. 1d, the different colors in the EBSD-normal direction (ND) maps indicate the different orientations inside the rotating crystal islands. Within one island, orientation changes are visible. It can be observed that, in each rotational crystal island, there is obviously only one snowflake-like crystal “grain” without any significate crystal misorientation boundary. The image analysis shows that the central area of every rotational crystal island spherulites has a homogeneous orientation rotational velocity and, upon further growth, it branches into larger features with slightly different orientation rotational velocity, however, it should be noted that such rotational crystal is still a whole grain, which is completely different from the previously reported ones [33,34], in which the spherulites are spherical (or circular) structures that consist of fibers growing radially and quasi-isotopically from a single nucleation point.

The orientation of the different island centers varies within limited numbers of orientations, including [222], [440], [400], and [622] zone axes, which is consistent with the observation from our XRD experiment. With our experiment XRD map, the index of the lattice can be pointed out with our EBSD map, which is shown in Fig. 1c.

### 2.6 Simulated SEM images
This extremely special Kikuchi pattern observed in the SEM images is similar to the formation mechanism of the ECP Kikuchi pattern. They both originate from the angular variation in the BSE yield as a function of the target's crystal lattice orientation with respect to the incoming SEM electron beam. The difference is that in conventional ECP, the formation of the Kikuchi pattern is achieved by keeping the lattice direction constant and changing the direction of the incident electron beam, i.e., the rocking beam; while in presented SEM images of rotating crystal samples, the continuous change in the angle between the electrons incidence direction and the crystal is achieved by keeping the direction of the incident electron beam constant while rotating the lattice in the sample according to a specific rule.

The electron interaction with solid and its surface are studied with several theoretical approaches [35-48]. Among them, the most popular method is the Monte Carlo simulation method [49-57]. However, in Monte Carlo method, the jelly model is used to describe the dielectric properties of solid, which is powerful in amorphous material, however cannot include the lattice properties in crystals. In order to accurately describe the electron diffraction inside lattice for crystals, we developed a dynamical method [58,59] to obtain ECP patterns by simulating the diffraction processes of electrons in $\text{In}_2\text{O}_3$ single crystals with different incident directions to construct SEM patterns of rotating crystals as shown in Fig. 1e. We have simulated the reconstruction of the experimental SEM images based on the information of the crystal orientation distribution of the rotating crystal obtained from the EBSD-ND map (Fig. 1d). In these simulations, we neglected the fluctuation of the crystal rotation velocity in each crystal island and used the average rotation velocity, which is the uniform variation of the angle between the electron incidence direction and the lattice of the $\text{In}_2\text{O}_3$ single crystal, and we will report the detailed simulation method in a separate article.
The reconstructed simulated images reproduce the experimental SEM images well, and the number of Kikuchi bands in each crystal island, as well as their distribution, are consistent with the reconstructed images, which indicates that the Kikuchi patterns in these SEM images are indeed caused by crystal rotation. The Kikuchi patterns in the experimental SEM patterns, on the other hand, exhibit inhomogeneous bandwidths and distortions in shape, which are different from the simulated ones. In the center of the rotating crystal island, the Kikuchi bandwidth and its direction are closer to the simulated results; when at the edge of the rotating crystal island, the fluctuations in the Kikuchi bandwidth and its direction become significantly larger. This means that in the actual rotating crystal, the rotation speed of the rotating crystal is approximately the same in the central region; while it becomes different in the local position away from the center of the rotating crystal island. Accordingly, by analyzing factors such as the bandwidth of the Kikuchi band and the distortion, as well as comparing them with the simulated results, it is also possible to roughly estimate the rotation information of the crystal island.

2.7 TEM cross-sectional images

For further verification, the cross section profile of the presented rotating crystal island with (400) plane direction is demonstrated in fig 1f using TEM technique. It can be clearly seen that the lattice structure of Sapphire as a substrate, as well as the crystalline region, belongs to the lattice structure of cubic bixbyite In$_2$O$_3$.

In these TEM cross section images, the angle between the (110) plane in InSiO cross section (marked by the red dashed line), and the (100) plane in Sapphire (marked by the blue dashed line), varies with the observation location of the TEM images. The angle is 37 degrees when the
observation location of TEM images is at the leftmost edge of the rotating crystal island. As the observation location gradually moves to the right, the corresponding angle gradually increases at an approximately constant rate. This angle reaches 55 degrees when the observation location is moved to the rightmost edge of the rotating crystal island (1.019 μm from the leftmost edge). It is clear that the (110) crystal plane in the rotating crystal island rotates continuously along the direction of cross section as the observation location changes in the rotating crystal island with a rotation speed of roughly 17.67 degree/μm.

Additionally, the lattice constant of the rotating lattice island is determined by measuring the (110) lattice plane distance. The average lattice spacing of the (110) crystalline planes measured using these TEM cross-sectional images is, 0.711 nm. Thus, the lattice constant determined from this lattice spacing, by the equation $a = d_{\text{hkl}} \sqrt{h^2 + k^2 + l^2}$, is 10.055 Å for rotating crystal islands, which is almost identical to the value measured using the XRD technique (10.048 Å ± 0.018 Å).

However, the lattice constants of both measured rotating islands are slightly smaller than the original lattice constant of 10.094 Å ± 0.012 Å for polycrystalline In$_2$O$_3$ films prepared in the same way, indicating a 0.4% shrinkage of the unit lattice. Considering that the lattice parameters in the films are usually expanded due to tensile stresses. Therefore, the shrinkage observed here is caused by Si dopants. It is clear that although a small amount of Si does not significantly change the crystal structure of cubic bixbyite, the lattice parameters of the presented rotating crystalline InSiO island greatly differ from those of pure In$_2$O$_3$.

In the TEM cross-sectional images of the two edge regions of the rotating islands (Fig. 1f), the interface between the low-density amorphous InSiO and the high-density InSiO rotating crystal islands is observed. It can be clearly seen that this sub-interface, which can also be seen as the
crystallization front, located at the middle depth of the film, is clearly ahead of the crystallization front near the surface and the front near the substrate. It can be considered that during the crystallization process, when this front as a whole propagates in a direction parallel to the interface between the film sample and the substrate, a significant amount of densification by shrinking must occur along with the glass/crystal interface. For thin films, the increase in density will preferentially occur in the direction perpendicular to the free surface since the change in film shape is unrestricted only in this direction [60]. Once, the shrinkage rate along the highly diffuse crystalline front is slower than the propagation rate at the crystallographic interface. By using the analogy of heterogeneous epitaxial crystal growth, it is expected that a type of dislocation is formed on the crystalline side of this interface, with mismatches appearing periodically to compensate for the lattice parameter mismatch [61]. Due to the small interatomic distance (higher density) of the crystalline state, an additional half-plane appears on the crystalline side. According to the speculation in Ref. [34], these unpaired dislocations, which initially appear at the interface, remain in the growing crystal volume. They may act as the building-in geometrically necessary dislocation (GND), which generates continuous lattice bending and are responsible for the formation of rotating crystal structures.

Here, the near edge X-ray absorption fine structure (NEXAFS) measurement at the BL01B1 beamline of the SPring-8 synchrotron radiation facility is performed to verify this speculation. The resulting NEXAFS spectra of the completely annealed rotating crystal InSiO sample demonstrate that the octahedral structure of the InO$_6$ units, and tetrahedral SiO$_4$ units are preserved in the rotating crystal InSiO sample. The small number of tetrahedral SiO$_4$ units in the InO$_6$ octahedral network presumably causes local distortion in the bixbyite In$_2$O$_3$ matrix. These local distortions,
in turn, become the GNDs necessary to rotate the InSiO thin-film crystal, which makes the InSiO film internally bent of the crystal lattice planes itself around an axis lying in the film plane.

One thing should be noticed that there is a similar phenomenon found earlier for crystal growth in amorphous films of different substances [33,34,60-63], in which the local crystal orientation rotates uniformly along the cross-section that passes through the center point of the circular crystal film, therefore, they are referred to as transrotational crystals. However, here the presented InSiO thin-film crystal not only possess continuous rotation in the local crystal direction but also the relative local crystal orientation refers to the central of this sample satisfying the cylindrical symmetry. Another thing that should be noted is that the observed boundary of the interface propagation differs slightly from the assumptions made in previous reports [34,60]. In these previous reports, they assumed that the crystallization front near the surface leads to the crystallization front near the substrate. Here, the observed crystallization front near the surface and the crystallization front near the substrate both lag behind the crystallization front in the medium depth region of the thin InSiO film. This difference, potentially, leads to this rotating crystal, where the crystalline surface rotates not only along the radius direction as shown in the TEM cross section images, but also around the center of the circle, resulting in a circularly symmetric pattern that makes the observed SEM pattern appear as a Kikuchi pattern.
Fig. 2 (a), Top: The BSE image measured from rotating crystal island whose central region near [440] zone axis with sample stage tilt angle for (-5, -3, -1, +1, +3, +5) degrees. (b), The simulated ECP patterns of In$_2$O$_3$ single crystals in the [440] zone axis direction with sample stage tilt angle for (-5, -3, -1, +1, +3, +5) degrees. (c), The measured deviation of [440] zone axis position, i.e. brightest central complex polygon in SEM images, as the function of sample stage tilt angles.

2.8 SEM images when tilting sample stage
Whether the experimentally observed Kikuchi pattern originates from electron diffraction can also be verified by observing the movement of the pattern when the sample stage is tilted. Fig. 2a shows SEM images of a rotating crystal island whose central region is near the [440] zone axis as the sample stage tilted at an angle from -5° to 5°. When focusing on the brightest central complex polygon (the intersecting region of multiple Kikuchi bands) in the SEM image, which is the most striking feature, it can be noticed that the brightest central polygon moves with the tilt of the sample stage. Using -5 degrees as the origin, it can be observed that the brightest central polygon shifts downward whenever the sample stage angle is changed.

Fig. 2b shows the ECP simulations of In$_2$O$_3$ single crystals in the [440] zone axis direction for the corresponding tilt angles of the sample stage. It can be seen that the simulated Kikuchi pattern of In$_2$O$_3$ in the [440] zone axis direction, spanning an angle of 30 degrees, has a similar Kikuchi band width to the experimental SEM pattern, as well as a similar size of the brightest central polygonal region. This represents that, the local crystal orientation of this presented rotating crystal island spans a rotation of about 30 degrees from one side to the other. In addition, the movement of the brightest central polygon in the simulated Kikuchi pattern, also moves downward as the changes of the tilting angle of the sample stage. The distance of the movement of the brightest central polygon with different tilt angles of the sample stage is generally consistent with the experimentally observed ones.

The rotation velocity of the crystal plane in the rotating island can also be estimated by measuring the offset distance of the brightest central polygon in these SEM images against the tilt angle of the sample stage. Fig. 2c demonstrates that the brightest central polygonal region in the SEM image shifts in distance as the sample stage tilt angle is changed, where the distance is measured with the
sample stage tilt angle at -5 degrees as the origin. According to the linear fit of these data, the reciprocal of the slope of the fitted line describes the average rotational velocity of the crystal plane in this rotating crystal island, which is about 20°/μm. It should be noted that the Kikuchi patterns in the SEM image shift downward by about 50 nm per degree. Such a large shift distance suggests that the contrast of the Kikuchi pattern observed in these SEM images must be diffraction contrast due to electron crystal diffraction rather than surface topography contrast or bulk chemical composition contrast caused by the growth of crystals in real space, such as those observed SEM images from some special crystals that grow radial filamentary structures [64]. Knowing that the thickness of the presented rotating crystals is only 30 nm, any contrast due to crystal growth in real space should shift to less than 0.53 nm (30 nm * tan1°) per degree when the angle of the sample stage changes.
Fig. 3 (a), The BSE image measured from rotating crystal island whose central region near [440] zone axis with incident electron energy of 3 keV, 5 keV, 10 keV, 15 keV, 20 keV, 25 keV, 30 keV. (b), The simulated ECP patterns of In$_2$O$_3$ single crystals in the [440] zone axis direction with the electron incident energies of 3 keV, 5 keV, 10 keV, 15 keV, 20 keV, 25 keV, 30 keV. (c), The relationship between the measured and simulated bandwidth of the ($\mp 4$, $\pm 4$, 0) Kikuchi band as the function of incident electron wavelength. The horizontal axis is the incident electron energy and the corresponding wavelength, the vertical axis is the measured bandwidth of the ($\mp 4$, $\pm 4$, 0) Kikuchi band, and the corresponding misorientation of the rotating crystal under the Kikuchi band range, where the rotational velocity of the crystal plane is set to 20°/μm.
2.9 SEM images for different incident energy

Fig. 3a gives the SEM images of the rotating crystal island whose central region is near the [440] zone axis at different incident electron voltages to further investigate the electron energy dependence of the observed Kikuchi patterns. It can be found that with the increase of the incident electron energy in the range from 3 keV to 30 keV, the width of all observed Kikuchi bands in these SEM images, becomes progressively narrower, and the size of the brightest central hexagonal region consisting of the intersection of these Kikuchi bands also shrink. This result is strong evidence that the Kikuchi pattern observed in these SEM images originates from the diffraction interaction of electrons with the lattice in the rotating crystal sample.

Fig. 3b shows the simulated ECP patterns in the [440] zone axis direction of the In$_2$O$_3$ single crystal with different incident electron energies. It can be found that the widths of the Kikuchi bands in the simulated Kikuchi patterns obtained with different incident electron beam energies, as well as the size of the brightest central polygonal region, are roughly consistent with the experimentally observed Kikuchi patterns. This indicates that the rotating crystal island shown in the SEM image, with a diameter of about 1.64 μm, has a rotation span of about 30 degrees along the diameter direction under its pattern.

In the Kikuchi pattern, the angular width of the Kikuchi band is equal to twice its corresponding Bragg angle. According to Bragg's law, $\lambda = 2d \sin \theta_B$, the angular width of the Kikuchi band at small-angle approximation is $2\theta_B \approx \lambda/d$, where the $\theta_B$ is the Bragg angle, $\lambda$ is the wavelength of the incident electrons and the $d$ is the crystal plane spacing.
Since the sample measured here is a special type of rotating crystal, its internal crystal planes rotate continuously at an approximately equal rotational velocity in any direction. Therefore, by measuring the bandwidth of the Kikuchi band in the SEM image, the span of rotation of the crystal direction in the rotating crystal film, under the coverage of this Kikuchi band, can be determined by the following expression, \( 2\theta_B = l d\Phi/dx \), where \( l \) is the bandwidth measured in the SEM image and \( d\Phi/dx \) is the rotation velocity of the rotating crystal. If we set the rotation velocity of the rotating crystal as \( 20^\circ/\mu\text{m} \) according to Fig. 2, the simulated angular width \( 2\theta_B \) of the Kikuchi band and the measured bandwidth \( l \) in the Kikuchi band could be plotted in the same figure as shown in Fig. 3c.

Fig. 3c shows the bandwidth of measured the \( (\mp 4, \pm 4, 0) \) Kikuchi band observed in the SEM images and the angular width \( 2\theta_B \) of the simulated \( (\mp 4, \pm 4, 0) \) Kikuchi band as the function of the wavelength of the incident electrons, as well as the corresponding energy of incident electrons. It is clear that there two data sets agree well with each other, which indicates the correctness of setting the rotation velocity of the rotating crystal as \( 20^\circ/\mu\text{m} \). Furthermore, linear variation in both the measured bandwidth and simulated angular width is observed in Fig. 3c.

In addition, substituting the correlation between the Bragg angle and the lattice plane spacing \( d \) and the wavelength \( \lambda \) of the incident electrons, \( d = \lambda / 2\theta_B \), the lattice plane spacing can be further determined by the expression, \( d = \lambda / (l d\Phi/dx) \). Thus, with the electron wavelength as the x-axis and the angle spanned by the rotation of its rotating crystal lattice plane under the cover of the Kikuchi bandwidth as the y-axis, i.e., \( 2\theta_B \), the lattice plane spacing is the reciprocal of the slope of the line to the measured bandwidth data were linearly fitted. According to those measured bandwidths of the Kikuchi band, the average lattice plane spacing of the \( (\mp 4, \pm 4, 0) \) plane is about 1.91 Å. Also, because the lattice constant \( a \), which can be determined by \( a = d_{hkl} \sqrt{h^2 + k^2 + l^2} \). The lattice
constant \(a\) of the presented rotating crystal island is further calculated as 10.80 Å, which is similar to the value of the lattice constant 10.05 Å measured from the TEM cross section image (Fig. 1f).

Fig. 4 The SEM image of the rotating crystal islands whose central region is (a) [222], (b) [400], and (c) [440] crystal direction, obtained by backscattered electron (BSE) detector, secondary electron (SE) detector with incident electron energy of 15 keV, and corresponding simulated SEM images by the dynamical method. Some separated out In\(_2\)O\(_3\) nanocrystal from the InSiO surface are marked by small blue arrows. The Miller indexes are labeled on the simulated Kikuchi pattern. The red lines indicate the projections of lattice planes (hkl) shown by labels. The size of the
Brillouin zone (inner bright hexagon) in the projection plane for the rotating crystal island with [440] direction is demonstrated in both measured SEM images and simulated images.

2.10 Kikuchi patterns with a center of symmetry

Since that we have determined that the Kikuchi pattern observed from the rotational crystal sample in SE images should be regarded as diffraction contrast which origin from the diffraction effect between the incident electron and the atomic lattice of the sample. It is interesting to simultaneously observe the diffraction contrast and also the morphology contrast in the SEM image at the same time. Here, the InSiO film is first annealed in air to 250 °C and then transfer to the SEM chamber for further annealing to 300 °C. After that, a large number of small bulbous protrusions whose radius is about 20-30 nm could be observed in the SEM images which is the In$_2$O$_3$ nanocrystal separated out from the InSiO surface because of the interaction of amorphous In$_2$O$_3$ with water gas in air. These In$_2$O$_3$ nanocrystals on the InSiO film surface, provide us with clear morphology contrast in both the BSE and SE images.

Fig. 4 shows the BSE and SE image of three selected rotational crystal islands whose central region is the [222], [400], and [440] crystal direction respectively. It is clear that the characteristic Kikuchi pattern observed in both BSE and SE images originates from Bragg reflection from sets of lattice planes oriented perpendicular to the surface. In the rotating crystal island with [222] direction (fig. 4a), three major Kikuchi bands visible arise from the one set of (0-44)-type planes. As for the rotating crystal island with [400] direction (fig. 4b), four major Kikuchi bands visible arise from the two sets of (004)-type planes, two sets of (0-44)-type planes. As for the rotating crystal island with [440] direction (fig. 4c), six major Kikuchi bands visible arise from the two sets of (-22-6)-
type planes, two sets of (-22-2)-type planes, one (4-40)-type planes and one (00-4)-type planes. In each rotating crystal island, different sets of Kikuchi bands process different bandwidths, and they intersect in a "pole", forming a bright polygon with filigree internal structures, respectively. The shape of the bright intersected region for the rotating crystal island with [222] direction, and [400] direction, are quite clear, while that for the rotating crystal island with [440] direction is relatively complex. The bright intersected region for the rotating crystal island with [440] direction is a complex polygon that is composed of a bright central hexagon being the zone axis, i.e. the intersection region of (004) Kikuchi band, (-22-6) Kikuchi bands and (-22-6) Kikuchi bands, as well as six adjacent triangles being the intersection of two of these three bands. The sets of blurry lines originated from the higher order reflection of the (-440) plane, and flanks those major Kikuchi bands, could be observed in the rotating crystal island with [440] direction as a rounded bright region with a clear dark outline. The intersections between these blurry lines and those major Kikuchi bands, form some brighter nodes in those major Kikuchi bands.

It is clear that the highly-symmetrized Kikuchi pattern can be observed in both SE and BSE images, they have almost the same features, while the contrast of SE image is significantly lower than that of BSE image. This result implies that the diffraction information observed in SE images is not coming from the interaction when the SE is transported inside the crystal. It is coming from the cascade SE produced by the backscattered electrons yield modulated by the incident high energy electron interacting with the crystal lattice. Therefore, the quantities for SE emitted from the different regions of the crystal surface are influenced by the local crystal direction where primary electrons land on different regions of this rotational crystal.

Compared to the simulated Kikuchi pattern, it is clear that the simulated Kikuchi pattern agrees well with the experimentally observed Kikuchi pattern in the presented BSE and SE images. In
fact, besides the bandwidths of different types of Kikuchi bands and their angles among them, even the fine features in the intersection region of the Kikuchi bands, i.e. the bright complex polygons with filigree internal structures, as well as the rounded bright region and its dark outline are well reproduced by the simulated Kikuchi pattern.

Another thing that should be noticed is that the major Kikuchi bands observed in the rotating crystal island with the same direction, their widths all remain relatively consistent, even when they are far from the center of the rotating crystal island edges, except for the (-404) plane in rotational crystal island with [222] zone axis because of the distorted lattice as shown in Fig. 4a. This represents that the presented rotating crystal island has a highly consistent rotation velocity. This allows us to further analyze the Kikuchi pattern in the SEM image, to obtain the useful physical quantities related to the crystal structure, which is generally not able to be obtained from the SEM image of a conventional sample.

2.11 Measure the lattice constant

According to the previous analysis, for the rotating crystal sample, twice the Bragg angle, could be determined by multiplying the rotation speed of the rotating crystal by the bandwidth of the Kikuchi band in the SEM images, as $2\theta_B = l \cdot d\Phi / dx$, therefore lattice plane spacing could be determined by the expression, $d = \lambda / (l \cdot d\Phi / dx)$. In the presented SEM images, the width of bands for each plane direction in these rotating crystal islands can be measured as listed in Table 1 and the calculated corresponding lattice plane spacings are also listed in Table 1.
Table.1. The plane index, Bragg angle, bandwidth, plane spacing, reciprocal lattice vectors, lattice constant, strain, Burgers vector and building-in geometrically necessary dislocations (GNDs) density in the observed rotating crystal island with the zone axis of [222], [400], and [440].

Thus through the equation \( a = d_{:\text{hkl}} \sqrt{h^2 + k^2 + l^2} \), the lattice constant, \( a \), determined from the lattice plane spacing are also listed in Table 1. The average value of the lattice constant obtained on the rotating crystal islands with zone axis of [222], [400] and [440] are 8.49 Å, 13.03 Å ± 2.11 Å, 11.61 Å ± 2.84 Å, respectively. And the total average value of the lattice constant is 11.57 Å ± 2.65 Å. These valued are slightly larger than that obtained from the analysis of the variation of the

| Zone Axis | h  | k  | l  | Bragg angle (degree) | Bandwidth (nm) | Plane spacing(Å) | Reciprocal lattice vectors(Å⁻¹) | Lattice constants(Å) | Strain (%) | Burgers vector(Å) | GND density(10¹⁴ m⁻²) |
|-----------|----|----|----|----------------------|-----------------|------------------|----------------------------------|---------------------|------------|------------------|----------------------|
| [222]     | -4 | 4  | 0  | 1.91                 | 190.6           | 1.5              | 4.19                             | 8.49                | 0.52       | 24.00            | 14.57                |
| [400]     | 0  | -4 | 4  | 1.40                 | 140             | 2.04             | 3.08                             | 11.54               | 0.52       | 32.64            | 10.73                |
| [400]     | 0  | -4 | 0  | 0.79                 | 78.8            | 3.63             | 1.73                             | 14.52               | 0.52       | 29.04            | 12.04                |
| [440]     | 0  | 0  | 4  | 1.06                 | 105.5 9         | 2.71             | 2.32                             | 10.83               | 0.52       | 21.67            | 16.14                |
| [440]     | -2 | 2  | -6 | 1.66                 | 166.1 5         | 1.72             | 3.65                             | 11.42               | 0.52       | 37.87            | 9.23                 |
| [440]     | -2 | 2  | -2 | 1.14                 | 113.8 9         | 2.51             | 2.50                             | 8.70                | 0.52       | 15.07            | 23.21                |
| [440]     | -4 | 4  | 0  | 1.05                 | 104.3 7         | 2.74             | 2.29                             | 15.50               | 0.52       | 43.84            | 7.97                 |
width of bands in the SEM image of the rotating crystal island with different electron incident energies.

2.12 Mark two-dimensional Brillouin zone

Based on the determined lattice plane spacing $d$, the lengths $|g_{hk'l}|$ of the reciprocal lattice vectors could as be calculated by the formula, $g_{hk'l} = \frac{2\pi}{d}$. Therefore, the reversed reciprocal lattice vectors for different planes in three different crystals can be further calculated as listed in Table 1. In fact, according to the special relationship between the bandwidth of Kikuchi band, $l$, and the Bragg angle, $\theta_B$, in the presented rotating crystal sample, i.e. $2\theta_B = l \cdot \frac{d\Phi}{dx}$, the the lengths $|g_{hk'l}|$ of the reciprocal lattice vectors could be rewritten as

$$g_{hk'l} = \frac{2\pi}{d} = \frac{l \cdot \frac{d\Phi}{dx}}{2\pi \lambda},$$

where, the $d\Phi/dx$ is the rotation speed of the rotating crystal, and the $\lambda$ is the wavelength of the electron. Since the $d\Phi/dx$ is 20 degree/μm, and $\lambda$ is 10 pm for electrons with 15 keV, the reversed reciprocal lattice vector, $g_{hk'l}$ is only dependent on the the bandwidth of Kikuchi band, $l$, measured in the SEM images. So in the SEM image of the rotating crystal sample, each pixel is corresponding to both the different spatial positions on the sample surface according to real-space coordinate and also the different angle between the electron beam and a particular crystal surface according to reciprocal-space coordinate, i.e. on a momentum scale. And the transformation coefficient from the spatial scale to the momentum scale is, $\frac{d\Phi}{dx} \cdot \frac{1}{2\pi \lambda}$. It is interesting to note the momentum scale of the presented SEM image of the rotating crystal sample at a given incident electron energy. For the incident electron energy of 15 keV, the transformation coefficient from
the spatial scale to the momentum scale is, $0.0022 \text{ Å}^{-2}$. Therefore, the presented SEM images are composed of 512 by 512 pixels, every pixel corresponds to 20.8 Å, 24.1 Å, 23.2 Å, in the real space coordinate, and 0.046 Å$^{-1}$, 0.053 Å$^{-1}$, 0.051 Å$^{-1}$ in the momentum coordinate at the same time in the BSE and SE images of three rotational crystal islands at the zone axis of [222], [400], [440], respectively. This property makes it possible to determine the size of the two-dimensional Brillouin zone in the projection plane in momentum scale whose outline could be determined by extending prominent Kikuchi bands. Here, the side length of the two-dimensional Brillouin zone in the projection plane for the [440] crystal is marked in the BSE and SE images as shown in Fig. 4c. The conventional SEM adopted for this special rotating crystal sample can be regarded as a momentum-microscopy similar to those reported in Ref. [65].

2.13 Measure the strain value

A simple estimation of the strain value in the rotating crystal is possible from the SEM image of this rotating crystal island, by assuming an elastic cylindrical bending of the rotating crystal in the direction of radius. The maximum strain $\varepsilon$ of the surface film could be estimated according to Ref. [62]:

$$\varepsilon = \frac{t \theta_B}{l} = \frac{t d\Phi/dx}{2}$$

where $\theta_B$ is the Bragg angle, $l$ is the bandwidth of the Kikuchi band, $t$ is the thickness of the rotating crystal film, and $d\Phi/dx$ is the average measured gradient of monotonously increasing misorientation angle, for a rotational crystal that could be expressed in the ratio of the angle spanned by the rotating crystal of the Kikuchi band to its bandwidth as $d\Phi/dx = 2\theta_B/l$. Considering
the thickness of the presented film is 30 nm, and the dΦ/dx is 20 degree/μm determined according to Fig. 2c, the strain values calculated from each plane in three rotating crystal islands are consistent as listed in Table 1. The presented strain value of rotating crystal islands is 0.52%, which is a relatively small value. According to [66], the microstrains of In$_2$O$_3$ NPs for different calcination temperatures (200, 300, 400, 500 and 600 °C) were 0.0736, 0.0944, 0.1144, 0.1500 and 0.1647, respectively. Our resulting strain value is far lower than the measured microstrain of In$_2$O$_3$ nanoparticles, so at the annealing procedure of the crystal, the inter force did not exceed the maximum strain value, thus the crystal can rotate homogeneously without breaking, finally forming the rotational crystal different from the normal crystals.

2.14 Measure the GNDs density

Furthermore, if we assume that the lattice rotation in the presented rotating crystal is contributed by the building-in geometrically necessary dislocation (GNDs) which accommodate a lattice curvature from a deformation gradient [67], we could even roughly estimate the lower bound of the GNDs density from the SEM image of the rotating crystal.

If one assumes a homogeneous distribution of GNDs in the rotating crystal, a lower bound estimation of the density of GNDs can be done by [68],

$$\rho_{\text{hom}} = \frac{\theta_{\text{tot}}}{|b| \Delta x},$$

where $\theta_{\text{tot}}$ is the lattice bending angle over distance $\Delta x$, realized by means of dislocations with Burger’s vector $b$ [69]. For the presented rotating crystal, therefore, the $\theta_{\text{tot}}/\Delta x$ can be expressed as
the ratio of the angle spanned by the rotating crystal of the Kikuchi band to its bandwidth, \(2\theta_B/l\).

Therefore, the formula could be rewritten as,

\[
\rho_{\text{hom}} = \frac{2\theta_B}{|b|l} = \frac{d\Phi/dx}{|b|},
\]

Substituting our experimental values of \(d\Phi/dx\) that taken from the SEM image, and \(|b|\) being the size of the crystal axis along each plane direction, \(|b| = \frac{a}{2}\sqrt{h^2 + k^2 + l^2}\) for bcc crystal as listed in Table 1. The GND densities calculated from each plane in three rotating crystal islands are listed in Table 1. The average value of the GNDs density obtained on the rotating crystal islands with zone axis of [222], [400], and [440] are \(1.46\times10^{15} \text{ m}^{-2}\), \(1.14\times10^{15} \text{ m}^{-2}\), \(1.41\times10^{15} \text{ m}^{-2}\), respectively. Thus the total averaged GND density is \(1.34\times10^{15} \text{ m}^{-2}\). Such a large dislocation density has also been observed in nanostructured In\(_2\)O\(_3\) thin film. For example in Ref. [70], it is found that the dislocation density of In\(_2\)O\(_3\) thin film is \(4.25\times10^{15} \text{ m}^{-2}\) when the substrate temperature is about 350 °C, it could be dropped to \(1\times10^{15} \text{ m}^{-2}\) when the substrate temperature reaches 500 °C.
Fig. 5 (a), SEM and EBSD measurement of rotating crystal island whose central region near [440] zone axis. From top left to bottom right are BSE image, EBSD map in the normal direction (EBSD-ND), EBSD image quality map (EBSD-Q), EBSD misorientation map (EBSD-M), EBSD relative misorientation-angle map (EBSD-angle), EBSD relative misorientation-axis map (EBSD-axis), respectively. (b), Top: The schematic diagram of crystal direction rotation in an ideal rotating crystal island in the presented work, where the direction of the red arrow represents the direction of any crystal plane. Middle: Crystal misorientation angle profiles measured along the white circle around the center of the rotational crystallization island. The reference line for the theoretical calculation of point-origin misorientation around an ideal circular symmetry rotating crystal is shown as the blue dashed curve, which is determined by

\[
\alpha = \arccos \frac{\vec{r}_{\text{origin}} \cdot \vec{r}_{\text{point}}}{|\vec{r}_{\text{origin}}| |\vec{r}_{\text{point}}|} = \arccos\left(\sin^2 \alpha_0 \cos \theta + \cos^2 \alpha_0\right),
\]

where the \( \alpha_0 \) is 4.86 degree measured from EBSD measurement. The inset is the sketch for calculating the corresponding point-origin
misorientation from an ideal circular symmetry rotating crystal. Bottom: Crystal misorientation angle profiles measured along the white arrow from the center to the edge of the rotational crystallization island. (c), Top: The schematic diagram of crystal direction rotation in previous observed rotating crystal island [34]. Middle: Crystal misorientation angle profiles measured along a circle around the center of the rotational crystallization island from Fig. 1 in Ref. [34]. Bottom: Crystal misorientation angle profiles measured along a radius from the center to the edge of the rotational crystallization island.

3. Discussion

3.1 Misorientation in rotating crystal island

Fig. 5a shows SEM and EBSD measurements of rotating crystal islands. The BSE images of the rotating crystal island show clear Kikuchi patterns, which indicate the central region of the presented rotating crystal island should be near the [440] zone axis. This is further confirmed by EBSD measurement. However, the EBSD map in the normal direction ND (EBSD-ND) is not homogeneous. Slightly various colors in this map indicate the slightly various misorientations of the local region of the rotating crystal island referring to the [440] crystal direction. In this case, the relative misorientation of the angle distribution (EBSD-angle), and the relative misorientation of the axis distribution (EBSD-axis), are plotted in Fig. 5a, respectively.

In the EBSD-angle map, different colors represent the relative out-plane direction between the local orientation and the average crystal direction of the whole crystallization region. From the EBSD-angle map, it is found that the angle of the central region in the rotating crystal island almost overlaps with the average crystal direction of the whole crystallization region, which implies 2D
spherulitic crystal growth of the crystal. Furthermore, the relative misorientation angle change from 0° to 14° from the center to the edge and forms a roughly circular symmetry distribution. This circular symmetry distribution implies that the relative misorientation angle only increases at a constant rate with the distance from the central region.

In the EBSD-axis map, different colors represent the relative in-plane direction between the local orientation and the averaged crystal direction of the island. The presented EBSD-axis map shows a significant 180° rotational symmetry pattern, which implies that the local crystal direction along any circle around the central region gradually and uniformly rotated by 360°. Furthermore, in this EBSD-angle map, any diameter of the rotating crystal island has approximately the same color. This implies that the in-plane relative misorientation between the local orientation and average orientation of the whole crystallization region is approximately the same along each diameter.

Based on the EBSD-angle map and EBSD-axis map, it is clear that in the presented rotating crystal island the crystal plane not only rotates uniformly along any radial outward direction of crystal growth, but also rotates permanently around an axis located in the center of the rotating crystal island. This is even clearer in the EBSD misorientation map shown in Fig. 5a. It can be seen that the misorientation is almost the same on this rotating crystal island, which is about 2 degrees. This means that the difference between the crystal orientation of any pixel on this rotating crystal island and the crystal orientation of its surrounding neighboring pixels, is almost equal. In other words, this disk-shaped rotating crystal island, with crystal orientations at any local position, along any adjacent direction, is continuously rotating at an almost uniform velocity.

EBSD-Q plots are also presented in Fig. 5a. This snowflake shaped crystal quality distribution indicates that the rotation of the crystal orientation in the rotating crystal sample originates from the density difference between the amorphous and crystalline InSiO films during the crystalline
process. According to the EBSD-Q map, it can be seen that the central region of the rotating crystal island has a relatively uniform crystal quality, which branches into larger features of decreasing crystal quality after further growth and forms finger-like patterns at the front end of its growth. It is clear that at a certain distance from the core, more anisotropic growths appear that clear fiber structure with finger patterns at their growth fronts. Such growth usually can be realized by two different mechanisms. The first one is crystallographic branching [71] when one primary fiber grows from the nucleation center without the formation of small angle grain boundaries (< 3°). The other one is the so-called non-crystallographic branching [72-75], when new sub-crystals (secondary fibers) with a small misorientation (> 3°) heterogeneously nucleate on the side of an already growing fiber and grow further as new fibers, also mostly in radial directions. During crystal formation, the above two effects are presented, it holds the possibility to grow into a rotating crystal. If the crystallographic branching effect dominates, it grows into radial spherical crystals with finger patterns and leads to a snowflake-like crystal quality distribution, as observed in the EBSD-Q map. Conversely, if the non-crystallographic branching effect dominates it grows into radial spherical crystals grow as radial spherical crystals with many slender fibers grow, as reported in the previous observation in Se [76-78], Fe₂O₃ [77-79], and V₂O₃ [80] fibers in spherulites, as well as for Cr₂O₃ [80], V₂O₃ [80], Ta₂O₅ [81], Ge-Te, Tl-Se, Cd-Te alloys [82], and single crystals Cu-Te alloys [83], among others [84].

The top of Fig 5b and 5c shows the local crystal rotation distribution for an ideal rotating crystal in this paper and the previously reported rotating crystals, respectively. The presented rotating crystalline film forms a special crystallization region as a whole in the shape of a disk with different local crystal orientations at any position because only the crystallographic branching effect exists in the crystallization process. However, in this disk-shaped crystallization region, the relative local
crystal orientation with reference to the crystal orientation at the crystal center position not only rotates in the radial direction of crystal growth, but also permanently rotates around the axis located at the crystal center position with the same rotational velocity. In contrast, in the previously reported rotating crystal films, the non-crystallographic branching effect dominates the crystallization process during growth, thus forming a crystallization region consisting of fibers. Its local crystal orientation changes only gradually with the growth of the fibers, i.e., it rotates continuously in the radial direction of growth. Its local crystal orientation changes only gradually with the growth of the fibers, i.e., it rotates continuously in the radial direction of growth. Due to the difference in crystal orientation between fibers, the local crystal orientation does not undergo any coherent rotational behavior in the vertical direction of the radial direction.

The middle part of Fig. 5b and 5c show the lattice misorientation of the presented rotating crystal and the rotating crystal previously reported in reference [34], respectively, running at one radius along the white arrow, from the center to the edge. It can be seen that the crystal misorientation curves for these two different types of rotating crystals show similar results, i.e. obvious linear behavior. From Fig. 5b and 5c, the gradient of the crystal rotation angle, i.e., the crystal rotation velocity along this selected radius can be obtained by a linear fitting. The crystal direction rotation velocity is 17.6 degree/μm for the presented rotating crystal and 0.74 degree/μm for the rotating crystal in Ref. [34]. Such a large difference arises from the widely different sizes of the two rotating crystals. The diameter of the presented rotating crystal island is about 1.6 μm, while the diameter of the rotating crystal island in Ref. [34] is about 70 μm. It should be noted that for the presented rotating crystal island, its rotation velocity determined by the EBSD technique roughly agrees with that determined from the analysis of the SEM image of the Kikuchi pattern with different tilt angles of the sample stage.
The bottom of Fig. 5b and 5c show the crystal lattice misorientation profile along the white circle around the crystal center for the presented rotating crystal and the previously reported rotating crystal in Ref. [34], respectively. It is obvious that the point-to-point misorientation curves for the presented rotating crystal and the previously reported rotating crystal are completely different. As for the presented rotating crystal, the point-to-point misorientation curve is a slightly fluctuating horizontal line close to 1 degree, and its maximum fluctuation is 2 degrees. Considering the overall accumulated misorientation for a vector of continuous rotation around the axis is 360° for one circle, the average value of 1 degree of the point-to-point misorientation curve in the range of 0-360°, indicating that the misorientation observed along the white circle is solely contributed by local crystal direction rotation at an approximately constant velocity. In order words, the whole region in the presented rotating crystal island is composed of one primary fiber growth from the nucleation center in which the crystal orientation is rotated without the formation of small angle grain boundaries.

As for the previously reported rotating crystal, the point-to-point misorientation curve is constantly and dramatically fluctuating, with an average value of 3.42 degrees and a maximum misorientation of even 22 degrees. This means that the previously reported rotating crystal are consisted of fibers growing radially from a single nucleation point, therefore, there is no continuous crystal direction rotation occurring along the white circle, but only local crystal direction rotation occurring at varying velocities inside each fiber. The dramatic rise and fall observed in the point-to-point misorientation curve are contributed by the misorientation between each neighboring fiber varies intensely and randomly.

In order to show how the local crystal orientation rotates along the white circle in the presented rotating crystal, the point-origin misorientation curve of the presented rotating crystal is shown in
Fig. 5b. The point-origin misorientation formed by a vector rotating continuously around the axis is also plotted as a reference curve in Fig. 5b and can be obtained by the following equation,

\[
\vec{r}_{\text{origin}} = (r, 0, \frac{r}{\tan \alpha_0}); \vec{r}_{\text{point}} = (r \cos \theta, r \sin \theta, \frac{r}{\tan \alpha_0})
\]

\[
\alpha = \arccos \left( \frac{\vec{r}_{\text{origin}} \cdot \vec{r}_{\text{point}}}{|\vec{r}_{\text{origin}}| \cdot |\vec{r}_{\text{point}}|} \right) = \arccos \left( \frac{\sin^2 \alpha_0 \cos \theta + \cos^2 \alpha_0}{r} \right)
\]

where \(\alpha_0\) is the mean misorientation referred to the rotation axis at the radius of \(r\), \(\theta\) is the azimuth on the plane perpendicular to the rotation axis. The angle \(\alpha_0\) between the vector and the rotation axis is 4.86° measured from the EBSD-angle map. This reference point-origin misorientation curve could be regarded as the point-origin misorientation curve measured from an ideal circular symmetry rotating crystal, as shown in Fig. 5a.

It can be seen that the point-origin misorientation curve for the presented rotating crystal is consistent with that determined from a vector rotating continuously around the axis. In other words, the point-origin misorientation curve for the presented rotating crystal is consistent with that for the ideal circular symmetry rotating crystal. In fact, in the presented rotating crystal, the local crystal orientation rotates under the same law along a circular trajectory with any radius, once the center of the circular trajectory overlaps with the center of the presented rotating crystal. Therefore, the relative local crystal direction, using the rotating crystal center direction as a reference, satisfies the circular symmetry throughout the presented rotating crystal region. And this extra circular symmetry is the essential difference between the presented rotating crystal and the previously reported rotating crystal, which is also the essential reason for the observation of the Kikuchi pattern in the SEM images of the presented rotating crystal.
Fig. 6 (a). The geometry of the EBSD measurement. The central one of the Kikuchi pattern distribution matrix region was projected on a planar screen to a perpendicular surface. The Kikuchi pattern of the emitted electrons, which are within ±15° of the surface normal of the sample, is coiled in the planar screen. (b) EBSD Kikuchi pattern matrix map observed in charge-coupled device (CCD) camera in EBSD measurement at intervals of 200 nm from the rotating crystal island sample, which is formed by electrons escaped within ±15° along the surface normal direction. Enlarged Kikuchi pattern region in the CCD image at the central region of the rotating crystal island. The rotating crystal island is marked by a dashed curve. (c) The measured BSE image of InSiO rotational crystal islands. The contrast of the image is adjusted to highlight the fine structures of the Kikuchi pattern.

3.2 Banach fixed-point theorems in Kikuchi patterns

As shown in Fig. 6a, in the geometry of the EBSD measurement, the specimen is largely tilted (about 70°) from the horizontal plane, therefore, those electrons escaping from the specimen along the surface normal form a corresponding Kikuchi pattern. This pattern appears on the upper side of the center of the Charge-coupled device (CCD) image, about 50 degrees (assuming, the center of the CCD camera is on the same level as the center of the sample). Fig. 6b shows the EBSD
Kikuchi pattern matrix maps observed in a selected region of CCD camera in the EBSD measurement at different electron landing positions in the rotating crystal sample, which is corresponding to the emitted electron within ±15° of the surface normal of the sample. It is obvious that the Kikuchi pattern is clear when the electron landing positions within the boundary of the rotating crystal island as marked in the white dashed curve according to Fig. 6c, and no pattern could be found when the electron landing positions in the amorphous region out of the rotating crystal island.

Because the crystal orientation of each location on the rotating crystal island is changing with its crystal position, the Kikuchi pattern observed in the CCD image also changes slowly and continuously with the moving of the electron landing position at the rotating crystal island.

This is because the resolution of the current adopted EBSD equipment is about 10 nm, so the angular rotation of the crystal in the region of 10 nm on the rotating crystal island is only 0.2 degrees. At such a small resolution, the Kikuchi pattern generated at each local electron landing position can be considered as the Kikuchi pattern measured at a single crystal of In$_2$O$_3$ in a certain crystal orientation. Thus for an ideal disk-shaped rotating crystal island crystal, such a special Kikuchi pattern will be observed when electrons land on the center of the rotating crystal island.

More interestingly, this phenomenon is a concrete manifestation of the “Banach fixed-point theorems” in mathematics. The famous “Banach fixed-point theorems” [85] is an important tool in the theory of metric spaces; it guarantees the existence and uniqueness of fixed points of certain self-maps of metric spaces, and provides a constructive method to find those fixed points. In fact, the Banach fixed-point theorem is also known as the contraction mapping theorem or contractive mapping theorem. One example is that if a map of a country is printed in a reduced size inside the territory of that country, there is one and only one such point on the map, whose position in the
map also happens to indicate the location of the land on which it falls. Here, the Kikuchi pattern observed in the SEM image from a rotating crystal sample must coincide with one of the Kikuchi patterns in the upper region of CCD image taken at a certain location in this rotating crystal sample in the EBSD Kikuchi pattern matrix map. It is very interesting to note that this small Kikuchi pattern region in the observed CCD image which is originated from those electrons emitted near the surface normal of the rotating crystal island, shows almost the same pattern as those Kikuchi patterns observed in the SEM image of this rotating crystal sample (Fig. 6c), which is also an example of Banach fixed-point theorems.

4. Summary

It is worth mentioning here that although a large number of rotating crystals have been observed in previous reports as well. These rotating crystals share the common property that the crystal orientation at the local position of the crystal rotates depending on the position. However, the large number of rotating crystals reported are different in both physical properties and appearance. Therefore, we need a more detailed classification of these special crystals based on the most essential differences in the way their local crystal orientation rotates. Just as in the case of crystalline materials, they are classified according to the different symmetries they exhibit. Here we also advocate applying similar ideas to classify the found rotating crystals in a more refined way. For example, the two-dimensional rotating crystalline film found in this paper not only satisfies the basic characteristics of a rotating crystal, i.e., the crystal orientation at the local position of the crystal rotates depending on the position, but also satisfies the cylindrical symmetry with respect to the crystal orientation. Therefore, the rotating crystals found in this paper should be called cylindrically symmetric rotating crystals.
Finally, it is worth mentioning that this extremely special thin film of cylindrically symmetric rotating crystals prepared in this paper possesses physically interesting properties. For, the cylindrically symmetric rotating crystal film prepared in this paper provides a very extreme case study of SEM contrast. The cylindrically symmetric rotating crystal film, in the raster scan mode of SEM, will not only demonstrate in the image both the particle properties of the incident electrons, i.e., the surface morphology of the sample detected by the interaction of the incident electrons with the sample, but also the wave properties of the incident electrons, i.e., the lattice-related information of the sample carried by the diffraction interaction of the incident electrons with the sample lattice. In addition, the special cylindrically symmetric rotating crystal film prepared in this paper is also mathematically interesting. The SEM image of the cylindrically symmetric rotating crystal film prepared in this paper has an interesting mathematical property associated with the EBSD Kikuchi pattern map, namely the immobility principle. The SEM image of the cylindrically symmetric rotating crystal film is like a thumbnail of the EBSD Kikuchi pattern map. In fact, the association of SEM images with the EBSD Kikuchi pattern map is a concrete example of the Banach fixed-point theorems in the field of materials science. This makes the SEM image we took with the EBSD Kikuchi pattern map demonstrate one of the common properties that the map should have under the mathematical definition, the Banach fixed-point theorems.
References

[1] McMullan D. Scanning electron microscopy 1928–1965[J]. Scanning, 1995, 17(3): 175-185.

[2] "Half-Tone Photo-Engraving". The Photographic Times. Scoville Manufacturing Co. 25: 121–123. 1894.

[3] Zworykin VA, Hillier J, Snyder RL (1942) A scanning electron microscope. ASTM Bull 117, 15–23.

[4] K. C. A. Smith and C. W. Oatley, Br. J. Appl. Phys. 6, 391 (1955).

[5] Hejna J. Detection of topographic contrast in the scanning electron microscope at low and medium resolution by different detectors and detector systems[J]. Scanning Microscopy, 1994, 8(2): 1.

[6] L J Allen et al 2010 J. Phys.: Conf. Ser. 241 012061

[7] Geoffrey E. LLOYD, Mineralogical Magazine, March 1987, 51, 3-19

[8] Shibata N, Findlay S D, Sasaki H, et al. Imaging of built-in electric field at a pn junction by scanning transmission electron microscopy[J]. Scientific reports, 2015, 5(1): 1-8.

[9] Hachtel J A, Idrobo J C, Chi M. Sub-Ångstrom electric field measurements on a universal detector in a scanning transmission electron microscope[J]. Advanced structural and chemical imaging, 2018, 4(1): 1-10.

[10] Jones G A. Magnetic contrast in the scanning electron microscope: an appraisal of techniques and their applications[J]. Journal of Magnetism and Magnetic Materials, 1978, 8(4): 263-285.
[11] Kotera M, Katoh M, Suga H S H. Observation technique of surface magnetic structure using type-I magnetic contrast in the scanning electron microscope[J]. Japanese journal of applied physics, 1995, 34(12S): 6903.

[12] Kriaa, H., Guitton, A. & Maloufi, N. Fundamental and experimental aspects of diffraction for characterizing dislocations by electron channeling contrast imaging in scanning electron microscope. Sci Rep 7, 9742 (2017).

[13] Howie A, Whelan M J. Diffraction contrast of electron microscope images of crystal lattice defects-II. The development of a dynamical theory[J]. Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences, 1961, 263(1313): 217-237.

[14] Hata S, Furukawa H, Gondo T, et al. Electron tomography imaging methods with diffraction contrast for materials research[J]. Microscopy, 2020, 69(3): 141-155.

[15] Crimp M A. Scanning electron microscopy imaging of dislocations in bulk materials, using electron channeling contrast[J]. Microscopy research and technique, 2006, 69(5): 374-381.

[16] Gutierrez-Urrutia I, Zaefferer S, Raabe D. Electron channeling contrast imaging of twins and dislocations in twinning-induced plasticity steels under controlled diffraction conditions in a scanning electron microscope[J]. Scripta Materialia, 2009, 61(7): 737-740.

[17] A.J. Schwartz, M. Kumar, B.L. Adams, D.P. Field, Electron Backscatter Diffraction in Materials Science, 2nd ed., Springer, Boston, 2009

[18] Hussain A, Yang L, Mao S, et al. Determination of electron backscattering coefficient of beryllium by a high-precision Monte Carlo simulation[J]. Nuclear Materials and Energy, 2021, 26: 100862.
[19] Bragg W (1913). "The Diffraction of Short Electromagnetic Waves by a Crystal". Proceedings of the Cambridge Philosophical Society. 17: 43–57.

[20] P. Hirsch; A. Howie; R. Nicholson; D. W. Pashley; M. J. Whelan (1977). Electron microscopy of thin crystals. Butterworths/Krieger, London/Malabar FL. ISBN 978-0-88275-376-8.

[21] Kikuchi S. Electron diffraction in single crystals[J]. Japanese Journal of Physics, 1928, 5(3061): 83-96.

[22] Coates D G. Kikuchi-like reflection patterns obtained with the scanning electron microscope[J]. The Philosophical Magazine: A Journal of Theoretical Experimental and Applied Physics, 1967, 16(144): 1179-1184.

[23] Joy D C, Newbury D E, Davidson D L. Electron channeling patterns in the scanning electron microscope[J]. Journal of Applied Physics, 1982, 53(8): R81-R122.

[24] Venables J A, Harland C J. Electron back-scattering patterns—A new technique for obtaining crystallographic information in the scanning electron microscope[J]. Philosophical Magazine, 1973, 27(5): 1193-1200.

[25] Langer E, Däbritz S. Investigation of HOLZ rings in EBSD patterns[J]. physica status solidi, 2007, 4(6): 1867-1872.

[26] Mitoma N, Da B, Yoshikawa H, et al. Phase transitions from semiconductive amorphous to conductive polycrystalline in indium silicon oxide thin films[J]. Applied Physics Letters, 2016, 109(22): 221903.

[27] S. Aikawa, T. Nabatame, and K. Tsukagoshi, Appl. Phys. Lett. 103, 172105 (2013).
[28] N. Mitoma, S. Aikawa, X. Gao, T. Kizu, M. Shimizu, M.-F. Lin, T. Nabatame, and K. Tsukagoshi, Appl. Phys. Lett. 104, 102103 (2014).

[29] Venables J A, Spiller G D T. Nucleation and growth of thin films[M]//Surface Mobilities on Solid Materials. Springer, Boston, MA, 1983: 341-404.

[30] Evans J W, Thiel P A, Bartelt M C. Morphological evolution during epitaxial thin film growth: Formation of 2D islands and 3D mounds[J]. Surface science reports, 2006, 61(1-2): 1-128.

[31] Shigeto K, Kizu T, Tsukagoshi K, et al. Radial Interference Contrast in in-situ SEM Observation of Metal Oxide Semiconductor Film Crystallization[J]. Microscopy and Microanalysis, 2017, 23(S1): 1512-1513.

[32] Gonzalez D, Kelleher J F, da Fonseca J Q, et al. Macro and intergranular stress responses of austenitic stainless steel to 90 strain path changes[J]. Materials Science and Engineering: A, 2012, 546: 263-271.

[33] Zhou S, Antoja-Lleonart J, Nukala P, et al. Crystallization of GeO2 thin films into α-quartz: From spherulites to single crystals[J]. Acta Materialia, 2021, 215: 117069.

[34] Lutjes N R, Zhou S, Antoja-Lleonart J, et al. Spherulitic and rotational crystal growth of Quartz thin films[J]. Scientific reports, 2021, 11, 14888.

[35] B. Da, S.F. Mao and Z.J. Ding

Validity of the Semi-classical Approach for Calculation of the Surface Excitation Parameter

J. Phys.: Condens. Matter 23 (2011) 395003.

[36] B. Da, S.F. Mao, G.H. Zhang, X.P. Wang and Z.J. Ding
Monte Carlo Modeling of Surface Excitation in Reflection Electron Energy Loss Spectroscopy Spectrum for Rough Surfaces

J. Appl. Phys. 112 (2012) 034310.

[37] B. Da, Y. Sun, S.F. Mao, Z.M. Zhang, H. Jin, H. Yoshikawa, S. Tanuma and Z.J. Ding

A Reverse Monte Carlo Method for Deriving Optical Constants of Solids from REELS Spectra

J. Appl. Phys. 113 (2013) 214303.

[38] B. Da, S. F. Mao, Y. Sun and Z.J. Ding

A New Analytical Method in Surface Electron Spectroscopy: Reverse Monte Carlo Method

e-J. Surf. Sci. Nanotech. 10 (2012) 441-446.

[39] B. Da, Z.Y. Li, H.C. Chang, S.F. Mao and Z.J. Ding

A Monte Carlo Study of Reflection Electron Energy Loss Spectroscopy Spectrum of a Carbon Contaminated Surface

J. Appl. Phys. 116 (2014) 124307.

[40] B. Da, L.H. Yang, J.W. Liu, Y.G. Li, S.F. Mao and Z.J. Ding

Monte Carlo Simulation Study of Reflection Electron Energy Loss Spectroscopy of a Fe/Si Layered Nanostructure

Surf. Interface Anal. 52 (2020) 742-754.

[41] D.B. Lu, K. Goto, B. Da, J.W. Liu, H. Yoshikawa, S. Tanuma and Z.J. Ding
Secondary Electron-, Auger Electron- and Reflected Electron-Spectroscopy Study on sp2-Hybridization Carbon Materials: HOPG, Carbon Glass and Carbon Fiber

J. Electr. Spectrosc. Rela. Phenom. 250 (2021) 147086.

[42] D.B. Lu, Z.F. Hou, X. Liu, B. Da and Z.J. Ding

Ab-Initio Simulation of Position-Dependent Electron Energy Loss and Its Application to the Plasmon Excitation of Nanographene

J. Phys. Chem. C 123 (2019) 25341-25348.

[43] X. Liu, L.H. Yang, Z.F. Hou, B. Da, K. Nagata, H. Yoshikawa, S. Tanuma, Y. Sun and Z.J. Ding

Machine Learning Approach for the Prediction of Electron Inelastic Mean Free Paths

Phys. Rev. Mater. 5 (2021) 033802.

[44] X. Liu, Z.F. Hou, D.B. Lu, B. Da, H. Yoshikawa, S. Tanuma, Y. Sun and Z.J. Ding

Unveiling the Principle Descriptor for Predicting the Electron Inelastic Mean Free Path Based on a Machine Learning Framework

Sci. Technol. Adv. Mater. 20 (2019) 1090-1102.

[45] B. Da, H. Shinotsuka, H. Yoshikawa, Z.J. Ding and S. Tanuma

Extended Mermin Method for Calculating the Electron Inelastic Mean Free Path

Phys. Rev. Lett. 113 (2014) 063201.
[46] Bo Da, Jiangwei Liu, Yoshitomo Harada, Nguyen T. Cuong, Kazuhito Tsukagoshi, Jin Hu, Lihao Yang, Zejun Ding, Hideki Yoshikawa, Shigeo Tanuma. Observation of Plasmon Energy Gain for Emitted Secondary Electron in Vacuo. The Journal of Physical Chemistry Letters. 10 [19] (2019) 5770-5775

[47] B. Da, J.W. Liu, M. Yamamoto, Y. Ueda, K. Watanabe, N.T. Cuong, S.L. Li, K. Tsukagoshi, H. Yoshikawa, H. Iwai, S. Tanuma, H.X. Guo, Z.S. Gao, X. Sun and Z.J. Ding

Virtual Substrate Method for Nanomaterials Characterization

Nature Commun. 8 (2017) 15629.

[48] B. Da, Y. Sun, Z.F. Hou, J.W. Liu, N.T. Cuong, K. Tsukagoshi, H. Yoshikawa, S. Tanuma, J. Hu, Z.S. Gao and Z.J. Ding

Measurement of the Low-Energy Electron Inelastic Mean Free Path in Monolayer Graphene

Phys. Rev. Appl. 13 (2020) 044055.

[49] L.H. Yang, K. Tőkési, J. Tóth, B. Da and Z.J. Ding

High Precision Determination of Optical Properties of Silicon and Germanium from Reflection Electron Energy Loss Spectroscopy Spectra

Phys. Rev. B 100 (2019) 245209.

[50] L.H. Yang, K. Tőkési, J. Tóth, B. Da and Z.J. Ding

Revision of Optical Property of Silicon by a Reverse Monte Carlo Analysis of Reflection Electron Energy Loss Spectroscopy Spectra

J. Phys.: Conf. Ser. 1412 (2020) 202026.
[51] L.H. Yang, B. Da, K. Tőkési and Z.J. Ding

Individual Separation of Surface, Bulk and Begrenzungs Effect Components in the Surface Electron Energy Spectra

Sci. Rep. 11 (2021) 5954.

[52] L.H. Yang, A. Hussain, S.F. Mao, B. Da, K. Tőkési and Z.J. Ding

Electron Backscattering Coefficients of Molybdenum and Tungsten Based on the Monte Carlo Simulations

J. Nucl. Mater. 553 (2021) 153042.

[53] D.B. Lu, K. Goto, B. Da, J.W. Liu, H. Yoshikawa, S. Tanuma and Z.J. Ding

Secondary Electron-, Auger Electron- and Reflected Electron-Spectroscopy Study on sp2-Hybridization Carbon Materials: HOPG, Carbon Glass and Carbon Fiber

J. Electr. Spectrosc. Rela. Phenom. 250 (2021) 147086.

[54] L.H. Yang, J.M. Gong, A. Sulyok, M. Menyhárd, G. Sáfrán, K. Tőkési, B. Da and Z.J. Ding

Optical Properties of Amorphous Carbon Determined by Reflection Electron Energy Loss Spectroscopy Spectra

Phys. Chem. Chem. Phys. 23 (2021) 25335-25346.

[55] H. Xu, B. Da, J. Tóth, K. Tőkési and Z.J. Ding

Absolute Determination of Optical Constants by Reflection Electron Energy Loss Spectroscopy Spectra
Phys. Rev. B 95 (2017) 195417.

[56] H. Xu, L.H. Yang, J. Tóth, Tőkési, B. Da and Z.J. Ding

Absolute Determination of Optical Constants of Three Transition Metals Using Reflection Electron Energy Loss Spectroscopy

J. Appl. Phys. 123 (2018) 043306.

[57] L.H. Yang, J. Tóth, K. Tőkési, B. Da and Z.J. Ding

Calculation of Electron Inelastic Mean Free Path of Three Transition Metals from Reflection Electron Energy Loss Spectroscopy Spectrum Measurement Data

Eur. Phys. J. D 73 (2019) 21.

[58] Cheng L, Ming Y, Ding Z J. Bohmian trajectory-bloch wave approach to dynamical simulation of electron diffraction in crystal[J]. New Journal of Physics, 2018, 20(11): 113004.

[59] Cheng L. Novel Quantum Trajectory Approaches to Simulation of Electron Backscatter Diffraction[J]. e-Journal of Surface Science and Nanotechnology, 2020, 18: 121-125.

[60] Kooi, B. J. & De Hosson, JTh. M. On the crystallization of thin films composed of Sb3.6Te with Ge for rewritable data storage. J. Appl. Phys. 95, 4714–4721 (2004).

[61] Savytskii, D., Jain, H., Tamura, N. & Dierolf, V. Rotating lattice single crystal architecture on the surface of glass. Sci. Rep. 6, 36449 (2016).

[62] Kolosov, V. Y. & Thölén, A. R. Transmission electron microscopy studies of the specific structure of crystals formed by phase transition in iron oxide amorphous films. Acta Mater. 48, 1829–1840 (2000).
[63] Shtukenberg, A. G., Punin, Y. O., Gujral, A. & Kahr, B. Growth actuated bending and twisting of single crystals. Angew. Chem. Int. Ed. 53, 672–699 (2014).

[64] Krumdieck S P, Boichot R, Gorthy R, et al. Nanostructured TiO2 anatase-rutile-carbon solid coating with visible light antimicrobial activity[J]. Scientific reports, 2019, 9(1): 1883.

[65] Fedchenko O, Winkelmann A, Medjanik K, et al. High-resolution hard-x-ray photoelectron diffraction in a momentum microscope—The model case of graphite[J]. New Journal of Physics, 2019, 21(11): 113031.

[66] Goh, K. W., Johan, M. R., & Wong, Y. H. (2018). Enhanced structural properties of In2O3 nanoparticles at lower calcination temperature synthesised by co-precipitation method. Micro & Nano Letters, 13(2), 270-275.

[67] J.F. Nye, Some geometrical relations in dislocated crystals, Acta Mater., 1 (1953), pp. 153-162

[68] Konijnenberg P J, Zaefferer S, Raabe D. Assessment of geometrically necessary dislocation levels derived by 3D EBSD[J]. Acta Materialia, 2015, 99: 402-414.

[69] Callister, William D. Jr. "Fundamentals of Materials Science and Engineering," John Wiley & Sons, Inc. Danvers, MA. (2005)

[70] Dasari S G, Nagaraju P, Yelsani V, et al. Nanostructured indium oxide thin films as a room temperature toluene sensor[J]. ACS omega, 2021, 6(27): 17442-17454.

[71] A.G. Shtukenberg, Y.O. Punin, E. Gunn, B. Kahr, Spherulites, Chem. Rev., 112 (3) (2012), pp. 1805-1838
[72] Sun C Y, Gránásy L, Stifler C A, et al. Crystal nucleation and growth of spherulites demonstrated by coral skeletons and phase-field simulations[J]. Acta biomaterialia, 2021, 120: 277-292.

[73] A. Shtukenberg, J. Freundenthal, E. Gunn, L. Yu, B. Kahr, Glass-crystal growth mode for testosterone propionate, Cryst. Growth Des., 11 (10) (2011), pp. 4458-4462

[74] A.G. Shtukenberg, C.T. Hu, Q. Zhu, M.U. Schmidt, W. Xu, M. Tan, B. Kahr, The third ambient aspirin polymorph, Cryst. Growth Des., 17 (6) (2017), pp. 3562-3566

[75] X. Cui, A.L. Rohl, A. Shtukenberg, B. Kahr, Twisted aspirin crystals, J. Am. Chem. Soc., 135 (9) (2013), pp. 3395-3398

[76] I. E. Bolotov, V. Yu. Kolosov, A. V. Kozhyn, Phys. Status Solidi A 1982, 72, 645–654.

[77] V. Yu. Kolosov, C. L. Schwamm, R. V. Gainutdinov, A. L. Tolstikhina, J. Phys. Conf. Ser. 2008, 100, 082037.

[78] V. Yu. Kolosov, K. L. Shvamm, R. V. Gainutdinov, A. L. Tolstikhina, Bull. Russ. Acad. Sci. Phys. 2007, 71, 1442–1446.

[79] V. Yu. Kolosov, A. R. Thörlen, Acta Mater. 2000, 48, 1829–1840.

[80] A. G. Bagmut, S. N. Grigorov, V. A. Zhuchkov, V. Yu. Kolosov, V. M. Kosevich, D. V. Mel’nichenko, Russ. Phys. J. 2007, 50, 1071–1078.

[81] V. Yu. Kolosov, C. L. Schwamm, J. W. Steeds, J. Phys. Conf. Ser. 2008, 100, 082038.

[82] V. Yu. Kolosov, L. M. Veretennikov, Yu. B. Starseva, C. L. Schwamm, Semiconductors 2005, 39, 955–959.
[83] V. Yu. Kolosov, A. V. Kozhin, L. M. Veretennikov, C. L. Schwamm, EMC 2008, Vol. 2 (Eds.: S. Richter, A. Schwedt), Springer, Berlin, 2008, pp. 343–344.

[84] V. Yu. Kolosov, EMC 2008, Vol. 2 (Eds.: S. Richter, A. Schwedt), Springer, Berlin, 2008, pp. 657–658.

[85] Brown, R. F., ed. (1988). Fixed Point Theory and Its Applications. American Mathematical Society. ISBN 0-8218-5080-6.
Support materials

Fig. S1 SEM image in different working distances. These BSE images were taken with a work distance of (a) 5.3 mm and (b) 20.5 mm. The figures were for the BSE of InSiO film where the rotational crystal islands are observed after annealing at 300 °C.

Fig. S2 (a) the structure of BSE detector. (b)(c)(d) BSE images taken using the different parts of BSE detectors. The red part is the detector inside, while the blue part is the outside. The rotational crystal islands are observed after annealing at 300 °C.
Fig. S3 (a) SE and (b) BSE image for the final state of the annealing under the temperature of 300 °C and electron energy of 3 keV.

Fig. S4 In$_2$O$_3$ Kikuchi pattern spherical projection with the incident electron energy of 15 keV. The Bragg wave method with an incident energy of 15 keV was used to simulate the different crystallographic atomic structure maps of In$_2$O$_3$. The angular range of theoretical [222], [211], [440] ECP patterns is ±15°. The experimental rotational crystal islands correspond to the
crystallographic orientation found in the rotating crystal of InSiO. The Miller indexes are labeled on the figure.

Mov. S1 The mp4 file is the crystallization process of InSiO film from 250°C to 300°C, with the incident electron energy of 15keV. In-situ SEM observations were performed utilizing the combination of heating sub-stage Gatan Murano and Schottky SEM Hitachi SU5000. This combined system allows us to obtain images of crystallization dynamically along a precisely fixed observation area.