1. Introduction

The interaction of energetic ions with solids is being investigated with a continuous interest, which is demonstrated by the numerous theoretical and experimental studies that have been published in recent years. Intensive research activity is going on with materials with pyrochlore structures because of their possible applications for the immobilization and disposal of actinides produced in nuclear plants.\cite{1,2} In contrast, these materials are very interesting from a scientific point of view, as well, as they exhibit a high chemical and structural flexibility, and their structure has a high radiation tolerance.\cite{3}

The stability of these solids under irradiation is a key problem for applications. The irradiation with high-energy heavy ions provides a suitable simulation method for investigation with respect to the long-term effect of actinides. Systematic studies would be required for making reliable predictions. However, in spite of the considerable number of publications, only a single pyrochlore \( \text{Gd}_2\text{Ti}_2\text{O}_7 \) was studied systematically in a broad range of ion energies \( E \) and electronic stopping power \( S_e \)\cite{4-6} including track formation, as well.

In this article, quantitative relationships observed in different insulators between the \( R^2 - S_e \) track evolution curves \( (R_e: \text{track radius}) \) are taken into account. These were ignored in previous analyses. Based on such observations, recently, there has been considerable progress in the understanding of track formation induced by electronic excitation.\cite{7} Concerning the effects of materials parameters (MPs), it has been shown that track formation is controlled only by the melting temperature \( T_m \) in a broad range of ion energies.\cite{7} Besides practical aspects, this is of high theoretical interest, as \( T_m \) is an equilibrium parameter; nevertheless, it controls a process under spike conditions. In this article, the validity of the model is checked for \( \text{Gd}_2\text{Ti}_2\text{O}_7 \).

The ion-induced temperature distribution \( \Delta T(r,t) \) denotes the increase in local temperature \( T \) over the irradiation temperature \( T_i \). It is one of the most important information characterizing the response of solids to the impact of the projectiles. Therefore, the reliability of theories depends on a great extent on the accuracy of the calculation of the temperature. The direct comparison with experiments is rather limited in this case and new possibilities are considered.

An emphasis is made throughout the article on the demonstration of some common features of track formation in various solids. In this respect, the initial values of the track radii \( R_m \) have exceptional importance as various subsequent processes may destroy the existing relationship between them. In this article, the track evolution is described in \( \text{Gd}_2\text{Ti}_2\text{O}_7 \) at low- and high-ion velocities, by applying the Analytical Thermal Spike Model (ATSM).\cite{8} The results in \( \text{Gd}_2\text{Ti}_2\text{O}_7 \) and other insulators are compared and scaling features are demonstrated.

2. Theoretical Background

The most widely studied irradiation effect is the formation of ion-induced tracks. It was first observed in 1959 in mica after exposing to uranium fission fragments.\cite{9} In the following more than 60 years, the theoretical efforts were concentrated in finding the right mechanism linking the physical properties of the actual target and the irradiation parameters with the track size.

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However, it was shown later that this could not lead to satisfactory results as there was found additionally a clear quantitative relationship between track radii induced in different solids by different ions of different energies.[10] These results have been incorporated into the ATSM published first in 1995.[8] In the ATSM, it is assumed that the Gaussian function is a good approximation for the ion-induced temperature distribution. Equations are derived for the threshold value $S_{ct}$ for track formation and for the track evolution function $R^2(S_e)$. [8] When ATSM was applied to track forming insulators, a simple relationship was observed between track radii measured in different insulators.[7] According to this relation, track radii do not depend on MPs other than $T_m$. Track radii are controlled by the ion-induced temperature distribution. When a quantitative relation exists between track radii, there must be a quantitative relation between the induced temperatures, either.

A systematic study of track formation led to revealing that at the moment of maximum peak temperature $T_p$, ($t = 0$), the ion-induced temperature increase $\Delta T(t=0)$ is identical in various insulators for $S_e = $ constant.[7] This universal-type temperature distribution is given by

$$\Theta(r) = \frac{f(S_e)}{3\pi w^2} \exp\left[-r^2/w^2\right]$$

where $S_e = S_e/N$ ($N$: number density of atoms), $f$ is the efficiency, $c$ is the Boltzmann constant, and $w = 4.5$ nm for insulators. In insulators, low-velocity ions induce larger tracks than high-velocity ions at $S_e = $ constant and this is called velocity effect (VE).[11] In ATSM, the efficiency $f$ is responsible for VE and $f \approx 0.17$ is valid for $E > 8$ MeV/nucleon (HI) and it may change to $f \approx 0.4$[7] in the range $E < 2$ MeV/nucleon (LO). Currently, there is no sufficient experimental information for irradiation within the range $8$ MeV/nucleon > $E > 2$ MeV/nucleon, and the shape of the transition of the efficiency between the LO and HI values is not known reliably in this range.

The presence of the efficiency $f$ in Equation (1) is the demonstration that the deposited electronic energy is not completely transferred to thermal energy. Recently, this was confirmed by direct experiments on $Y_3Fe_5O_{12}$. [12] The comparison of Equation (1) with the experiments in insulators leads to the conclusion that the spatial localization of energy deposition in the lattice is characterized by the same value $w = 4.5$ nm, whatever is $E$. The term MP is used in the sense of an individual materials property. Therefore, the common parameters $f$ and $w$ in Equation (1) are not considered as an MP in insulators, though they are obviously parameters that are related to materials.

In semiconductors, however, $w$ is an MP.

Recently, the uniform temperature distribution described by Equation (1) was checked in the analysis of experimental data on electronic sputtering. Systematic data of ten solids (amorphizable and nonamorphizable insulators and semiconductors) were analyzed assuming a thermal activation mechanism.[13] Excellent agreement was found with the experiments for all solids in a broad range of $S_e$. The results confirm the validity of Equation (1) when it claims that the ion-induced temperature does not depend on MPs. It was also found that $w = $ constant in a broad range of induced temperature.

Complete uniformity of the process of track formation is not possible as the atomic stopping power $S_e/N$ depends on the composition of the target and $S_e = $ constant is required for the uniform temperature distribution.

Actually, ATSM predicts that the track evolution can be described by the equations:[8]

$$R^2_e = w^2 \ln(S_e/S_{ct}) \quad \text{for } S_e < 2.7S_{ct}$$

$$R^2_e = \frac{w^2 S_e}{2.7S_{ct}} \quad \text{for } S_e > 2.7S_{ct}$$

$$S_{ct} = \pi \rho c(T_m - T_w)w^2$$

where $\rho$ and $c$ are the density and specific heat. Often the $\rho c = 3Nk$ approximation is used in the equations. The same equations can be derived from Equation (1) as well. In the analyses according to ATSM, a logarithmic function is fitted to the experimental data in the range $S_e < 2.7S_{ct}$ (see Equation (2)) and $w$ and $f$ are the two parameters of the model derived from this fit. In principle, $w$ and $f$ are fitting parameters, but due to specific features of this effect, they have the same values for various insulators.

When applying ATSM, this procedure cannot be avoided. When the value of $S_{ct}$ provided by another source is used in the analysis, this necessarily modifies the derived values of the $w$ and $f$ parameters and in this case Equation (2)–(4) of ATSM do not describe the track evolution any more. In a review of ATSM by Dufour and Toulemonde, such improper action was made both in the LO and in the HI ranges[14] and the equations with the derived parameters were applied to track data measured in $Y_3Fe_5O_{12}$. Earlier the same data had been already analyzed using ATSM correctly.[15] The comparison of the results of the two analyses with the original experimental data is very instructive. While the corresponding plot in the study by Szenes et al. [15] shows an excellent agreement with the data, the track sizes calculated by Dufour et al.[14] do not even remind us of the original ones. There are also several similar elementary problems with this review.[14]

It is important that Equation (2)–(4) are valid for the initial radius $R_{m1}$ of the amorphous cylindrical volume formed in the thermal spike. In agreement with the identical temperature distribution expressed by Equation (1), there is a simple unambiguous quantitative relationship between these track sizes and also between the $R^2_e-(S_e)$ track evolution curves measured in different insulators. This has been proven for a number of insulators in which $R_{m1}$ was measured.[7,16]

However, in many experiments, $R_{m1}$ cannot be estimated because of the fast processes of recrystallization or phase transformation. By now, the validity of Equation (1) has been justified for about 20 insulators. As originally each one has been selected randomly for the experiment, it is reasonable assuming that the validity extends to a much higher number of insulators including those where $R_{m1}$ cannot be measured. Actually, the validity of the composition independent, uniform temperature distribution (Equation (1)) must not depend on whether $R_{m1}$ can be measured by the applied experimental techniques or cannot. It is not obvious, what could be the reason that swift heavy ions induce
\[ \Delta T(r,0) \], according to Equation (1), in one group of insulators, where \( R_m \) can be measured, whereas in another group of insulators, where \( R_m \) cannot be measured, the same ions would induce a totally different temperature distribution depending on many MPs. Much more systematic track data are necessary for the final solution of this problem.

The problems with any theory are revealed when the predictions are compared with the experiments. In this article, such comparison is extended to irradiation experiments on Gd$_2$Ti$_2$O$_7$ and it is shown that the predictions of ATSM are in good agreement with the experiments in a broad range of \( E \) and \( S_c \). This analysis diverges from any previous one on this solid as the track evolution is described without using any individual fitting parameters and MPs, except \( T_m \). This is a rather severe condition when the accuracy and the validity of a model is checked.

### 3. Experimental Data and their Analysis

In the study by Lang et al.,\textsuperscript{[5]} a detailed experimental and theoretical study is published on track formation induced by swift heavy ions in Gd$_2$Ti$_2$O$_7$ pyrochlore. The samples were irradiated by various ions of 11.1 MeV/nucleon initial energy, and transmission electron microscopy (TEM), Raman spectroscopy, and X-ray diffraction (XRD) were used for the study of the ion-induced structures. Bright-field and high-resolution TEM methods were also used. The samples were made either by crushing the irradiated specimens or by conventional TEM specimen preparation, including polishing and ion milling. The original depth in the irradiated sample is unknown for either types of TEM specimens.

It was found that radii measured by TEM correspond to tracks consisting of an amorphous core and a disordered defect fluorite shell. However, radii derived from the XRD maxima are related only to the amorphous track core. Consequently, track radii derived from TEM experiments always exceed the sizes obtained by the XRD experiment at the same values of \( S_c \).

Following other studies,\textsuperscript{[5]} only TEM results were used in this analysis. It is a reasonable decision as there is no individual adjustable parameter in this model. Therefore, it is more sensitive to the uncertainties of the experimental data than other models. For the same reason, there is always an endeavor to avoid including those data in the analysis for which the parameters of the model (\( T_m \), \( f \), \( N \)) or the experimental conditions (\( E \), \( S_c \)) are not well defined.

Similarly, those track data could not be used when inhomogeneous track structures were reported and there was no information about the initial track size. A practical solution of the problem is when the measurement of the total track radius is possible as it is close to the initial amorphous size with \( R_m \). This was done in the study by Lang\textsuperscript{[5]} for Gd$_2$Ti$_2$O$_7$ providing a possibility for the application of ATSM.

A further critical point is that there are no suitable systematic track measurements on pyrochlores other than Gd$_2$Ti$_2$O$_7$. Obviously, it is very disadvantageous for the application of any model, but especially those without individual adjustable parameters, when only a single track size is known. The uncertainty of the derived parameters is considerably increased in such cases. This was the reason that some experimental data had to be ignored.

Though this limitation reduces the available database, however, the unambiguous demonstration of a close quantitative relationship between track radii and Equation (1) would not be possible without such limitations.

In the experiment in the study by Lang et al.,\textsuperscript{[5]} the effect of the ion velocity changes in the 40 \( \mu \)m-thick samples as the ion energy varies in the range 11.1 MeV/nucleon \( < E < 2 \) MeV/nucleon along the trajectory.\textsuperscript{[5]} While \( S_c \approx \) constant along the trajectory, track radii change by more than 50% because of the VE. Different values of \( R_c \) could be obtained in this experiment depending on whether the original position of an actual TEM sample was closer to the front side (\( E = 11.1 \) MeV/nucleon, HI condition) or to the back side (\( E = 2 \) MeV/nucleon, LO condition) in the thick specimen. Thus, the experimental method led to an additional minimum \( \pm 25\% \) error in the track diameters in ref. [5]. According to the preceding considerations, VE is a source of considerable uncertainty for track studies in these types of irradiation experiments.

In another experiment, Gd$_2$Ti$_2$O$_7$ was irradiated by 120 MeV U ions.\textsuperscript{[4]} The irradiations were conducted with HI ions in the study by Lang et al.\textsuperscript{[5]} All data in the study by Jozwik-Biala\textsuperscript{[4]} belonged to the low-velocity range with \( E < 2 \) MeV/nucleon. Besides monoatomic irradiation by U ions, Gd$_2$Ti$_2$O$_7$ samples were also irradiated by 30 MeV C$_{60}$ ions that completed the LO condition.\textsuperscript{[16]} The experiments in these studies\textsuperscript{[4,5,16]} were conducted in a broad range of \( S_c \) (6–54 keV nm$^{-1}$) and \( E \) (0.04–11 MeV/nucleon).

In Figure 1, track radii induced by HI and LO projectiles\textsuperscript{[4,16]} in Gd$_2$Ti$_2$O$_7$ are shown in a normalized plot where \( r_c = 3Nk \) is used. The internal structure of the tracks is ignored here and the experimental \( R_c \) radii with an amorphous core+defect fluorite shell structure are used as discussed earlier. In the figure, the solid lines are the predictions according to Equation (2) with \( w = 4.5 \) nm, \( f = 0.17 \) for HI irradiations (\( E > 8 \) MeV/nucleon), and \( f = 0.4 \) for LO irradiations (\( E < 2 \) MeV/nucleon). Equation (2) is valid for \( S_c < 2.7S_m \) and predicts an \( R_c^2 \sim \omega^2 \ln S_c \) dependence in agreement with the experiments. When \( S_c \) is higher, \( R_c \) cannot be directly obtained from

\[
R_c^2 \sim \omega^2 \ln S_c
\]

\[
\left( \langle s_d \rangle / \sqrt{\pi \kappa w^2 (T_m - T_p)} \right)
\]

Figure 1. Variation of the track size in Gd$_2$Ti$_2$O$_7$ with the atomic stopping power (\( s_d \)); \( R_c \) and \( k \) are the track radius and the Boltzmann constant, \( w = 4.5 \) nm, and \( T_m = 2093 \) K\textsuperscript{[5,16]} is the melting and the irradiation temperatures. The irradiation by high-velocity projectiles is denoted by full symbols. The solid and dashed lines are drawn according to Equation (2) and (5), respectively, without fitting parameters.
Equation (2) as the maximum radius of the melt is reached at \( t > 0 \) during the cooling phase.\(^{[8]}\) In this case, Equation (3) and (4) are combined leading to

\[
R_s^2 = \frac{1}{2.73} \frac{f S_e}{3kN(T_m - T_r)} \quad \text{for } \frac{S_e}{S_0} > 2.7 \quad (5)
\]

It is remarkable that Equation (5) does not depend on \( w \). The same is true for Equation (3) as Equation (4) is always valid in ATSM. This was not realized in the review by Dufour et al.\(^{[14]}\) when the value of \( w \) was estimated using exclusively Equation (3) and ignoring Equation (4). This led to a considerable error.

Equation (5) is valid at high values of \( S_e \) and the dashed curve in Figure 1 was calculated by applying it. The good agreement between the data (\( \Delta R_s/R_s \approx 5\% \), \( \Delta S_e/S_e \approx 5\% \)) and the theoretical predictions for irradiation by LO ions including \( C_{60} \) irradiation when \( S_e \) and \( E \) are known accurately is emphasized.\(^{[4,16]}\) The agreement was achieved when \( S_e \) varied by an order of magnitude and no individual fitting parameters were used. This verifies that Equation (2)–(5) provide an accurate description of the track evolution in Gd\(_2\)Ti\(_2\)O\(_7\) that depends only on a single MP, \( R_m \), and the irradiation temperatures; \( k \) is the Boltzmann constant. The values for Gd\(_2\)Ti\(_2\)O\(_7\) are present results.

The comparison of the track data from the study by Lang et al.\(^{[5]}\) with the predicted HI curve shows lower than the expected uncertainty in the positions of TEM samples in the thick specimen. The figure suggests that except the largest track at \( S_e = 40.1 \text{ keV nm}^{-1} \), the original positions of the TEM samples were occasionally in that half of the irradiated specimen which was close to the impact of the projectile. The agreement of the predicted curve with the experimental data in the HI range is satisfactory when the possible consequences of the thick samples in ref. [5] (leading to \( \pm 50\% \) uncertainty for \( R_s^2 \)) are taken into account. As the random selection of the TEM samples along the thickness in ref. [5] and the existence of VE in Gd\(_2\)Ti\(_2\)O\(_7\) are without doubts, the above explanation seems to be reasonable. Thus, this high error is simply the consequence of VE and it is not related to the estimates using ATSM.

When Equation (1) is applied in Figure 1, \( fS_e = E_f \) is obtained for \( R_s = 0 \), where \( E_f = 3\pi Nkw^2(T_m - T_r) \) is the energy necessary for increasing the maximum temperature of the track to \( T_m \). Estimates for the thresholds are \( S_0 = 14.4 \text{ keV nm}^{-1} \) (HI), \( f = 0.17 \), \( T_m = 2093 \text{ K} \), \( 1/E_f = 0.408 \text{ nm keV}^{-1} \) and \( S_e = 6.13 \text{ keV nm}^{-1} \) (LO), \( f = 0.4 \), \( 1/E_f = 0.408 \text{ nm keV}^{-1} \).

In Figure 2, the normalized threshold values for track formation \( \langle s_0 \rangle \) are shown for various insulators. The comparison of the present results with the data in other insulators in Figure 2 shows that these estimates for Gd\(_2\)Ti\(_2\)O\(_7\) nicely fit the lines in the figure and verify their reliability. According to Equation (4), the slope of the line is \( m = \pi w f/3f \) in Figure 2 (\( w = 4.5 \text{ nm} \) and \( f = 0.17/0.4 \)), which is in excellent quantitative agreement (within 5%) with the experiments.

It is an interesting feature of the plot in Figure 2, that it can be used for estimates of the value of \( S_e \) for transformed tracks as well as they cannot skip this stage of formation.\(^{[15]}\)

The relationship between track sizes is demonstrated by another plot. When Equation (2) and (4) are combined, the result predicts that the \( R_s^2 - \langle s_0 \rangle \) track evolution curves follow the same lines in an \( R_s^2 - f/S_e/3kw^2(T_m - T_r) \) plot.\(^{[7]}\) In Figure 3, such a plot is shown for tracks induced by HI ions (SrFe\(_{12}\)O\(_{19}\))\(^{[17]}\) MgFe\(_2\)O\(_4\),\(^{[18]}\) NiFe\(_2\)O\(_4\),\(^{[19]}\) ZrSiO\(_4\),\(^{[20]}\) Y\(_2\)O\(_3\)(\(^{[21]}\)), together with data induced by LO ions (mica\(^{[7]}\), Y\(_2\)Al\(_5\)O\(_{12}\),\(^{[22]}\) TeO\(_2\),\(^{[23]}\) Y\(_2\)Fe\(_2\)O\(_{12}\),\(^{[11,24]}\) Al\(_2\)O\(_3\),\(^{[15]}\) KTiOPO\(_4\),\(^{[25]}\) LiNbO\(_3\)).\(^{[26]}\) The lines cross the axis at \( \langle s_0 \rangle/3kw^2(T_m - T_r) = 1/f \) providing \( f \approx 0.4 \) (LO) and \( f \approx 0.17 \) (HI). The figure proves that track formation proceeds identically in these solids including Gd\(_2\)Ti\(_2\)O\(_7\). This means that similar to the plot in Figure 1, the same description of the track evolution is valid for solids in Figure 3, as well.

It is noted that in Figure 3 the track sizes with highest deviations were measured in Y\(_2\)Fe\(_2\)O\(_{12}\) HI samples having thicknesses typically over 60 \( \mu \)m in the experiments. These high deviations are attributed partly to VE along the track length, leading to the formation of larger tracks approaching the backside of the samples. VE had a similar effect also in Gd\(_2\)Ti\(_2\)O\(_7\) as 40 \( \mu \)m-thick samples were irradiated, as in the study by Lang et al.\(^{[5]}\)

An unexpected relationship between track radii measured in different insulators is shown in Figure 4 where the appropriate
We note that, previously, track evolution has been already analyzed in Gd$_2$Ti$_2$O$_7$, applying the i-TS model requiring an individual adjustable parameter and the knowledge of a considerable number of MPs under spike conditions and assuming a superheating scenario. The advantage of the description given by ATSM seems to be without doubt as it is valid in a broader range of $S_c$ by applying only a single MP without any individual fitting parameter.

4. Discussion

4.1. The Role of MPs

According to the thermal spike philosophy, the most important information is the accurate description of the ion-induced temperature. ATSM offers a common solution for insulators, whereas it ignores the heat of fusion $L$ and other MPs. Otherwise, the contribution of $L$ might affect considerably the temperatures. In a recent paper, this problem was discussed in detail and most of the available systematic experimental track data were analyzed. The results confirmed that $L$ might have only a minor effect on track formation. The present analysis of track evolution in Gd$_2$Ti$_2$O$_7$ also refutes the usual argument according to which the good agreement with the experimental track sizes is a sufficient evidence for the validity of a model. In this case, it would not be possible that several models could lead to equally good agreement with a given set of data, while only one of the proposed models may be valid. The correct model must provide a complete description of the irradiation effect and explain beyond the track sizes the existing relations between track formations in different solids (e.g., Figure 2–4).

In this article, the theoretical lines in Figure 1 are drawn in agreement with Equation (1), which does not contain any contribution of $L$. As $L/E_f$ varies in a broad range in insulators, this large variation ought to lead to considerable shifts of $S_c$ (crossing of the lines with the X-axis) to higher values in insulators. Thus, the induced temperature would reach $T_m$ by the fast cooling and solidification of the melted cylindrical volume along the trajectory of the projectiles.

It is emphasized that the relationship in Figure 4 has been predicted using ATSM. For deriving this plot, Equation (2) and (4) were combined and a plot was drawn at $S_c = 0.3 \times 10^5$ and $7.5 \times 10^2 \text{ nm}^2 \text{ K}$, respectively. This is a remarkable important experimental evidence shown by this plot that MPs apart from $T_m$ in insulators irradiated by low- ($E < 2 \text{ MeV/nucleon}$) and high-velocity ($E > 8 \text{ MeV/nucleon}$) ions at $S_c = 3.42 \times 10^5$ and $3.61 \times 10^5 \text{ nm}^2 \text{ K}$, respectively; for track data, see the study by Szenes.

It is emphasized that the message of Figure 4 is not modified by any individual adjustable parameter and the knowledge of a considerable number of MPs under spike conditions and assuming a superheating scenario. The advantage of the description given by ATSM seems to be without doubt as it is valid in a broader range of $S_c$ by applying only a single MP without any individual fitting parameter.

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4. Discussion

4.1. The Role of MPs

According to the thermal spike philosophy, the most important information is the accurate description of the ion-induced temperature. ATSM offers a common solution for insulators, whereas it ignores the heat of fusion $L$ and other MPs. Otherwise, the contribution of $L$ might affect considerably the temperatures. In a recent paper, this problem was discussed in detail and most of the available systematic experimental track data were analyzed. The results confirmed that $L$ might have only a minor effect on track formation. The present analysis of track evolution in Gd$_2$Ti$_2$O$_7$ also refutes the usual argument according to which the good agreement with the experimental track sizes is a sufficient evidence for the validity of a model. In this case, it would not be possible that several models could lead to equally good agreement with a given set of data, while only one of the proposed models may be valid. The correct model must provide a complete description of the irradiation effect and explain beyond the track sizes the existing relations between track formations in different solids (e.g., Figure 2–4).

In this article, the theoretical lines in Figure 1 are drawn in agreement with Equation (1), which does not contain any contribution of $L$. As $L/E_f$ varies in a broad range in insulators, this large variation ought to lead to considerable shifts of $S_c$ (crossing of the lines with the X-axis) to higher values in insulators.

It is emphasized that the relationship in Figure 4 has been predicted using ATSM. For deriving this plot, Equation (2) and (4) were combined and a plot was drawn at $S_c = 0.3 \times 10^5$ and $7.5 \times 10^2 \text{ nm}^2 \text{ K}$, respectively. This is a remarkable important experimental evidence shown by this plot that MPs apart from $T_m$ in insulators irradiated by low- ($E < 2 \text{ MeV/nucleon}$) and high-velocity ($E > 8 \text{ MeV/nucleon}$) ions at $S_c = 3.42 \times 10^5$ and $3.61 \times 10^5 \text{ nm}^2 \text{ K}$, respectively; for track data, see the study by Szenes.

It is emphasized that the message of Figure 4 is not modified by any individual adjustable parameter and the knowledge of a considerable number of MPs under spike conditions and assuming a superheating scenario. The advantage of the description given by ATSM seems to be without doubt as it is valid in a broader range of $S_c$ by applying only a single MP without any individual fitting parameter.

We note that, previously, track evolution has been already analyzed in Gd$_2$Ti$_2$O$_7$, applying the i-TS model requiring an individual adjustable parameter and the knowledge of a considerable number of MPs under spike conditions and assuming a superheating scenario.

The advantage of the description given by ATSM seems to be without doubt as it is valid in a broader range of $S_c$ by applying only a single MP without any individual fitting parameter.
show a close relationship between \( S_e \) and \( T_m \) in Figure 2. \( S_e \) is related to a much higher superheating temperature, \( T_{ps} > T_m \), in the study by Lang et al.\(^5\) This is the reason that the temperature is supposed to be raised to \( T_m \) at a much lower value of \( S_e \) in that model.

A possible origin of the discrepancy might be that the estimates for superheating were made for solids without taking into account the ion-induced transient processes and the highly damaged structure of the targets. These specific features may affect considerably the proceeding of various processes including phase transition.

As the reliable knowledge of the temperature is the basis for any calculation in the spike, therefore, various estimates made for the irradiated Gd\(_2\)Ti\(_2\)O\(_7\) are problematic in the study by Lang et al.\(^5\) The same is valid for all those solids whose data are included in Figure 3 and 4 and similar plots. It is important in this respect that while Y\(_3\)Fe\(_5\)O\(_{12}\) is an emblematic insulator for i-TS, the most popular model denying scaling properties of ion-induced tracks,\(^29\) nevertheless, its experimental data fit well with other ones in all those figures which demonstrate the interdependence of tracks. This means that the \( R_c^2 - S_e \) track evolution curve can be calculated easily for Y\(_3\)Fe\(_5\)O\(_{12}\) and other solids in Figure 3 and 4 using track data of any other insulator in these figures. To do that only \( T_m \) values must be known, and other MPs are indifferent. This has been demonstrated in the study by Szennes,\(^7\) and agreement between the calculated and measured values was found within experimental error. In the same time, up to ten MPs are required for the description of the same tracks in Y\(_3\)Fe\(_5\)O\(_{12}\)\(^29\) and Gd\(_2\)Ti\(_2\)O\(_7\).\(^3\) However, it remains an open question whether solutions in other studies\(^5,29\) can be considered as a complete solution of the problem when the interdependence of these tracks is not even mentioned.

The possibility of these quite different two solutions is provided by the fact that the width of \( \Delta T(r,0) \) can be checked by track measurements only at a single temperature, \( T = T_m \). The shape of \( \Delta T(r,0) \) cannot be controlled at any other temperature. Thus, a great number of various distributions with the same width at \( T = T_m \) satisfy this simple condition and any deviation from real distribution at \( T \neq T_m \) is indifferent for the agreement with experiments. However, when a quantitative relationship is valid between track radii in different solids, then the width of \( \Delta T(r,0) \) is also determined at several temperatures (see Figure 4); thus, its shape can be checked in several points. These considerations may be useful for a correct understanding of an agreement between measured and calculated values.

The results indicate that the correct estimation of the temperature is highly important and indispensable in cases when a quantitative comparison is possible with experiments. The plots in Figure 2–4 and the accurate prediction of the \( R_c^2 - S_e \) track evolution curve of Gd\(_2\)Ti\(_2\)O\(_7\) are sound evidences confirming the validity of Equation (1). Up to now, the meaning of these figures has not been discussed and explained in the past 15 years using any other model. Thus, the readers have no information on how the various models comply with the new experimental limitations.

We see that some important features of track formations can be obtained directly from Equation (1) and Figure 2–4. Evidently, the specific features of the amorphous track core + disordered shell with a defect fluorite structure in Gd\(_2\)Ti\(_2\)O\(_7\) cannot be derived from Equation (1). This is a very complex nanostructural problem.\(^3\) Its discussion is beyond the scope of this article.

5. Conclusion

When initial track radii are known in insulators, common scaling features show up and \( T_m \) is the only MP which controls the track size. These scaling features include 1) \( \langle S_e \rangle \propto (T_m - T_e) \) in Figure 2 for the threshold atomic stopping power; 2) common \( R_c^2 - \langle S_e \rangle / (T_m - T_m) \) track evolution curves in Figure 3; and 3) common \( R_c - T_m - T_e \) Gaussian curve controlled by \( T_m \) in Figure 4, connecting track data measured in various solids for \( \langle S_e \rangle = \text{constant} \).

The validity of these relations is supported by experimental data on ion-induced tracks measured in various insulators including Y\(_3\)Fe\(_5\)O\(_{12}\). This is a serious contradiction as this solid is an emblematic prototype for the i-TS model. The scaling features are also demonstrated in Gd\(_2\)Ti\(_2\)O\(_7\) where the measured \( R_c \) total track radii with an amorphous core + defect fluorite shell structure are close to the initial radii. The experimental \( R_c^2 - S_e \) values satisfy the equations of ATSM for a very broad spectrum of \( S_e (6-54 \text{ keV nm}^{-1}) \) and \( E (0.04-11 \text{ MeV/nucleon}) \) without applying any individual fitting parameter and without MPs except \( T_m \). LO and HI tracks in Gd\(_2\)Ti\(_2\)O\(_7\) are included in Figure 4 that demonstrates the interdependence of tracks induced in different insulators when \( \langle S_e \rangle = \text{constant} \).

The results on Gd\(_2\)Ti\(_2\)O\(_7\) confirm the validity of Equation (1), claiming that the ion-induced temperature distributions agree quantitatively in numerous insulators for identical values of \( \langle S_e \rangle \). The estimates of the thresholds for Gd\(_2\)Ti\(_2\)O\(_7\) are \( S_e = 6.13 \text{ and } 14.4 \text{ keV nm}^{-1} \) for LO and HI projectiles, respectively, in agreement with the values derived from the scaling properties. The heat of fusion \( L \) may have only minor if any effect on the track size. Thus, the assumption of the superheating mechanism is not justified by the experiments. It is a source of a systematic error in the calculation of the ion-induced temperatures that may exceed even 50%.

Conflict of Interest

The author declares no conflict of interest.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

Keywords

heavy ions, pyrochlore, thermal spike, tracks, velocity effect

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\[1\] R. C. Ewing, W. J. Weber, J. Lian, J. Appl. Phys. 2004, 95, 5949.
[2] A. Chroneos, M. J. D. Rushton, C. Jiang, L. H. Tsoukalas, J. Nucl. Mater. 2013, 441, 29.
[3] J. Shamblin, C. L. Tracy, R. I. Palomares, E. C. O'Quinn, R. C. Ewing, J. Neuhefeind, M. Feygenson, J. Behrens, C. Trautmann, M. Lang, Acta Mater. 2018, 144, 60.
[4] I. Jozwik-Biala, J. Jagielski, L. Thomé, B. Arey, L. Kovarik, G. Sattonnay, A. Debelle, I. Monnet, Nucl. Instrum. Methods B 2012, 286, 258.
[5] M. Lang, M. Toulemonde, J. Zhang, F. Zhang, C. L. Tracy, J. Lian, Z. Wang, W. J. Weber, D. Severin, M. Bender, C. Trautmann, R. C. Ewing, Nucl. Instrum. Methods B 2014, 336, 102.
[6] S. Moll, G. Sattonnay, L. Thomé, J. Jagielski, C. Legros, I. Monnet, Nucl. Instrum. Methods B 2010, 268, 2933.
[7] G. Szenes, Nucl. Instrum. Methods B 2013, 312, 118.
[8] G. Szenes, Phys. Rev. B 1995, 51, 8026.
[9] E. C. M. Silk, R. S. Barnes, Philos. Mag. 1959, 4, 970.
[10] G. Szenes, Radiat. Eff. Defects Solids 2007, 162, 557.
[11] A. Meftah, F. Brisard, J. M. Costantini, M. Hage-Ali, J. P. Stoquert, M. Toulemonde, Phys. Rev. B 1993, 48, 920.
[12] G. Szenes, L. Toth, Phys. Scr. 2019, 94, 115810.
[13] G. Szenes, Phys. Scr. 2021, 96, 035703.
[14] C. Dufour, M. Toulemonde, in Ion Beam Modification of Solids (Eds: W. Wesch, E. Wendler), Springer Series in Surface Sciences, Vol. 61, Springer, Cham 2016, pp. 63–104.
[15] G. Szenes, J. Nucl. Mater. 2005, 336, 81.
[16] J. M. Zhang, M. Toulemonde, M. Lang, J. M. Costantini, S. Della-Negra, R. C. Ewing, J. Mater. Res. 2015, 30, 2456.
[17] C. Houpert, F. Studer, H. Pascard, J. Y. Fan, M. Toulemonde, Nucl. Tracks Radiat. Meas. 1991, 19, 85.
[18] M. Toulemonde, S. Bouffard, F. Studer, Nucl. Instrum. Methods B 1994, 91, 108.
[19] M. Toulemonde, F. Studer, Solid State Phenom. 1993, 30/31, 477.
[20] L. A. Bursill, G. Braunhausen, Philos. Mag. A 1990, 62, 395.
[21] S. Hemon, A. Berthelot, C. Dufour, F. Courbilleau, E. Dooryhée, S. Bégin-Colin, E. Paumier, Eur. Phys. J. B 2001, 19, 517.
[22] M. Izerrouken, A. Meftah, M. Nekkab, Nucl. Instrum. Methods B 2007, 258, 395.
[23] G. Szenes, F. Pászti, Á. Péter, A. I. Popov, Nucl. Instrum. Methods B 2000, 166–167, 949.
[24] J. Jensen, A. Dunlop, S. Della-Negra, M. Toulemonde, Nucl. Instrum. Methods B 1998, 146, 412.
[25] T. Opfermann, T. Höche, S. Klaumünzer, W. Wesch, Nucl. Instrum. Methods B 2000, 166, 954.
[26] B. Canut, S. M. M. Ramos, Radiat. Eff. Defects Solids 1998, 145, 1.
[27] I. Jozwik-Biala, J. Jagielski, B. Arey, L. Kovarik, G. Sattonnay, A. Debelle, S. Mylonas, I. Monnet, L. Thome, Acta Mater. 2013, 61, 4669.
[28] G. Szenes, Radiat. Eff. Defects Solids 2020, 175, 241.
[29] M. Toulemonde, W. Assmann, C. Dufour, A. Meftah, F. Studer, C. Trautmann, Mat. Fys. Med. 2006, 52, 263.
[30] G. Szenes, Nucl. Instrum. Methods B 2012, 280, 88.