

Article

Simulation of Two-Dimensional Images for Ion-Irradiation Induced Change in Lattice Structures and Magnetic States in Oxides by Using Monte Carlo Method

Akihiro Iwase *  and Shigeru Nishio

The Wakasa Wan Energy Research Center (WERC), 64-52-1 Nagatani, Tsuruga, Fukui 914-0192, Japan; snishio@werc.or.jp

* Correspondence: aiwase@werc.or.jp or iwase@mtr.osakafu-u.ac.jp

Abstract: A Monte Carlo method was used to simulate the two-dimensional images of ion-irradiation-induced change in lattice structures and magnetic states in oxides. Under the assumption that the lattice structures and the magnetic states are modified only inside the narrow one-dimensional region along the ion beam path (the ion track), and that such modifications are affected by ion track overlapping, the exposure of oxide targets to spatially random ion impacts was simulated by the Monte Carlo method. Through the Monte Carlo method, the evolutions of the two-dimensional images for the amorphization of TiO_2 , the lattice structure transformation of ZrO_2 , and the transition of magnetic states of CeO_2 were simulated as a function of ion fluence. The total fractions of the modified areas were calculated from the two-dimensional images. They agree well with the experimental results and those estimated by using the Poisson distribution functions.

Keywords: high energy irradiation; ion track overlapping; oxides; Monte Carlo simulation for two-dimensional images; lattice structures and magnetic states; binomial and Poisson distribution functions



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1. Introduction

It is well known that, in a lot of polymers, the energetic ion irradiation and the subsequent chemical etching produce one-dimensional holes with small diameters [1]. Resultant perforated membranes have been used as filters for small particles [1,2]. In some ceramics irradiated with swift heavy ions, one-dimensional areas, in which the lattice structures and the physical properties are strongly modified, are produced along the ion beam path. Such one-dimensional structures are called “ion-tracks” [3]. The ion-tracks originate from the energetic ion induced high-density electronic excitation, and their production mechanisms have been explained by using the thermal spike models [4–6] and by the Coulomb explosion model [7–9]. A lot of studies have been performed in order to investigate the individual ion-track structures and their dependence on the electronic stopping power, S_e , of the irradiating ions [10–15]. The effects of the ion-track overlapping on lattice structures of target materials, which appear for the high fluence irradiation, have also been investigated so far [10,16–22].

Ishikawa et al. have explained the ion track overlapping effect on the TiO_2 amorphization by using the binomial distribution function [23]. In our recent study, we analyzed the ion-track overlapping effect on the magnetization of CeO_2 by using the Poisson distribution function, and succeeded in the reproduction of the experimentally observed ion-fluence dependence of the magnetization [24]. As shown later in this report, the Poisson distribution function, which is the approximated formula of the binomial distribution function under some extreme condition, can also describe the ion-track overlapping effects on the evolution of the amorphization in TiO_2 and the lattice structure change in ZrO_2 . Such analytical methods, however, only reproduce the total fraction of the areas modified by

high energy ions as a function of ion fluence. In the present study, we used the Monte Carlo method in order to simulate the two-dimensional images of the lattice structures and the magnetic states as a result of the ion-track overlapping for TiO_2 , ZrO_2 and CeO_2 . The total fractions of the modified area were calculated from the two-dimensional images, and were compared with the experimental results and those estimated by using the Poisson distribution function.

2. Binomial and Poisson Distribution Functions for the Analysis of the Ion Track Overlapping Effect

The binomial distribution function,

$$f(n, k, p) = \frac{n!}{k!(n-k)!} p^k (1-p)^{n-k}, \quad (1)$$

generally presents the discrete probability distribution of the number of successes, k , in a sequence of n independent trials, where p is the probability of the success and $1-p$, that of the failure for each trial. The paper of Ishikawa et al. [23] and our previous report [24] have shown that the probability of the ion-track overlapping can be described by the binomial distribution function. When the fluence of ions is Φ and the total area of the target is S_0 , the number of irradiating ions, n ($=\Phi S_0$) corresponds to the trial number. The ratio of the cross section of each ion track, S , to S_0 ($s = S/S_0$) and the number of track impacts, r , correspond to p and k in Equation (1), respectively. The probability of r times track impacts is given as

$$a(n, r, s) = \frac{n!}{r!(n-r)!} s^r (1-s)^{n-r} \quad (2)$$

If the ion track overlapping is discussed for the unit irradiation area ($S_0 = 1 \text{ cm}^2$), n is replaced by the ion fluence, Φ , and s is replaced by the track cross section itself, S . Then, the fraction of the area for the r times track impacts in the unit area of the target, $A(\Phi, r)$ is,

$$A(\Phi, r) = \frac{\Phi!}{r!(\Phi-r)!} S^r (1-S)^{\Phi-r} \text{ for } r = 1, 2, 3, \dots, \Phi \quad (3)$$

Although the dimensions of Φ and S are cm^{-2} and cm^2 as experimental parameters, these parameters can be treated in Equation (3) and other equations in this paper as dimensionless.

For the usual irradiation experiments of the ion tracks in materials, as a value of Φ is very large and a value of S is very small, the binomial distribution function can be approximated by the following Poisson distribution function [25],

$$A(\Phi, r) = \frac{(\Phi S)^r \exp(-\Phi S)}{r!} \quad (4)$$

We have confirmed that the result calculated by the Poisson distribution function (Equation (4)) is completely the same as that calculated by the binomial distribution function (Equation (3)) in the case of the irradiation studies referred in the present report [16,21,24]. Equation (4) will be used later for the comparison of the analytical result with that by the Monte Carlo simulation.

3. Monte Carlo Method for Two-Dimensional Imaging of Ion Track Overlapping Effect

We have developed the following Monte Carlo algorithm for the present study. The target square for the Monte Carlo calculation consisted of 1000×1000 grid of cells. Its dimension was defined to be $100 \text{ nm} \times 100 \text{ nm}$ throughout this work; a single impact on the target square was equivalent to the ion fluence of 10^{10} cm^{-2} . The target was randomly bombarded with energetic ions until the ion fluence reached the given value, Φ .

Numbers of ion impacts for all cells were examined after every single bombardment to visualize the two-dimensional impact map and to calculate the fraction of ion overlapping area, $A(\Phi, r)$ for all the number of the impacts by the ion-track, r , to compare the results by using the Poisson distribution function. It is worth noting here that the analysis by using the Poisson distribution function ignores temporal decay and spatial distribution of an effect of an impact. Hence, these conditions can also be included in the Monte Carlo calculations.

All the Monte Carlo calculations were carried out with the homemade code written in LabVIEW 2019 (32 bit) for Windows.

4. Results of the Monte Carlo Simulation and the Analysis Using the Poisson Distribution Function

4.1. Amorphization of TiO_2 by High Energy Heavy Ion Irradiation

Here, we refer the result of anatase TiO_2 by Ishikawa et al. as an example of the ion irradiation induced amorphization [21]. They irradiated TiO_2 films with 230 MeV Xe^{+15} ions and measured the x ray diffraction (XRD) spectra. They have shown that the XRD peak intensity decreases in an exponential manner as a function of ion fluence. This experimental result can be explained as a result of the overlapping of amorphous ion tracks. Along the ion beam path in TiO_2 , the crystal structure becomes amorphous, and the amorphous areas still remains amorphous irrespective of the number of impacts by ion-tracks. As only the non-amorphized area contributes to the x ray diffraction, the intensity of the XRD peak is proportional to the fraction of zero impact ($r = 0$) area, $A(\Phi, 0)$, which is not amorphized by the irradiation. By using Equation (4), the relative XRD peak intensity, $I(\Phi)/I(0)$, is related to the ion-fluence, Φ , by the following function,

$$I(\Phi)/I(0) = A(\Phi, 0) = \exp(-S\Phi) \quad (5)$$

or

$$\ln(I(\Phi)/I(0)) = \ln A(\Phi, 0) = -S\Phi \quad (6)$$

where $I(0)$ is the XRD peak intensity for the unirradiated target and $I(\Phi)$, the XRD peak intensity after the irradiation with the fluence of Φ . From the slope of $\ln(I(\Phi)/I(0)) - \Phi$ plot, the cross section of the ion track, S , has been determined as 72.3 nm^2 , which corresponds to the track diameter of 9.6 nm. Because the total fraction of amorphized and non-amorphized areas is unity, the fraction of the amorphized area is expressed as,

$$1 - A(\Phi, 0) = 1 - \exp(-S\Phi), \quad (7)$$

which is well known as “the Poisson law”.

The above model that TiO_2 sample is amorphized only inside the ion -track, and the amorphized region never contributes to the XRD diffraction seems to be too simple. The previous TEM (transmission electron microscope) observations, however, clearly show that just inside the ion-track in TiO_2 , the structure becomes amorphous, and outside the ion-track, the crystal structure is maintained [26,27]. The XRD peaks for TiO_2 samples which are almost completely amorphized by high energy heavy ion irradiation are much smaller than for the unirradiated crystalline TiO_2 and are scarcely observed [22]. These experimental results, therefore, justify the above model for the analysis of ion track overlapping in TiO_2 , and the fraction of amorphized area can be estimated by the decrease in XRD peak intensity.

The result of the Monte Carlo simulation for the Xe ion induced amorphization of TiO_2 is shown in Figure 1. The figure represents the two-dimensional images of amorphized (yellow) and non-amorphized (blue) areas for the ion-fluences of 5×10^{11} , 1×10^{12} , 2×10^{12} and $2.5 \times 10^{12} \text{ cm}^{-2}$. The track diameter is 9.6 nm which has been determined by the experiment.

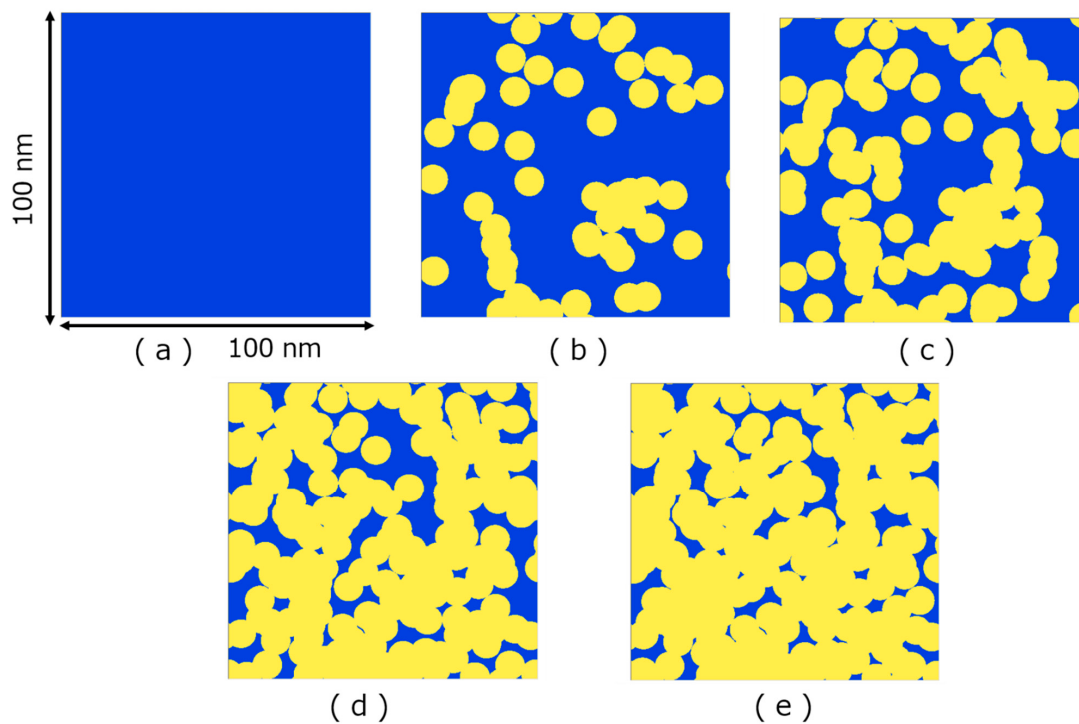


Figure 1. Two-dimensional images of amorphized (yellow) and non-amorphized (blue) areas for various ion fluences; (a) 0 cm^2 (unirradiated), (b) $5 \times 10^{11} \text{ cm}^2$, (c) $1 \times 10^{12} \text{ cm}^2$, (d) $2 \times 10^{12} \text{ cm}^2$ and (e) $2.5 \times 10^{12} \text{ cm}^2$. The diameter of ion track is assumed to be 9.6 nm.

Figure 2 shows the fraction of the non-amorphized area as a function of ion-fluence, which has been calculated from the two-dimensional images. The figure also shows the experimental result [21] and the result calculated using Equation (6). The result of the Monte Carlo simulation well agrees with the experimental result and that estimated by the Poisson distribution function. Figure 3 confirms that the ion fluence dependence of the fraction of the amorphous area is expressed by the Poisson law (Equation (7)).

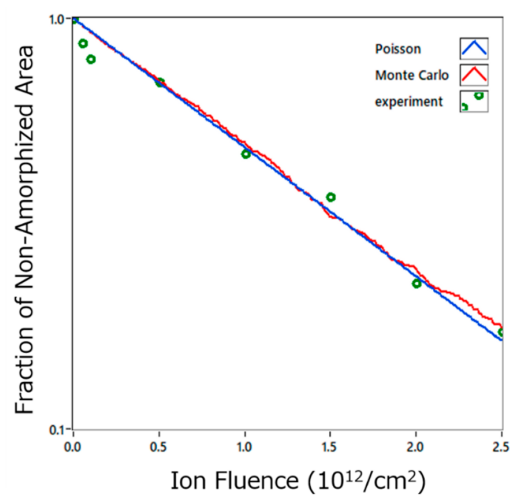


Figure 2. Fraction of non-amorphized area as a function of Xe ion fluence. Green circles, blue line, and red line represent the experimental result [21], result calculated by Equation (6) and the result from two-dimensional images, respectively. The logarithmic scale is used on the vertical axis.

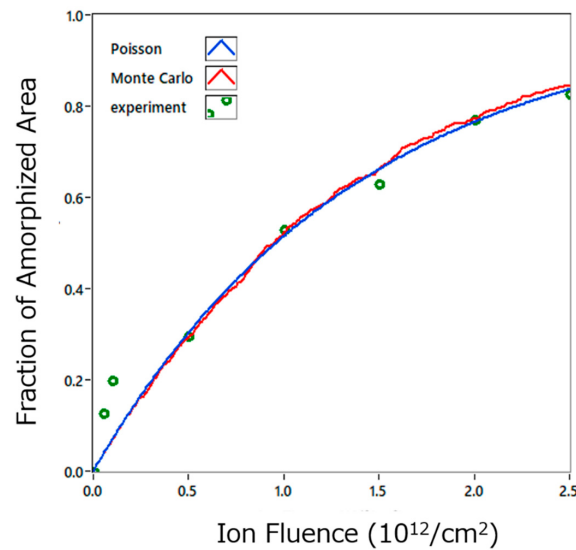


Figure 3. Fraction of amorphized area as a function of Xe ion fluence. Green circles, blue line, and red line represent the experimental result [21], result calculated by Equation (7) and the result from two-dimensional images, respectively.

4.2. Lattice Structure Change of ZrO_2 by High Energy Heavy Ion Irradiation

Next, one of the experimental results of the lattice structure transformation by high energy ion irradiation is referred. Benyagoub et al. irradiated monoclinic zirconia with 135 MeV Ni ions, and the lattice structures of the irradiated samples were characterized by XRD [16]. They have found that the lattice structure of monoclinic ZrO_2 gradually changes to the tetragonal structure by the ion irradiation. They have also shown that the evolution of the fraction of the tetragonal phase with the ion fluence shows a sigmoidal shape. This experimental result suggests that only one impact of the ion track does not cause the lattice structure transformation, but two or more impacts of the ion tracks are needed for the lattice structure transformation. This phenomenon can be explained by using the Poisson distribution function (Equation (4)) as follows. Since the total fraction of areas which are affected and not affected by ion tracks is unity:

$$\sum_{r=0}^{\Phi} A(\Phi, r) = A(\Phi, 0) + A(\Phi, 1) + \sum_{r=2}^{\Phi} A(\Phi, r) = 1. \quad (8)$$

According to Equation (4),

$$A(\Phi, 0) = \exp(-S\Phi) \quad (9)$$

and

$$A(\Phi, 1) = S\Phi \exp(-S\Phi) \quad (10)$$

Therefore, the fraction of two or more impacted areas, or the area of the tetragonal structure is,

$$\sum_{r=2}^{\Phi} A(\Phi, r) = 1 - \exp(-S\Phi) - S\Phi \exp(-S\Phi). \quad (11)$$

The result of the Monte Carlo simulation is shown in Figure 4 for the lattice structure transformation of ZrO_2 . The figure represents the two-dimensional images of monoclinic (blue) and tetragonal (yellow) areas for the ion-fluences of 2.5×10^{12} , 5×10^{12} , 1.5×10^{13} and $3 \times 10^{13} \text{ cm}^{-2}$. The track diameter is assumed to be 4.4 nm. This value was determined by the comparison of the experimental data and the result calculated by Equation (11).

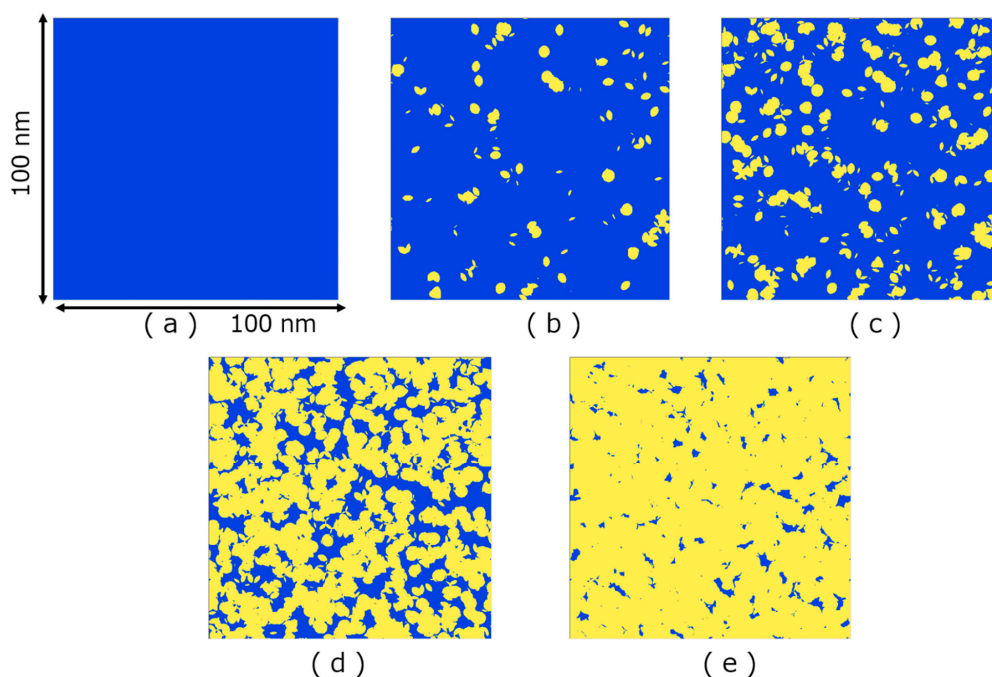


Figure 4. Two-dimensional images of tetragonal (yellow) and monoclinic (blue) areas for various ion fluences; (a) 0 cm^2 (unirradiated), (b) $2.5 \times 10^{12} \text{ cm}^2$, (c) $5 \times 10^{12} \text{ cm}^2$, (d) $1.5 \times 10^{13} \text{ cm}^2$ and (e) $3 \times 10^{13} \text{ cm}^2$. The diameter of ion track is assumed to be 4.4 nm.

Figure 5 shows the fraction of the tetragonal structure area as a function of ion-fluence, which has been calculated from the two-dimensional images. The figure also shows the experimental result and the result estimated using Equation (11). The result of the Monte Carlo simulation well agrees with the experimental result and that calculated by using the Poisson distribution function with the track cross section, S , of $1.5 \times 10^{-13} \text{ cm}^2$, or the track diameter of 4.4 nm. Benyagoub et al. have shown that the similar equation to Equation (11) can reproduce their experimental result for ZrO_2 [16]. The equation used in ref. [16] was deduced through a quite complicated manner [28]. Meanwhile, in our case, Equation (11) can be simply given as an approximated formula of the binomial distribution function.

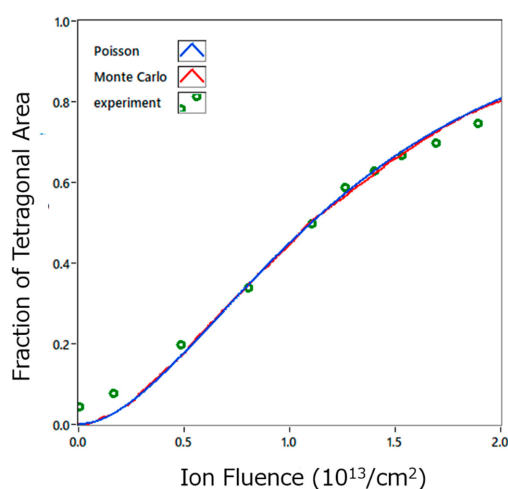


Figure 5. Fraction of tetragonal structure area of ZrO_2 as a function of ion fluence. Green circles, blue line, and red line represent the experimental result [16], result calculated by Equation (11) and the result from two-dimensional images, respectively. The value of S used in Equation (11) is $1.5 \times 10^{-13} \text{ cm}^2$ which corresponds to the track diameter of 4.4 nm.

4.3. Change in Magnetic States of CeO₂ by High Energy Heavy Ion Irradiation

Concerning the appearance of magnetism in CeO₂ at room temperature, a lot of experimental and theoretical studies have been performed [29]. Although the mechanism of the appearance of the magnetism has never been fully clarified, previous studies have suggested that defects of O anions and Ce³⁺ state of cations somehow contribute to the magnetism of CeO₂ [29]. The measurements of EXAFS (extended x-ray absorption fine structure) and XPS (X-ray photoelectron spectroscopy) using synchrotron radiation facilities have revealed that 200 MeV Xe ion irradiation induces the oxygen deficiency around Ce cations in CeO₂, and the resultant change in valence state of cations from Ce⁴⁺ to Ce³⁺ [30,31]. The change in Ce valence state due to oxygen disorders has also been confirmed by the first principles calculation [32]. The SQUID (super quantum interference device) measurement shows that the irradiation with 200 MeV Xe ions induces the ferromagnetic state in CeO₂ [24,33]. These experimental and theoretical results imply that the appearance of the magnetism is attributed to the magnetic moment of localized 4f electrons on Ce³⁺ cations. Takaki and Yasuda have shown by the TEM observations that 200 MeV Xe ion irradiation produces one-dimensional defective regions (ion-tracks) in CeO₂ samples, and that only inside the ion-tracks, the arrangement of oxygen atoms is preferentially disordered [11]. Based on such previous results, we use the following model for the analysis of the ion-track overlapping effect on the magnetic state of Xe⁺¹⁴ ion irradiated CeO₂. Only inside the ion track, disorders of oxygen atoms and the resultant Ce³⁺ valence state are produced, and the ferro magnetic state appears. Outside the ion track, the sample is still nonmagnetic. With increasing Xe ion fluence, arrangements of not only oxygen atoms, but also cerium atoms become disordered by the ion track overlapping, resulting in the decreases in magnetization [24,33]. The effect of the ion track overlapping on the magnetic states for CeO₂ irradiated with 200 MeV Xe⁺¹⁴ ions is, therefore, more complicated than the cases of TiO₂ or ZrO₂. Our previous paper has shown that if the following effect of track overlapping on the saturation magnetization is assumed, the ion fluence dependence of the saturation magnetization calculated by using the Poisson distribution function well reproduces the experimental result [24]. The saturation magnetization is $M_0 = 0$ emu/g, $M_1 = 0.1$ emu/g, $M_2 = 0.05$ emu/g, $M_3 = 0.025$ emu/g and $M_4 = 0.005$ emu/g for the non-impacted area ($r = 0$), one-impacted area ($r = 1$), two-impacted area ($r = 2$), three-impacted area ($r = 3$), and the area for four or more impacts ($r \geq 4$), respectively. From Equation (4), $A(\Phi, r)$ for $r = 0, 1, 2, 3$ and for r of 4 or more impacts, are given by,

$$\begin{aligned}
 A(\Phi, 0) &= \exp(-S\Phi) \\
 A(\Phi, 1) &= (S\Phi) \exp(-S\Phi) \\
 A(\Phi, 2) &= \frac{(S\Phi)^2}{2} \exp(-S\Phi) \\
 A(\Phi, 3) &= \frac{(S\Phi)^3}{6} \exp(-S\Phi) \\
 A(\Phi, r \geq 4) &= 1 - \sum_{r=0}^3 A(\Phi, r)
 \end{aligned} \tag{12}$$

and the irradiation induced saturation magnetization as a function of ion fluence is given as,

$$M(\Phi) = M_1 \cdot A(\Phi, 1) + M_2 \cdot A(\Phi, 2) + M_3 \cdot A(\Phi, 3) + M_4 \cdot A(\Phi, r \geq 4) \tag{13}$$

Equation (13) will be used later for the comparison with the result of the Monte Carlo simulation.

The result of the Monte Carlo simulation is shown in Figures 6 and 7 for the transition of the magnetic states of CeO₂. The figure represents the two-dimensional images of areas having different saturation magnetization for the ion-fluence of 2.5×10^{12} , 5×10^{12} , 1.5×10^{13} and 3×10^{13} cm⁻². The track diameter is 4.7 nm which has been determined by the experiment [24].

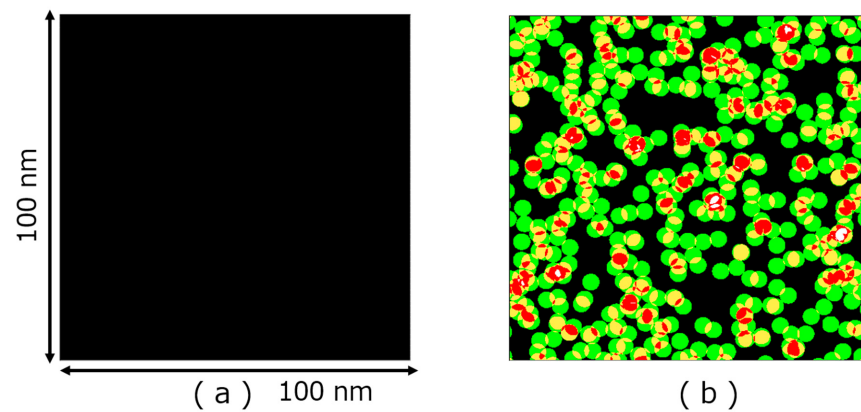
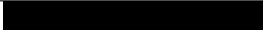


Figure 6. Two-dimensional images of irradiation-induced saturation magnetization of CeO₂ for ion fluences of (a) 0 cm² (unirradiated), and (b) 2.5×10^{12} cm². The diameter of ion track is assumed to be 4.7 nm. The correspondence relationship for colors and the values of magnetization is shown in Table 1.

Table 1. Correspondence relationship for colors in Figures 6 and 7, the number of ion track impacts and saturation magnetization.

	Color	Number of Ion Track Impacts, r	Saturation Magnetization (emu/g) Mi
black		0	0
green		1	0.1
yellow		2	0.05
red		3	0.025
white		≥ 4	0.005

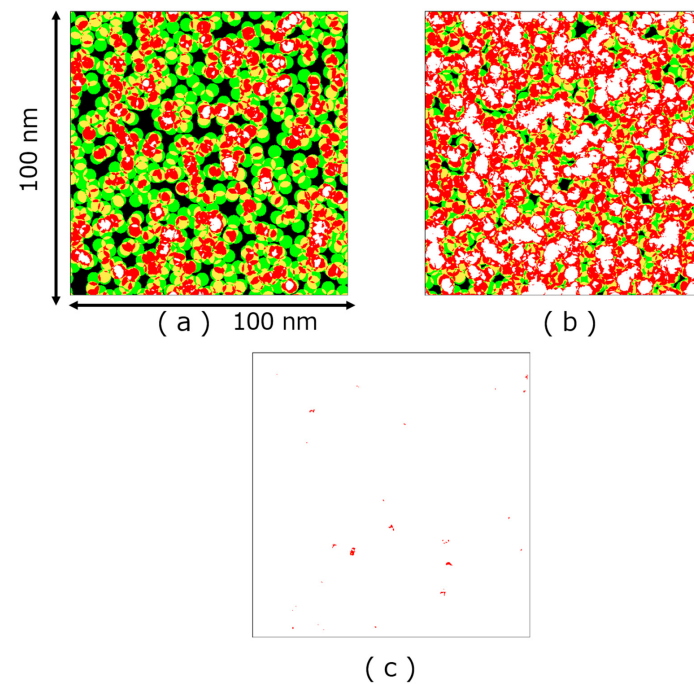


Figure 7. Two-dimensional images of irradiation-induced magnetization for ion fluences of (a) 5×10^{12} cm², (b) 1.5×10^{13} cm² and (c) 3×10^{13} cm². The diameter of ion track is assumed to be 4.7 nm. The correspondence relationship for colors and the values of magnetization is shown in Table 1.

The correspondence relationship for colors in Figures 6 and 7, the number of ion track impacts, and the saturation magnetization is shown in Table 1.

Figure 8 shows the saturation magnetization of CeO₂ as a function of ion-fluence, which has been calculated from the two-dimensional images. The figure also shows the experimental result [24] and the result calculated using Equation (13). The result of the Monte Carlo simulation well agrees with the experimental result and that calculated by using the Poisson distribution function.

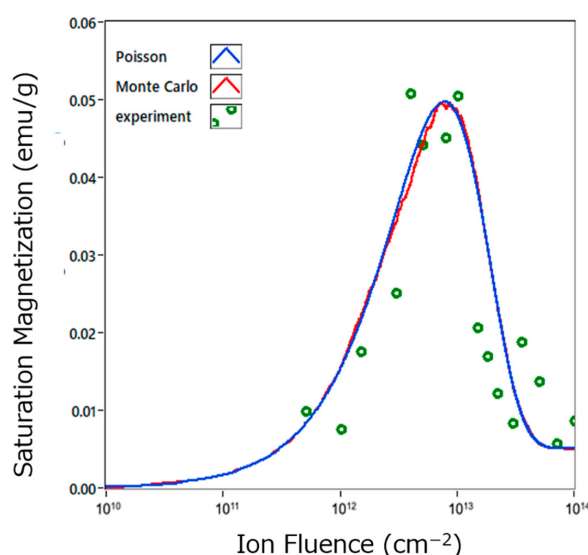


Figure 8. Saturation magnetization of CeO₂ as a function of ion fluence. Green circles, blue line, and red line represent the experimental result [24], result calculated by Equation (13) and the result from two-dimensional images, respectively. The value of S used in Equation (13) is $1.7 \times 10^{-13} \text{ cm}^2$ corresponding to the track diameter of 4.7 nm.

5. Discussion

In the previous section, by using the Monte Carlo method, we have simulated the two-dimensional images for the three kinds of the ion track overlapping effects on the oxides irradiated with high energy heavy ions. In the case of the amorphization of TiO₂, as can be seen in Figure 1, the target is dotted with disk-shaped amorphized areas with the same diameter for small ion fluence, because only one impact of the ion-track can make a target amorphous. With increasing the ion fluence, the overlapping of amorphous tracks occurs more frequently, and for higher ion fluences, most part of the target becomes amorphous. For the crystal phase transformation of ZrO₂, Figure 4 shows that the areas of the tetragonal phase, which are surrounded by the monoclinic area, appear for the small ion fluence. The tetragonal phase areas have various shapes, which are far from disk-shape. This is due to the fact that only one ion-track impact does not cause the crystal phase transformation, but two or more ion-track impacts are needed for the crystal phase transformation. As can be seen in Figure 1, Figure 4, Figure 6, and Figure 7, the overlapping of ion -tracks leads to the modulated lattice and magnetic structures with a nanometer scale. Therefore, the Monte Carlo simulation provides a good first approach for understanding the nanometer-sized two-dimensional structures of oxides, which are produced by the overlapping of the ion-tracks. Moreover, the complementary usage of the Monte Carlo simulation with some experimental techniques of precise imaging, such as TEM, AFM (atomic force microscope), MFM (magnetic force microscope), and PEEM (photo-emission electron microscope), will also be useful in order to promote the study of the effect of high energy ion irradiation in materials.

In the present report, we have only mentioned the lattice structure and magnetic property changes by the ion-track overlapping. If the electrical conductivity appears inside the

ion-tracks in insulators, and the overlapping of the ion-tracks affects their electrical conductivity, the formation of continuous electron paths or the conducting network will drastically change the macroscopic electrical property of the insulators. The two-dimensional images of ion track overlapping, which are simulated by the Monte Carlo method, may also be helpful for the understanding of such the percolation behavior in ion-irradiated insulators.

The ion-track overlapping effects have been analyzed so far by using complicated manners [20,28,34]. In the present study, the Monte Carlo method has confirmed that the effects of the ion-track overlapping can surely be described by a much simpler formula, the Poisson distribution function, which is the approximated formula of the binomial distribution function.

6. Summary

The two-dimensional images of the ion-track overlapping effects on the amorphization of TiO_2 , the crystal phase transformation of ZrO_2 , and the transition of the magnetic states of CeO_2 were simulated by the Monte Carlo method. From the two-dimensional images, the fractions of the areas which are modified by ion-tracks were calculated as a function of ion fluence. They agree well with the analytical result by using the Poisson distribution function.

Author Contributions: Monte Carlo simulation, S.N.; data analysis by Poisson distribution function, A.I.; data curation, S.N.; writing—original draft preparation, A.I.; writing—review and editing, A.I. and S.N. All authors have read and agreed to the published version of the manuscript.

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