

# Ion track formation in amorphous SiO<sub>2</sub>: a low velocity study

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## Abstract

Ion track formation in amorphous SiO<sub>2</sub> was investigated using infrared spectroscopy. For comparison, one set of samples was also irradiated using 1.25 MeV gamma rays. Increase of 1044 cm<sup>-1</sup> peak and decrease of 1078 cm<sup>-1</sup> peak was observed in all cases. Experimental results were analyzed using analytical thermal spike model and non-standard model parameters were found. This finding is attributed to the amorphous structure of the a-SiO<sub>2</sub> material.

**Keywords:** swift heavy ion, ion track, thermal spike, amorphous SiO<sub>2</sub>, infrared spectroscopy, gamma rays

## 1. Introduction

Permanent damage formed after the passage of a swift heavy ion (SHI) through a solid material is called the ion track [1–6]. Most common description of the ion track formation, the thermal spike model, proposes that the kinetic energy of the projectile, deposited as dense electronic excitation along the ion trajectory, can lead to melting of the material. In this case, ion tracks originate from the quenching of the molten phase on the nanosecond time scale, resulting in an amorphous inclusion. In crystal materials, ion tracks can be readily observed with various techniques like electron microscopy or Rutherford backscattering spectrometry in channeling [5,7,8]. Atomic force microscopy has also been found suitable for direct imaging of ion tracks on the surface after grazing SHI irradiations [9–16]. However, in amorphous materials, investigation of ion tracks is much more difficult due to the weak contrast between the ion track and the surrounding material, hence very few techniques like infrared (IR) spectroscopy and small angle X-ray scattering (SAXS) were found

suitable for this purpose [17–19].

Response of the amorphous  $\text{SiO}_2$  (a- $\text{SiO}_2$ , vitreous silica) to SHI irradiation has been thoroughly studied in the past because of the material's importance for industrial applications in electronics and photonics. Furthermore, a- $\text{SiO}_2$  is a role model for glass structures and therefore important for studies where similar materials are exposed to fission fragment damage during nuclear waste immobilisation [20]. Optical fibers based on a- $\text{SiO}_2$  can also be used for the distributed measurements of high temperatures and high radiation levels at various nuclear facilities [21,22]. Unlike crystalline materials that can undergo large structural changes due to exposure to high levels of radiation [23,24], amorphous materials should not alter much since their structure remains amorphous [25]. Therefore, ion track formation in a- $\text{SiO}_2$  due to SHI irradiation has been studied for many years now [17,18,26–33] and recently a review has been published [20]. Other SHI irradiation effects in a- $\text{SiO}_2$  have also been researched thoroughly, most notably plastic deformation (ion hammering effect) [34–38] and nanoparticles modification/synthesis [19,39–49], but also ion track etching [50–52], surface nanostructuring [11,53–55] and sputtering [56–60].

Often are these experimental results described using thermal spike models [18,20,32,35,61–66] which can quantitatively explain various observed features. Some difficulties, for example the role of self-trapped excitons, remained unclear in the ion track formation processes [3,6,64,65,67,68]. Also, theoretical efforts indicated that a significant amount of SHI deposited energy remains stored in the valence holes within the ion tracks in a- $\text{SiO}_2$  and is released only on the picosecond time-scale via Auger processes [69–72]. Experimental and theoretical efforts investigating these early stages of ion track formation via detection of SHI induced X-rays and secondary electrons are still ongoing [73–77]. It might be that these features of ion track formation in a- $\text{SiO}_2$  led some authors to apply diverse, non-standard parameters in thermal spike calculations [19,48,65,78].

Aim of this work is to obtain new experimental data from which thermal spike parameters for a- $\text{SiO}_2$  can be established. To accomplish this, we have performed low velocity SHI irradiations ( $E/A < 2 \text{ MeV/amu}$ ) of the samples and analysed them using IR spectroscopy. Additionally, we have performed high dose gamma ray (GR) irradiations and characterised the samples with the same method. This way, correlation in radiation effects between SHI and GR irradiation was established. In this work we follow well-established approach of using IR spectroscopy to characterise ion tracks in a- $\text{SiO}_2$  [17,18,26–28,32]. Evolution of the TO3 band seen as a decrease of  $1078 \text{ cm}^{-1}$  peak and increase of  $1044 \text{ cm}^{-1}$  peak has been observed with increasing SHI fluence. These changes have been

attributed to the decrease of the O-Si-O bond angle from  $144^\circ$  to  $129^\circ$  due to compaction of silica [28]. Microscopically, origin of this structural damage has been found in the change of  $\alpha\text{-SiO}_2$  from large (six member) ring structure to small (planar three and four member) ring structure [18,28,29]. It has also been experimentally observed that the absorption band attributed to LO4-TO4 pair at  $1165\text{-}1200\text{ cm}^{-1}$  does not change upon irradiation [18]. Ion track sizes evaluated from observed kinetics have been found in a very good agreement with SAXS data [17,18] and consistent with track etching results [52]. This enables comprehensive analysis of the ion track data in  $\alpha\text{-SiO}_2$  both from previous and present study.

## **2. Experimental details**

Thermally grown  $\alpha\text{-SiO}_2$  film on Si wafer and having 200 nm thickness was purchased from Crystec (Berlin, Germany). Oxide was grown on both sides of the Si wafer that was cut into roughly  $1\times 1\text{ cm}^2$  pieces before irradiation. No further sample preparation was done.

First set of samples was irradiated at normal incidence using 1 MeV O, 3 MeV O and 23 MeV I delivered by 6 MV EN Tandem Van de Graaff accelerator at the Ruđer Bošković Institute (RBI, Zagreb, Croatia). Second set of samples was irradiated at  $30^\circ$  off normal using 167 MeV Xe beams delivered by cyclotron at Joint institute for Nuclear Research (JINR, Dubna, Russia). Fluences applied were up to  $3\times 10^{15}$  ions/ $\text{cm}^2$  for oxygen beams, up to the  $3\times 10^{13}$  ions/ $\text{cm}^2$  for the iodine beam, and up to the  $10^{13}$  ions/ $\text{cm}^2$  for the xenon beam. Third set of samples was exposed to 1.25 MeV GR irradiation up to a dose of 90 MGy using panoramic  $^{60}\text{Co}$  source at the RBI.

For the sample analysis, IR spectroscopy was performed using PerkinElmer Spectrum GX spectrometer equipped with DTGS (Deuterated TriGlycine Sulfate) detector. FTIR spectra were recorded in normal transmission mode averaging 20 scans over the range  $400\text{-}2000\text{ cm}^{-1}$ . Before scanning the samples the background was recorded collecting 50 scans with a spectral resolution of  $4\text{ cm}^{-1}$ .

## **3. Results**

For a given SHI beam, samples were irradiated with different fluences and following this IR spectra were measured. The spectra were subsequently baseline corrected. As an example, IR spectrum

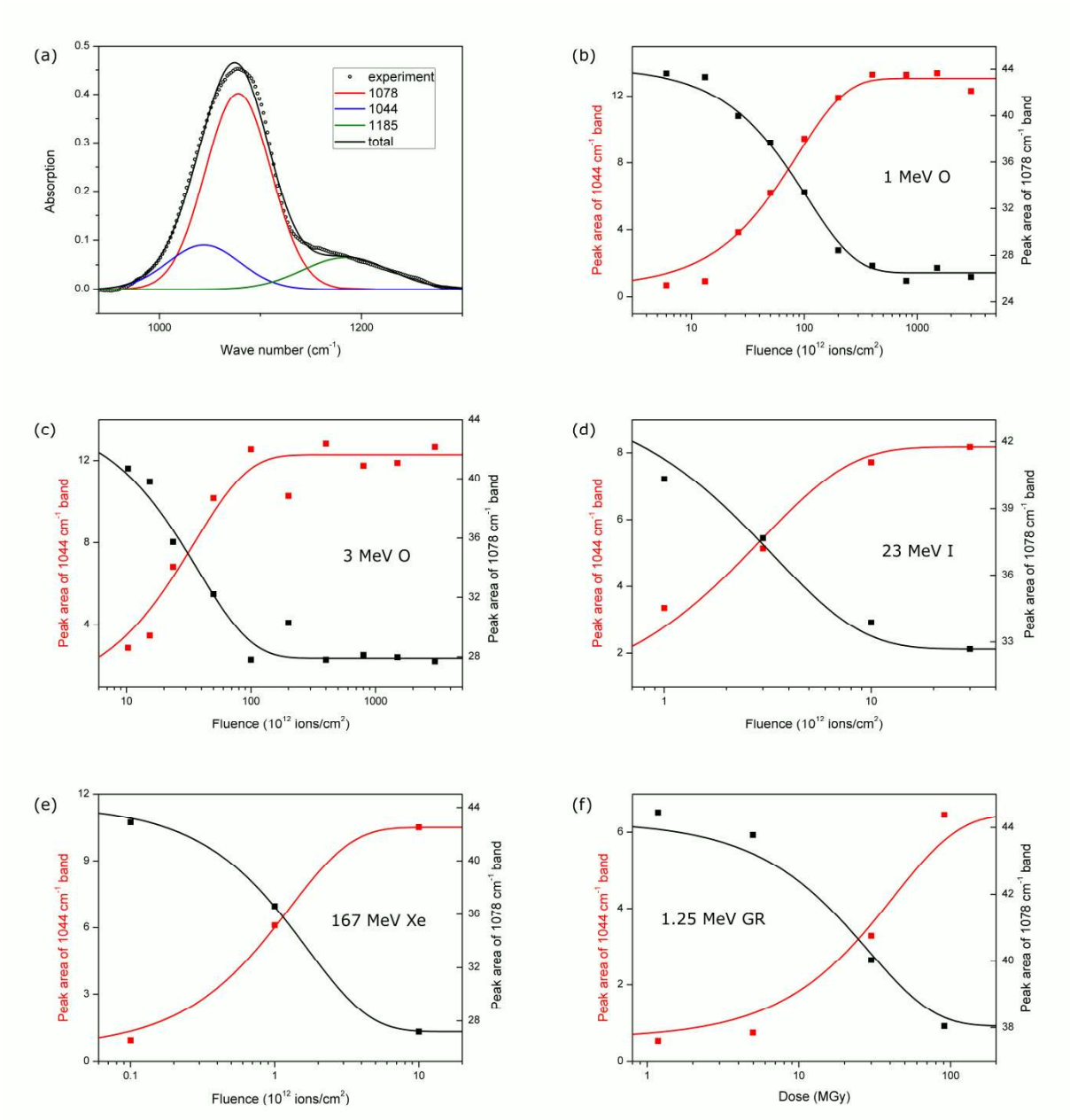
from the sample irradiated by 23 MeV I beam is shown on Fig. 1a. From the analysis of the recorded spectra, it is clearly seen how 1078  $\text{cm}^{-1}$  peak decreases with ion fluence, while 1044  $\text{cm}^{-1}$  peak increases, as shown on Figs. 1b.-1e for different ion beams. There is no significant change in the intensity of the pair LO4-TO4 band at 1165-1200  $\text{cm}^{-1}$ . It can also be seen that 1078  $\text{cm}^{-1}$  peak does not disappear because there is a contribution from the unirradiated a-SiO<sub>2</sub> film on the backside of the Si wafer. In contrast to the previous work [18], we find somewhat different film thickness on the backside because the Si wafer has been polished only on the side facing ion beam.

Since the positions of band maxima are well known [20], Gaussian fitting of the nonirradiated sample with fixed peak positions (wavenumbers 1044, 1078, 1185  $\text{cm}^{-1}$ ) was performed and virgin band areas were obtained. The same fitting procedure was repeated for all irradiated samples. Our fitting procedure is different from the ones in refs. [18, 32] where fitting was done with fixed full widths at half maximum (FWHM) of each band. Most of the oscillators in virgin SiO<sub>2</sub> samples vibrate with frequency of 1078  $\text{cm}^{-1}$  (144° bond angle) and there are, if any, only small number of oscillators with rather narrow energy distribution which vibrate with frequency of 1044  $\text{cm}^{-1}$  (129° bond angle). As seen from experiments, irradiation of the samples influences structural reordering of the molecules, attributed to the change of the O-Si-O bond angle, in which the number of oscillators with smaller bond angle increases. Apart from the change of the number of oscillators, it is to be expected that irradiation causes certain system disorder which would manifest as broader vibrational energy dispersion of the oscillators i.e. width of the 1044  $\text{cm}^{-1}$  band. On the other hand, positions of the band maxima, which correspond to the mean frequencies of the vibrational bands, are not changed with irradiation. Therefore, it is to be expected that Gaussian fitting with fixed positions of the band maxima would yield more accurate description of the physical situation. Indeed, we have found that our spectra showed better fits with fixed positions of each peak. In our fitting procedure, FWHMs of the 1078  $\text{cm}^{-1}$  and 1185  $\text{cm}^{-1}$  band in all samples vary approximately 5% (at most 10%), what can be attributed to the fluence uncertainties, while the FWHM of the 1044  $\text{cm}^{-1}$  band increases with fluence until saturation is reached. All this is in accordance with ref. [79] where it is shown that the width of the TO band is proportional to the mean square deviation of the bond angle in amorphous materials.

Gaussian peak areas obtained using above methodology exhibit uniform evolution with respect to the applied SHI fluence as shown on Fig. 1. To deduce ion track cross-section  $B=r^2\pi$ , the Poisson law is used for fitting evolution of 1078  $\text{cm}^{-1}$  peak area  $Z$  and 1044  $\text{cm}^{-1}$  peak area  $Y$  with increasing SHI fluence  $F$ :

$$Z(F) = [Z_0(F = 0) - Z_{min}(F = \infty)] \times e^{-B \times F} + Z_{min}(F = \infty) \quad (\text{eq 1})$$

$$Y(F) = [Y_{max}(F = \infty) - Y_0(F = 0)] \times (1 - e^{-B \times F}) + Y_0(F = 0) \quad (\text{eq 2})$$



**Figure 1.** (a) IR absorption spectrum from irradiated a-SiO<sub>2</sub> in the range of the TO3 band. Measured IR spectrum (23 MeV I, fluence 3x10<sup>13</sup> ions/cm<sup>2</sup>) is fitted using three Gaussians with peaks centered at 1044 cm<sup>-1</sup>, 1078 cm<sup>-1</sup>, and 1185 cm<sup>-1</sup>. Evolution of the TO3 band with increasing fluence for (b) 1 MeV O, (c) 3 MeV O, (d) 23 MeV I, and (e) 167 MeV Xe beam. (f) Evolution of the TO3 band with increasing dose for 1.25 MeV GR irradiation.

For the measured ion track cross sections, their average values determined from IR spectroscopy of 1078 cm<sup>-1</sup> and 1044 cm<sup>-1</sup> peaks are plotted, and all evaluated cross sections are reported in Table 1. Error bars represent dispersion of the 1078 cm<sup>-1</sup> and 1044 cm<sup>-1</sup> peak data [18]. In the case of non-normal irradiation geometry for the Xe beam, damage cross section was corrected by the geometrical factor. Finally, on Fig. 1f. we present spectra from the GR irradiated samples. Changes in the TO3 band of the IR spectra are visible only for the two highest doses, and show similar behaviour to the SHI irradiated samples. It should be noted that in this case both front side and backside of the a-SiO<sub>2</sub> film are affected by the GR irradiation.

Ion beam	Electronic stopping (keV/nm)	Nuclear stopping (keV/nm)	Range (μm)	Damage cross section $\sigma_{1078}$ (nm <sup>2</sup> )	Damage cross section $\sigma_{1044}$ (nm <sup>2</sup> )	Average damage cross section $\sigma$ (nm <sup>2</sup> )
1 MeV O	0.974	0.019	1.66	0.96 ± 0.05	1.15 ± 0.09	1.06 ± 0.1
3 MeV O	1.679	0.008	3.09	2.68 ± 0.26	2.86 ± 0.38	2.77 ± 0.09
23 MeV I	5.214	0.257	7.19	29.9 ± 3.7	34.1 ± 3.9	32 ± 2.1
167 MeV Xe	13.91	0.058	19.43	51.7 ± 2.1	68.6 ± 5.6	60.15 ± 8.45

**Table 1.** Damage cross sections  $\sigma$  measured by IR spectroscopy for the TO3 band (1078 cm<sup>-1</sup> and 1044 cm<sup>-1</sup> peaks). SHI irradiation parameters calculated using SRIM [80] for a-SiO<sub>2</sub> with density 2.2 g/cm<sup>3</sup>.

#### 4. Discussion

Despite many similarities, two of the most commonly used thermal spike models, namely inelastic thermal spike model (ITSM) and analytical thermal spike model (ATSM), are at present mutually incompatible [81]. It is no surprise that experimental evidence for either validation or refutation of both models is vigorously sought-after via ion-track experiments [5,7,82–85]. Also other SHI irradiation effects were considered with the same purpose, like sputtering [60,86,87], ion hammering [61,65], ion beam mixing [88–90] and nanohillock formation [14,91–95]. Since the present work aims to investigate applicability of ATSM for description of ion tracks in a-SiO<sub>2</sub>, here we summarize model equations [83]:

$$R^2 = a_0^2 \ln \frac{S_e}{S_{et}} \quad S_{et} < S_e < eS_{et} \quad (\text{eq 3})$$

$$R^2 = \frac{a_0^2 S_e}{e S_{et}} \quad eS_{et} < S_e \quad (\text{eq 4})$$

$$S_{et} = \frac{\rho c \Delta T_m a_0^2 \pi}{g} \quad (\text{eq 5})$$

For crystalline insulators, ATSM parameters have been established as  $a_0 = 4.5$  nm and  $g = 0.4$  or  $g = 0.17$ , for low ( $E/A < 2$  MeV/A) and high ( $E/A > 8$  MeV/A) velocity irradiations, respectively. For the intermediate velocities ( $2 \text{ MeV/A} < E/A < 8 \text{ MeV/A}$ ) parameter  $g$  can have intermediate value. These model parameters have been questioned in the case of quartz [5], but later experimental work seems to reaffirm canonical ATSM interpretation [23] as well as previous observation of unusual dispersion of the old experimental data [96]. In the case of a-SiO<sub>2</sub>, non-standard model parameters have been used previously, when increase in factor  $g = 0.6$  has been seen [65], or alternatively decrease of the  $a_0 = 3.5$  nm [19,48] or even to  $a_0 = 2.8$  nm [78] has been proposed. Furthermore, the velocity effect has been observed at lower ion velocities [7,48], and very recently it was reported that parameter  $g$  can be dependent on the electronic stopping power [97]. Given all these developments, and taking into account that a-SiO<sub>2</sub> is more susceptible to ion track formation due to stronger electron-phonon coupling [98] and lower melting temperature [99], it is of importance to reliably establish ATSM parameters needed to successfully describe ion tracks in amorphous SiO<sub>2</sub>. Procedure to establish ATSM parameters is the following [83]: from the logarithmic regime (i.e. data described by (Eq. 3)) parameter  $a_0$  can be extracted by applying the least squares fit on the data in the semilogarithmic graph, when the slope of the fitted line equals  $a_0^2$ . Also, from the same fit, the threshold  $S_{et}$  can be determined. Next, by substituting (Eq. 5) into (Eq. 4), one can use least squares method from data in the linear regime (i.e. data described by (Eq.4)) to determine ATSM parameter  $g$ . Ideally, this value should be consistent with the value of  $g$  that could be numerically calculated from (Eq. 5). In principle, the same procedure could be applied to high velocity data points, but usually very high stopping powers are needed to have sufficient amount of experimental data for thorough analysis.

#### 4.1. Establishing ATSM parameters for a-SiO<sub>2</sub>

Ion tracks in both quartz SiO<sub>2</sub> [23,100,101] and a-SiO<sub>2</sub> have shown some peculiarities that turned out to be important for thermal spike models. In the 1990s, when the amount of experimental data was not yet large, ion tracks in quartz SiO<sub>2</sub> and a-SiO<sub>2</sub> appeared of the same size [26,61,100], and respective thresholds for ion track formation have been found to be very close. Not surprisingly, thermal spike models initially used the same parameters for both quartz and a-SiO<sub>2</sub> [61,88]. In case of ITSM, the difference between these two materials (i.e. greater a-SiO<sub>2</sub> sensitivity to SHI irradiation)

has been noticed earlier and relevant model parameter, electron-phonon mean free path, has been evaluated as  $\lambda = 4$  nm for quartz  $\text{SiO}_2$  and  $\lambda = 2.5$  nm for a- $\text{SiO}_2$  [63]. These values have been fine tuned over the years and recently  $\lambda = 3.6$  nm for quartz  $\text{SiO}_2$  [86] and both  $\lambda = 2.5$  nm and  $\lambda = 3$  nm for a- $\text{SiO}_2$  have been reported [18,60]. Later it was recognized that standard ATSM parameters for crystalline insulators [83] are also not suitable for description of ion tracks in a- $\text{SiO}_2$  [19,48,65,78].

To establish ATSM parameters for a- $\text{SiO}_2$ , we now analyse both here presented new experimental data on ion tracks in a- $\text{SiO}_2$ , as well as previously published data using the same methodology (IR spectroscopy) [17,18,26–28,32]. For comparison, we also analyse recent results from optical reflectivity (OR) measurements [102]. First, we restrict our analysis only to data where contribution of the nuclear stopping can be neglected i.e. where ratio between the two is at least one order of magnitude. In the analysis, also included are experimental data from the transmission electron microscopy (TEM) and small angle X-ray scattering (SAXS) because total size of the core-shell track observed by SAXS is in agreement with the results of the IR measurements [18]. We list published and here measured experimental data in Table 2.

Ion	Energy (MeV)	Energy per nucleon (MeV/A)	Electronic stopping (keV/nm)	Nuclear stopping (keV/nm)	Ion track cross section (nm <sup>2</sup> )	Technique and Reference
Xe	762	5.77	15.7	0.02	80	IR, [26]
Ni	551	9.5	4.99	0.003	6	IR, [26]
Au	22.5	0.11	3.84	0.7	$18.1 \pm 3.0$	SAXS, [17]
Au	45.7	0.23	7.32	0.43	$55.4 \pm 2.6$	SAXS, [17]
Au	77.2	0.39	10.97	0.29	$75.4 \pm 3.1$	SAXS, [17]
Au	168.3	0.85	16.17	0.16	$91.6 \pm 3.4$	SAXS, [17]
Xe	628	4.76	16.02	0.02	$58.1 \pm 5.4$	SAXS, [17]
Xe	1416	10.73	13.67	0.01	$50.3 \pm 7.5$	SAXS, [17]
O	3.1	0.19	1.69	0.007	$3.8 \pm 0.7$	IR, [18]
Si	5.5	0.2	2.77	0.02	$14 \pm 4$	IR, [18]
Ni	11.4	0.2	3.89	0.08	$27 \pm 6$	IR, [18]
O	3.1	0.19	1.69	0.007	$3.8 \pm 1.4$	TEM, [18]
Si	5.5	0.2	2.77	0.02	$13.9 \pm 4$	TEM, [18]
Ni	11.4	0.2	3.89	0.08	$26.4 \pm 9.1$	TEM, [18]
C	66	5.5	0.47	0.0003	0	IR, [32]

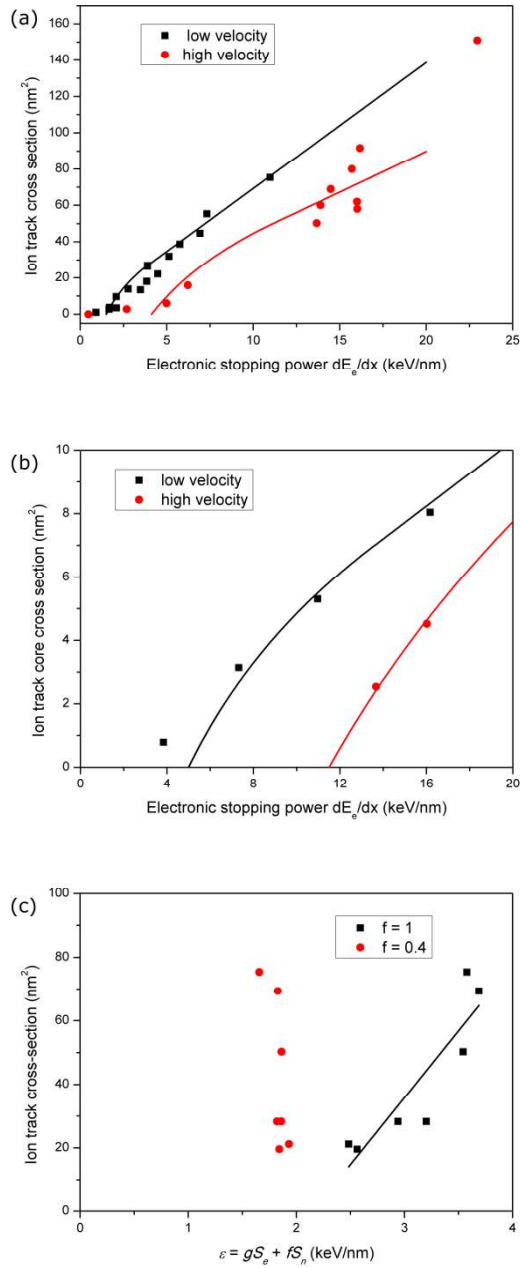
S	160	5	2.69	0.002	$2.8 \pm 0.5$	IR, [32]
Ni	320	5	6.22	0.005	$16 \pm 2$	IR, [32]
Sn	616	5.5	14.48	0.015	$69 \pm 9$	IR, [32]
Xe	645	5	16	0.018	$62 \pm 9$	IR, [32]
Pb	957	4.6	22.95	0.042	$151 \pm 25$	IR, [32]
F	5	0.26	2.09	0.004	3.5	OR, [102]
Br	5	0.06	2.08	0.265	9.7	OR, [102]
Br	10	0.13	3.48	0.16	13.5	OR, [102]
Br	15	0.19	4.48	0.119	22.2	OR, [102]
Br	25	0.32	5.76	0.08	38.7	OR, [102]
Br	40	0.51	6.92	0.055	44.7	OR, [102]
O	0.9	0.06	0.91	0.02	$1.06 \pm 0.1$	IR, here
O	2.92	0.18	1.67	0.008	$2.77 \pm 0.09$	IR, here
I	22.5	0.18	5.13	0.261	$32 \pm 2.1$	IR, here
Xe	166	1.26	13.89	0.058	$60.15 \pm 8.45$	IR, here

**Table 2.** Measured ion track radii and related cross sections in a-SiO<sub>2</sub> using IR, SAXS, TEM and OR, either previously published with corresponding reference, or measured in this work. The SHI kinetic energy is calculated at the mid-point of thin a-SiO<sub>2</sub> film. All stopping powers are calculated using SRIM 2013 code [80].

As shown on Fig. 2a, velocity effect is active already at 1 MeV/A, in agreement with our previous work [48]. Thus, for the analysis of the low velocity data points we consider only data acquired from irradiations with specific kinetic energies below  $E/A = 0.5$  MeV/A. Experimental data from irradiations with specific kinetic energy above 0.85 MeV/A are treated as high-velocity data points.

First, we consider low-velocity data points. The threshold for the ion track formation  $S_{et} = 1.50 \pm 0.25$  keV/nm is found analysing the data from the logarithmic regime (Eq. 3) occurring below 4 keV/nm in electronic stopping power. From the same data subset, ATSM parameter  $a_0 = 2.7 \pm 0.2$  nm is found, in agreement with previous work [78]. Analysing the data from the linear regime, ATSM parameter  $g = 0.53 \pm 0.02$  was established by fitting the experimental data to (Eq. 4). By taking into account established ion track formation threshold of  $S_{et} = 1.50 \pm 0.25$  keV/nm, the analysis yields  $a_0 = 3.4 \pm 0.3$  nm for data points from linear regime. This way we conclude the ATSM parameters for low velocity irradiations are  $a_0 = 3.0 \pm 0.5$  nm and  $g = 0.53 \pm 0.02$ . Using these values (Eq. 4),

calculated threshold for ion track formation  $S_{et} = 1.53 \pm 0.28$  keV/nm, in excellent agreement with experimentally determined value.



**Figure 2.** (a) Ion tracks in a-SiO<sub>2</sub> after low velocity irradiations (black) and high velocity irradiations (red). The ATSM model predictions are plotted for comparison. (b) Ion tracks cores in a-SiO<sub>2</sub> after low velocity irradiations (black) and high velocity irradiations (red). The ATSM model predictions are plotted for comparison. (c) Damage cross section dependence on the density of deposited energy for two different scenarios: very efficient energy transfer of nuclear stopping ( $f=1$ ) and standard transfer rate of electronic stopping ( $f=0.4$ ). The ATSM model prediction is plotted for comparison.

As often is the case, very small ion tracks can be found below the threshold established by the least squares method, which are usually neglected in the analysis [83]. In our case, this is 1 MeV O data point, which we do not include into the present analysis. However, as can be seen from comparing Figs. 1b and 1c, differences between 1 MeV O and 3 MeV O are not that large and could point to other track forming mechanism below the threshold established by the thermal spike model, like for example excitonic mechanism [3]. Present work can open up the way for future studies of near threshold ion track damage formation, that should shed light on this open question [23]. Generalization of the analysis to Avrami-type behaviour [103,104] could be well justified for these irradiation conditions.

Next, we consider the high-velocity data subset. There are only two data points above 8 MeV/A, but as can be seen on Fig.2a, 0.85 MeV/A gold and 1.26 MeV/A xenon irradiations can be included into this data set, as well as a number of irradiations with ion velocities of 5 MeV/A. Below-threshold datapoint of 5 MeV/A carbon ion beam is neglected. Still, two data points can be found in the logarithmic regime (Eq. 4), indicating larger value of the ATSM parameter  $a_0 = 4$  nm for high velocity irradiations. Using the same procedure as before, we analysed the linear regime between 10-20 keV/nm to obtain ATSM parameter  $g = 0.35 \pm 0.02$  for the high velocity irradiations. Data point from 5 MeV/A lead ion irradiation was not used in the analysis because as an outlier it was not possible to incorporate it in the present analysis. Taking into account changes to ATSM parameters, the high velocity threshold for ion track formation is estimated at  $S_{et} = 4.1$  keV/nm.

As shown in Fig. 2a, plotted curves using ATSM parameters ( $a_0 = 3$  nm,  $g = 0.53$ ) for low velocity and ( $a_0 = 4$  nm,  $g = 0.35$ ) for high velocity irradiations follow experimental data well. Velocity effect is clearly observed, but separation between high and low velocity data occurs already at 1 MeV/A, similar to the case of  $\text{CaF}_2$  [7] and Ge containing  $\alpha\text{-SiO}_2$  [48]. Although it is difficult to maintain assignment of the standard ATSM values  $g = 0.17$  to pure thermal spike and  $g = 0.4$  to Coulomb explosion contribution, we note high velocity ATSM parameters have values close to the standard ATSM parameters for the low velocity irradiation. This would implicate that Coulomb explosion is active even for irradiations using the highest ion velocities at 10 MeV/A, but more experimental data is needed to verify this.

However, the most puzzling values of the ATSM parameters are for the low velocity irradiations. These non-standard values are needed to correctly describe low threshold for ion track formation, as well as ion track sizes in this regime. As proposed before, possible explanation can be related to the amorphous structure of the  $\alpha\text{-SiO}_2$  material. The velocity effect, as it is conceived in

the ITSM, i.e. high density of deposited energy after low velocity irradiations as a consequence of limited spread of excited electrons in the primary stages of the ion track formation, could possibly be promoted even further by amorphous structure of the material and its low thermal conductivity. Thus, very narrow cylinder ( $a_0 < 4.5$  nm) of highly excited ( $g > 0.4$ ) matter along ion trajectory, as suggested by ATSM parameters, could be responsible for observed ion track features. Similar conclusion has very recently been reached by molecular dynamics studies of ion track formation in a-SiO<sub>2</sub> [102].

#### 4.2. Extending ATSM to description of ion track core and additive role of nuclear stopping in a-SiO<sub>2</sub>

Two features of ion tracks in a-SiO<sub>2</sub> have been used previously to support validity of the ITSM description of ion tracks in this material: appearance of the underdense ion track core for very high electronic stopping powers [17], and additivity of the electronic and nuclear stopping power as source term in the ITSM equations [18]. The observation of the underdense core in SAXS measurements have been assigned to very high densities of deposited energy leading to increase of the material temperature above the boiling point of 7000 K [17]. Results of the molecular dynamics simulations agree well with experimental data [17,102]. In the Table 3 we list available experimental data on measured radii of the ion track cores in a-SiO<sub>2</sub> [17].

Ion	Energy (MeV)	Energy per nucleon (MeV/A)	Electronic stopping (keV/nm)	Nuclear stopping (keV/nm)	Ion track core cross section (nm <sup>2</sup> )	Technique and Reference
Au	22.5	0.11	3.84	0.7	$0.79 \pm 0.47$	SAXS, [17]
Au	45.7	0.23	7.32	0.43	$3.14 \pm 0.94$	SAXS, [17]
Au	77.2	0.39	10.97	0.29	$5.31 \pm 0.82$	SAXS, [17]
Au	168.3	0.85	16.17	0.16	$8.04 \pm 1.01$	SAXS, [17]
Xe	628	4.76	16.02	0.02	$4.52 \pm 1.51$	SAXS, [17]
Xe	1416	10.73	13.67	0.01	$2.54 \pm 1.13$	SAXS, [17]

**Table 3.** Measured ion track core radii and calculated core cross sections in a-SiO<sub>2</sub> using SAXS [17]

Due to scarcity of data, we do not perform the analysis in the manner described in the section 4.1. Instead, as shown on Fig. 2b, we plot available experimental data together with estimated ATSM parameters that ensure satisfactory fit. We estimate initial widths of the thermal spike as  $a_0 = 1.5$  nm for low velocity irradiations, and  $a_0 = 2.1$  nm for high velocity irradiations. These

values are much smaller than ATSM parameter  $a_0$  deduced from ion track radii measurements in section 4.1., but decrease of  $a_0$  with increase in the deposited energy density is consistent with results from section 4.1., indicating highly localized ultrafast processes on the nanoscale. Furthermore, by estimating thresholds for appearance of the underdense ion track core at 5 keV/nm for low velocity and at 11.5 keV/nm for high velocity irradiations, by means of (eq. 5), other ATSM parameter  $g$  can be evaluated. By imposing material boiling at 7000 K as requirement for appearance of the underdense ion track core,  $g = 0.16$  for low velocity irradiations, and  $g = 0.135$  for high velocity irradiations can be found. These values are very close to standard ATSM parameter  $g = 0.17$  for high velocity irradiations, assigned to pure thermal spike process. Alternatively, by imposing material melting as requirement for appearance of the underdense ion track core,  $g = 0.64$  for low velocity irradiations, and  $g = 0.54$  for high velocity irradiations can be found, indicating Coulomb explosion mechanism, and in very good agreement with values obtained in section 4.1. At present it is not possible to rule out any of these two possibilities, because from (Eq. 5) only ratio between temperature increase and ATSM parameter  $g$  can be found. Boiling of the material as ITSM requirement for small ion track sizes in  $\text{CaF}_2$  has been criticized previously [7,105], and this criticism can be extended to track cores in  $\text{a-SiO}_2$  as well. ATSM opens up another route for ion track core formation, related to melting of the material, thus avoiding the controversy, but clearly more experimental data is needed to verify this hypothesis.

Next, we look into remaining experimental data published in ref. [18] where ion tracks were produced using Au ion beams having energies between 0.5 MeV and 15 MeV. In this energy range, electronic and nuclear stopping have comparable values. Good agreement between ITSM and experimental data has been achieved when nuclear stopping power has also been included as a source term of excitation in differential equation describing evolution of the temperature of the atomic subsystem. The agreement has been improved when stopping power values calculated by the SRIM have been determined applying reciprocity principle of Sigmund [106]. The same data is used here in order to extract ATSM parameters describing nuclear stopping contribution to the ion track formation, using the same approach that has been used previously by Szenes to describe ion tracks on muscovite mica surface [91]. In that work, increase of the ATSM parameter  $g$  up to 0.9 has been found, indicating very efficient energy transfer in case of nuclear stopping. Similar efficient transfer of deposited energy (via nuclear stopping) has been found in the case of amorphous  $\text{Al}_2\text{O}_3$  [78].

Ion	Energy (MeV)	Energy per nucleon (MeV/A)	Electronic stopping (keV/nm)	Nuclear stopping (keV/nm)	Ion track cross section (nm <sup>2</sup> )	Technique and Reference
Au	0.3	0.0015	0.71	3.2	75.4 ± 24.6	IR [18]
Au	0.6	0.003	1.11	3.1	69.4 ± 20.7	IR [18]
Au	1.3	0.0066	1.4	2.8	50.3 ± 15.1	IR [18]
Au	2.3	0.012	1.7	2.3	28.3 ± 9.4	IR [18]
Au	4.8	0.024	2.15	1.8	28.3 ± 9.4	IR [18]
Au	9.8	0.05	2.57	1.2	19.6 ± 6.3	IR [18]
Au	14.8	0.075	2.95	0.92	21.2 ± 6.5	IR [18]

**Table 4.** Measured ion track radii and calculated ion track cross sections in a-SiO<sub>2</sub> using IR spectroscopy measurements [18]. Electronic stopping power is tabulated from ref. [18] where reciprocity principle has been used.

As shown in Fig. 2c, considering electronic and nuclear stopping on equal footing is not realistic within ATSM framework. Only for very efficient transfer of the deposited energy from nuclear stopping into the thermal spike, reasonable behaviour can be achieved. The threshold for damage formation at 2 keV/nm is in reasonable agreement with the threshold for ion track formation found at 1.5 keV/nm. Still, ATSM parameters needed to describe observed damage behaviour originating from nuclear stopping are very much different than parameters found in section 4.1. Besides  $f = 1$  that is in agreement with previous works on nuclear stopping contribution to the thermal spike [78,91], another ATSM parameter should be close to the standard value for insulators  $a_0 = 4.5$  nm. At present, these ATSM parameters are given only as estimates because most of the data is clustered between 2.5 - 3.5 keV/nm. To establish their values reliably, more experimental data is needed in wider range of stopping powers.

#### 4.3. Gamma rays and swift heavy ions: a comparison

The original idea underlying this study was to investigate if GR irradiation effects in a-SiO<sub>2</sub> could be imitated by SHI irradiation. This would open up a way to simulate radiation response of the material to high dose GR irradiation using SHI irradiation, which should be doable on much shorter time-scale. For example, in this study we have exposed a-SiO<sub>2</sub> samples to a very high dose GR irradiation in multi-month exposures, while even highest fluences of SHI irradiation were accomplished in a

matter of hours. Clearly, ion track formation is a unique feature of SHI irradiation, so we turned to SHI irradiation under conditions when electronic stopping power was under threshold for ion track formation. This way we ensured that atomic rearrangements would occur after similar electronic excitation, but no large-scale defects (i.e. ion track formation) would take place. To accomplish this, we have applied 1 MeV O (below threshold) and 3 MeV O (at the ion track formation threshold) irradiations and compared the results with the IR spectra from GR irradiated samples.

As shown on Fig. 1f, significant changes to the TO3 band in the IR spectra of GR irradiated samples occur only for highest doses of 30 MGy and 90 MGy. The GR irradiations with doses up to 10 MGy do not produce significant changes. Comparing with the 1 MeV O irradiated samples, we observe similar IR spectra changes in the range of ion fluences between  $3 \times 10^{13}$  –  $3 \times 10^{14}$  ions/cm<sup>2</sup>. We note that required GR doses and SHI fluences agree very well with the criterion used in the work by Awazu et al. [28], where density of deposited energy equal to the total Si-O bonding energy of 3.8 eV in a unit volume of a-SiO<sub>2</sub> has been correlated with the structural changes. This critical density of deposited energy corresponds to  $3.4 \times 10^{23}$  eV/cm<sup>3</sup>. In our case, for 90 MGy GR irradiation, density of deposited energy equals  $1.25 \times 10^{24}$  eV/cm<sup>3</sup>, and for 1 MeV O irradiation with the fluence of  $1 \times 10^{14}$  ions/cm<sup>2</sup> we find deposited energy density equal to  $9 \times 10^{23}$  eV/cm<sup>3</sup> (we note damage cross section equals 1 nm<sup>2</sup>, and applied ion fluence equals 1/nm<sup>2</sup>). Therefore, in both cases similar changes to IR spectra occur at the same levels of deposited energy densities, and also similar values can be found for other SHI beams used. Therefore, SHI irradiations present very attractive option for simulating very high dose GR exposures to materials relevant for nuclear applications.

## 5. Conclusions

In this work, ion tracks in amorphous SiO<sub>2</sub> were measured using infrared spectroscopy. Obtained experimental results of ion track measurements show typical behaviour like thresholding and increase in size with stopping power. However, model parameters have to be tuned to unusual values to obtain satisfactory description, making this material suitable for comparison with other thermal spike models. Other ion track features like core-shell morphology and contribution of the nuclear stopping to final ion track size can be used for the same purpose. Here we show that analytical thermal spike model is able to reproduce ion track core features without invoking boiling criteria like in the case of inelastic thermal spike model. In addition, we show transfer of the nuclear stopping into the thermal spike is very efficient. Finally, it was shown how present approach could

be useful to simulate response of the  $\alpha$ -SiO<sub>2</sub> to very high dose gamma rays exposures using ion irradiations performed on much shorter timescales.

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## Data availability statement

The raw data required to reproduce these findings are available on request. The processed data required to reproduce these findings are available in Tables 1-4.

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