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Study of very thin crystals by rainbow scattering effect

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Abstract

This research introduces an innovative methodology for the analysis of very thin crystals, utilizing the rainbow scattering, a technique previously unexploited for this application. The focus is on assessing the capabilities of this method for an in-depth investigation of crystalline structures. To characterize the very thin crystals, we implemented numerical simulation techniques to model and evaluate rainbow scattering phenomena under channeling conditions. The simulations aimed to forecast the scattering patterns of protons as they traversed various cubic crystal types at different orientations. We identified distinct patterns of rainbow scattering, providing essential insights into the internal configurations and orientations of the cubic crystals examined. The combination of channeling mode rainbow scattering with numerical simulations presents a fresh and efficient approach for analyzing very thin crystals, setting the stage for subsequent experimental investigations. This advancement holds considerable promise for enhancing research in the fields of materials science and nanotechnology.

Introduction

Thin crystal characterization is a crucial field in materials science, particularly in the study of thin films and nanostructures. Over the years, a variety of techniques have been employed to characterize thin crystals, each providing valuable insights into the material's structure and properties (Benz 2014). Ion channeling stands out as a powerful technique for studying the internal structure of crystalline materials (Robinson 1963). When particles pass through a well-aligned crystal, they are "channeled" by the potential well created by the atomic lattice (Gemel 1974). In this process, if the crystal is sufficiently thin, the ions will be transmitted, and each ion preserves its unique spatial and angular information, providing detailed insights into the channeled trajectories. The challenges of analyzing angular distributions of channeled ions through thin crystals can be

easily and effectively overcome by applying the crystal rainbow theory (Petrović 2000). In the case of channeling, the scattering angles of ions exhibit distinct features and are constrained within specific values known as rainbow lines. This effect arises due to the singularity of the classical differential cross-section in ion scattering. The rainbow lines in the transmission angle (TA) plane are comparable with the angular distribution of channeled ions. Crystal rainbow theory accurately predicts the spatial and angular distributions of ions channeled through crystals.

Experimental proof of crystal rainbow theory was conducted in a high-resolution ion transmission channeling experiment (Motapothula 2012). Detailed analysis of crystal rainbow theory led to the correction of the ion-crystal interaction potential in the case of channeling (Petrović 2015). Based on these results, we constructed interaction potentials for other cubic crystals (Petrović 2019, Starčević 2021, 2023). Also influence of crystal electrons on channeled ions is determined (Starčević 2023). The "doughnut effect" which occurs when the ion beam is tilted away from a major crystallographic direction, was theoretically explained using crystal rainbow theory in interpretation of experimental results of angular distributions of channeled protons as the crystal tilting angles increase (Motapothula 2012). Applications of rainbow scattering for theoretical analysis of graphene sheets have led to the determination of several physical quantities, (Ćosić 2019, 2021, Hadžijojić 2021, 2023).

One of the remarkable features of rainbow lines is that by analyzing angular distributions of channeled ions, which are completely determined by the shape of rainbow lines, it is possible to determine the shape of the crystal channel. As the image of rainbow lines in the impact parameter (IP) plane lie within the channel region where the contributions of all atomic strings defining the channel are significant this line will be “repelled” by the atomic strings defining the channel, and the coordinates of its points are solely determined by the arrangement of the atomic strings. This means that the angular distributions of transmitted ions can be described, since rainbow lines in the TA plane appear as their “skeleton”. Further on, unknow crystal thickness is indicated by the reduced thickness parameter Λ (Krause 1994), which represents the crystal thickness in terms of the wavelength for the projectile's transverse oscillating motion $\Lambda = f(q, m_p)L/v$, where L is the crystal thickness, v is the projectile speed, and f is the transverse oscillation frequency of the projectile, q and m_p are its charge and mass. The evolution of the angular distributions with the increase of L can be precisely tracked using the reduced crystal thickness, so the evolution or rainbow lines can also be tracked using this variable. By considering crystal rainbow theory and the scaling law, it is possible to determine the type and thickness of a very thin crystal based on the recorded rainbow lines, provided the energy and charge of the projectile are known. Given these factors, we opted to use protons with energy in the MeV range, as we already have experience analyses with these projectiles (Motapothula 2012).

Analysis and results

We are investigating the scattering patterns of 2 MeV protons channeled through very thin cubic crystals in major crystallographic orientations. The main criterion for analysis is that all crystals are considered very thin. With body-centered cubic (BCC), face-centered cubic (FCC), and diamond cubic lattices, including vanadium, chromium, iron, niobium, molybdenum, barium, europium, tantalum, tungsten, aluminum, calcium, nickel, copper, strontium, rhodium, palladium, silver, cerium, ytterbium, iridium, platinum, gold, lead, thorium, silicon, germanium and tin crystals are investigated. The main morphological difference between these three types of cubic crystals lies in the number and position of atoms in the primitive lattice, as shown on Fig. 1.

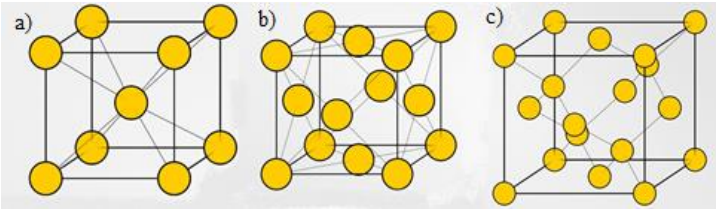

Fig. 1. Primitive lattice of: a) BCC, b) FCC and c) diamond crystals

The differences in the arrangement and positions of atoms in crystal lattices determine the channel shape and positions of its atomic strings. Fig. 2 shows square channel for in (001) orientation and hexagonal channels in (111). In FCC crystals, the channel side length will be smaller by a factor of $\sqrt{2}$ compared to that of BCC, and for diamond cubic crystals, the channel will have a side length half that of BCC crystals. Since the shapes of the channels presented in Fig. 2 a), b), and c), d) are completely different, a simple morphological comparison of the rainbow lines can determine the crystal orientation in question. However, a limitation of this morphological approach is that we cannot definitively identify the crystal type based solely on the shape of the rainbow line.

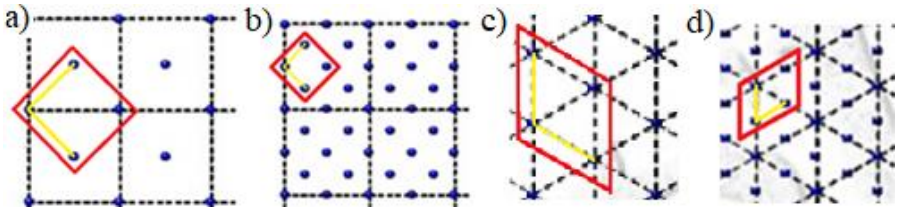

Fig. 2. a) BCC (001), b) diamond (001), c) BCC (111), d) diamond (111) channels

In Fig. 3, the primary rainbow lines of protons channeled through a) square (001) and b) hexagonal (111) channels are presented. The proton energy, crystal

type, and thickness are chosen such that only the primary rainbow line appears in this theoretical representation. Square channel exhibit inner rainbow lines shaped like a cusped square, with the cusps pointing toward the angular positions of the atomic strings defining the channel. In contrast, all hexagonal channels will generate rainbow lines resembling a symmetrical six-pointed star, composed of two cusped equilateral triangles oriented opposite each other. Solely based on the shape of these lines, we can conclude what crystal channel is in question. Thus, we can conclude that the shape of the primary rainbow line allows us to determine the orientation of an unknown crystal.

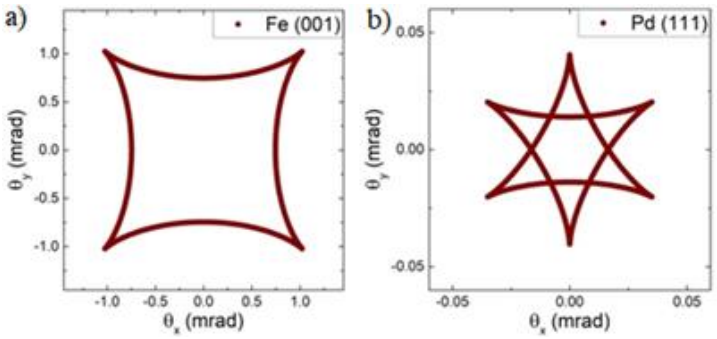

Fig. 3. Shape of rainbow lines for a) square and d) hexagonal channel

As is known, the size of the rainbow line depends on the interaction potential, which is a function of the crystal atomic number and the impact parameter (size of the channel). If we change (gradually reduce) the projectile energy in comparison to the first part of our analysis at some point, a secondary or outer rainbow line will emerge in the area surrounding the atomic strings in the IP plane, and its image will appear in the TA plane. The outer rainbow line will be highly sensitive to the atomic number of the crystal and will not be as sensitive to the influence of all atomic strings and their spatial positions. By measuring the angular distributions of channeled protons and comparing them to the rainbow lines - now consisting of one inner (primary) rainbow line and one outer (secondary) rainbow line - we can conduct a similar morphological analysis to identify the unknown crystal.

In Fig. 4, we present the experimental angular distribution of 2 MeV protons channeled through the (001) channel of a 55 nm thin silicon crystal, along with the calculated rainbow lines in the TA plane and the corresponding rainbow lines in the IP plane. This combination of crystal thickness and proton energy yields a reduced crystal thickness less than 0.25, so the silicon crystal can be considered very thin.

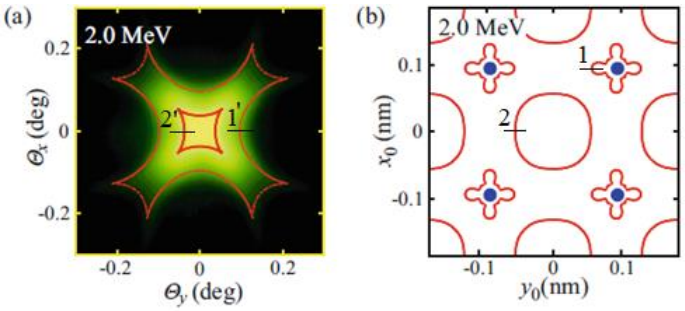

Fig. 4. a) Proton experimental angular distributions with rainbow lines, b) corresponding rainbow lines in the IP plane

Based on these results, we conducted an analysis of all cubic crystals in the (001) orientation. Our goal is to, by calculating the angular position of the outer rainbow line, deduce the atomic number of the crystal. We specifically calculated the positions of the outer rainbow lines along position 1 on Fig. 4, and determined its position 1' in the TA plane. From Fig. 5 a) we can see that as the atomic number of crystal increases, the position of the outer rainbow line in the TA plane exhibits a growing trend. Thus, by measuring this position, we can deduce which crystal is being analyzed. However, some minor flaws in this morphological methodology arise from the fact that some crystals have similar ratio of atomic number and channel size. This leads to a slight indeterminacy when distinguishing between these elements.

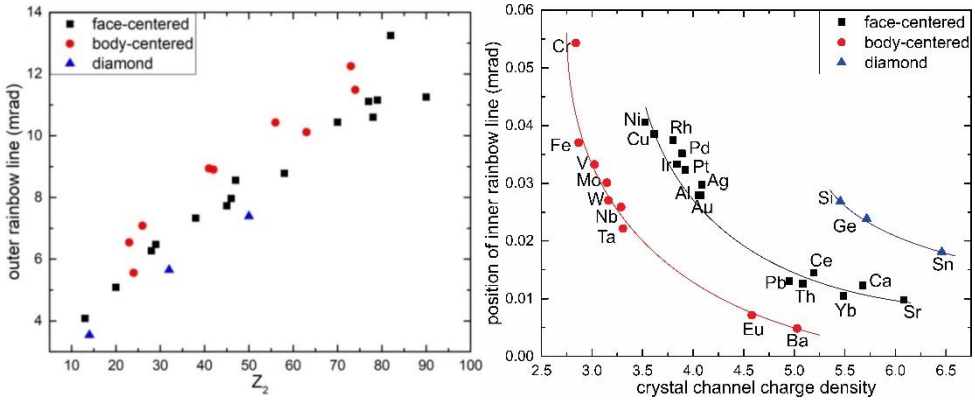

Fig. 5. Positions of a) outer rainbow lines, b) inner rainbow lines

However, if we consider the physical parameters that differentiate these crystals - such as channel area, thickness of one layer, crystal atomic number - and compare them with the positions of the inner rainbow lines for all these types of cubic crystals, as presented in Fig. 5 b), some ambiguities will be clarified. The surface charge density per crystal channel is defined as the ratio of the atomic number of the crystal to the number of atomic strings defining the crystal channel. This

approach allows us to overcome the morphological differences between the three types of cubic crystals, specifically addressing the total number of atoms in one crystal lattice as seen in cross-section for each different orientation. For all analyzed cubic crystals in the (001) orientation, the number of atomic strings per primitive crystal lattice is five for BCC crystals, nine for FCC crystals, and thirteen for diamond cubic crystals. This allows us to categorize these cubic crystals into three distinct groups, as illustrated in Fig. 5 b). All crystals with a BCC structure are fitted along the red line, those with a FCC structure along the black line, and diamond crystals are fitted along the blue line. Notably, while some elements may have similar positions for their outer rainbow lines, they can be easily distinguished by considering the morphological position of their inner rainbow line, making it straightforward to sort them based on these values. This way total separation of the different elements is achieved, with the main criterion that the crystal is considered very thin.

Conclusions

We applied crystal rainbow theory to characterize various cubic crystals. Our findings underscore the potential of using crystal rainbow theory as a powerful tool for the characterization of thin crystals. We demonstrated that the morphology of rainbow lines provides a reliable means to determine the orientation of unknown thin crystals. Our analysis revealed distinct differences in the shapes of channels and their corresponding rainbow lines across different orientations. We also showed that the position of the secondary rainbow line can be effectively used to distinguish between different crystals, as its position increases with the atomic number of the crystals. We introduced the concept of charge density per crystal channel, which allowed us to categorize the cubic crystals using inner rainbow line into three distinct groups, each corresponding to a different type of cubic crystal. Overall, our findings underscore the potential of using crystal rainbow theory as a powerful tool for the morphological characterization of thin crystals. Future studies could focus on extending this methodology to other crystals, thereby broadening its applicability and enhancing our understanding of ion channeling phenomena.

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