Swift heavy ion tracks in alkali tantalate crystals: a combined experimental and computational study

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Abstract
The formation of latent tracks with different damage morphologies in alkali tantalate crystals (KTaO\textsubscript{3} and LiTaO\textsubscript{3}) under the action of the extreme electronic energy loss induced by 358 MeV $^{58}$Ni\textsuperscript{19}\textsuperscript{+} irradiation was studied by experimental characterizations of the lattice damage and numerical calculations using the inelastic thermal spike model. Prism coupling measurements were used to analyze the refractive index profiles of irradiated regions. This approach is effective and very accurate for determination of the in-depth damage profile and its correlation with the energy loss curves. The calculated spatio-temporal evolution of the energy deposition densities and lattice temperatures theoretically demonstrate the experimentally observed latent tracks in Ni\textsuperscript{19+}-irradiated crystals. Based on the observed damage morphologies of individual and overlapped spherical defects, and discontinuous and continuous tracks, the corresponding threshold values of the electronic energy loss for track damage in alkali tantalate crystals were assessed. For irradiating ions with an energy of 6.17 MeV amu\textsuperscript{-1}, a threshold of $\sim$12.0 keV nm\textsuperscript{-1} for the production of spherical defects in KTaO\textsubscript{3} crystals is indicated, and the threshold for LiTaO\textsubscript{3} crystals is less than 12.0 keV nm\textsuperscript{-1}. For irradiating ions with an energy of 2.15 MeV amu\textsuperscript{-1}, owing to the ion-velocity dependence effect, an electronic energy loss of $\sim$13.8 keV nm\textsuperscript{-1} leads to overlapped spherical defects and discontinuous tracks in KTaO\textsubscript{3} and continuous tracks in LiTaO\textsubscript{3}. Compared with LiTaO\textsubscript{3}, a relatively higher damage tolerance and critical threshold for track formation in KTaO\textsubscript{3} crystals are proven. The determined lattice temperature threshold for continuous track production is 3410 K for KTaO\textsubscript{3} and slightly less than 3250 K for LiTaO\textsubscript{3}, demonstrating that, compared with the melting point, a much higher lattice temperature in the region surrounding the ion...
path needs to be achieved to produce stable track damage due to the non-negligible effect of melting damage caused by annealing during the cooling process.

Keywords: ion irradiation, electronic energy loss, thermal spike response, latent ion track

(Some figures may appear in colour only in the online journal)

1. Introduction

In the field of radiation research, study of the interactions between energetic ions and solid materials is fundamental to understanding the responses of materials to ion irradiation and the induced effects on microstructure and performance [1–4]. Irradiating a solid material with swift heavy ions leads to intense electronic excitation and thermal spike responses due to inelastic collisions between moving ions and target electrons [5–7]. One of the major consequences of this interaction is the molten phase induced along the path of ion penetration and the subsequent latent track with a width of a few nanometers and length of several tens of micrometers [8–11]. The properties in such nanoscale regions containing lattice disorder or an amorphous phase are drastically changed and modified compared with the surrounding virgin bulk. Thus, swift heavy ion irradiation is a powerful technique that is utilized to study the undesirable phenomena (creep, swelling, embrittlement, etc) occurring in irradiation environments [12–16] but also has beneficial applications in material modification and device fabrication on the nano- and micrometer scales [17–31]. Alkali tantalate crystals (KTaO$_3$, LiTaO$_3$, etc) with perovskite-like structures exhibit versatile properties that are attractive for numerous applications, such as electro-optic waveguides in microelectronics [32], pyroelectrics in neutron generation [33] and photocatalysts in pollutant degradation [34] and water splitting [35]. According to recent work, the abovementioned properties can be effectively modified by the introduction of ion-irradiation-induced defects and nano-structures [20, 36]. Utilizing swift Ni$^{19+}$ ion irradiation, the present work focuses on the thermal spike responses and latent track behaviors of alkali tantalate crystals under the action of different ion energies and electronic energy losses. Different damage morphologies, from individual and overlapped spherical defects to discontinuous and continuous tracks, are discussed, and the corresponding threshold values are determined. The temperature threshold is used to define the condition for generation of latent track damage from a thermodynamic perspective, and could be used to discuss the track behaviors corresponding to different electron energy losses and ion velocities under different irradiating ion species and ion energies. In this work, the introduced temperature threshold is demonstrated to be a more fundamental and essential parameter for determining the latent track behaviors; it is broadly applicable and makes the experimental results for different irradiation conditions comparable. Addressing these issues can not only provide a deeper scientific understanding of the puzzling scenario of ion track formation in insulators but also contribute to a foundation and assessment for ion-irradiation applications in the fields of material modification and nanostructure fabrication.

2. Materials and methods

2.1. Alkali tantalate crystals and ion irradiation processes

The single-crystal (100) KTaO$_3$ (perovskite structure with cubic $Pm\overline{3}m$ symmetry) and (006) LiTaO$_3$ (ilmenite structure with rhombohedral $R3c$ symmetry) samples with dimensions of 10 mm $\times$ 10 mm $\times$ 0.5 mm were irradiated at 300 K with 358 MeV $^{58}$Ni$^{19+}$ at fluences of $1 \times 10^{12}$, $3 \times 10^{12}$ and $3 \times 10^{13}$ cm$^{-2}$, respectively. The swift Ni$^{19+}$ ion irradiations were carried out at the Heavy Ion Research Facility in Lanzhou (HIRFL), Institute of Modern Physics, Chinese Academy of Sciences, which has relatively precise energy and fluence calibrations, and could guarantee the repeatability of the irradiation experiment. During the irradiation process, a relatively low Ni$^{19+}$-beam current density and ion flux (8.1 $\times$ 10$^{10}$ cm$^{-2}$s$^{-1}$) were used in order to avoid undesired ion beam annealing and charge accumulation on the samples [37].

2.2. Experimental characterizations of latent track damage

The track damage in alkali tantalate crystals induced by swift Ni$^{19+}$ irradiation was characterized through prism coupling, Rutherford backscattering spectroscopy in channeling configuration (RBS/channeling), high-resolution x-ray diffraction (HRXRD) and transmission electron microscopy (TEM) techniques. Prism coupling measurements were carried out utilizing a Metricon model 2010 prism coupler in a near-infrared wavelength band (1539 nm diode laser). In the RBS/channeling analysis, a 2.0 MeV He$^+$ beam with a probe size of 2 mm $\times$ 2 mm was extracted from an NEC 2 $\times$ 1.7 MV tandem accelerator, and a Si detector located at a scattering angle of 160° relative to the He$^+$ beam was used to collect the backscattered He$^+$ signal. The HRXRD measurements were performed using a Rigaku Smartlab high-resolution x-ray diffractometer, in which a Cu K$_{\alpha1}$ anticathode with a Ge monochromator was used to provide a parallel and monochromatic x-ray beam with a wavelength of 1.54 Å. In the measurements, $\omega$-2$\theta$ scans were recorded with a step size of 0.001°. Cross-sectional TEM samples of alkali tantalate crystals were prepared by a standard method including polishing, dimpling and then Ar ion milling with a Gatan 695 precision polishing system, and were observed with an FEI Tecnai G2 F20 transmission electron microscope. The damage characterization experiments were also carried out several times in order to confirm the accuracy and repeatability of the analysis results.
2.3. Simulations and analysis of the track damage effect

The electronic energy loss and displacements per atom (dpa) induced by irradiating alkali tantalates with 358 MeV Ni\(^{19+}\) were determined by using the Stopping and Range of Ions in Matter (SRIM) 2013 full-cascade simulation code [38, 39]. In these two crystals, the displacement energies of Li, K, Ta and O atoms were set as default values in the SRIM simulation code [25, 25, 25 and 28 eV, respectively]. Based on the results of prism coupling measurements [40], the refractive index profiles in Ni\(^{19+}\)-irradiated regions were reconstructed by utilizing the inverse Wentzel–Kramers–Brillouin (iWKB) procedure [41, 42]. The spatio-temporal evolutions for the energy deposition and lattice temperature induced by electronic energy loss were numerically calculated with the inelastic thermal spike (iTS) model [43, 44]. The experimentally observed lattice damage and latent track behavior in alkali tantalate crystals were analyzed based on the iTS calculation results.

3. Results and discussion

3.1. Experimental characterizations of latent tracks in alkali tantalate crystals

As shown in figure 1(a), the metallographic cross-section images of Ni\(^{19+}\)-irradiated KTaO\(_3\) and LiTaO\(_3\) samples clearly indicate the regions with irradiation-induced damage. Under \(^{58}\text{Ni}^{19+}\) irradiation with a fluence of \(3 \times 10^{12}\) cm\(^{-2}\), the LiTaO\(_3\) crystal sample was obviously darker than the KTaO\(_3\) crystal sample at the position of peak electronic energy loss, providing evidence that the transmissivity decreased considerably. Therefore, more severe irradiation damage was observed in the LiTaO\(_3\) crystal than in the KTaO\(_3\) crystal. Electronic energy loss and simulated dpa depth profiles corresponding to 358 MeV \(^{58}\text{Ni}^{19+}\) irradiation with a fluence of \(3 \times 10^{13}\) cm\(^{-2}\) are shown in figure 1(b), as indicated by the black solid curves and red dashed curves, respectively. The peak values of dpa corresponding to \(^{58}\text{Ni}^{19+}\) irradiation at a fluence of \(3 \times 10^{13}\) cm\(^{-2}\) are 0.037 and 0.028 for the KTaO\(_3\) crystal and LiTaO\(_3\) crystal, respectively. Based on the damage accumulation curve [36, 45], the disorder levels of the Ta sublattice in KTaO\(_3\) crystal and LiTaO\(_3\) crystal corresponding to the related dpa values are 1.9% and 3.2%, respectively. Owing to the low absolute dpa values in the present work, the nuclear energy loss process \((\text{Sn})\) is negligible. Therefore, more severe irradiation damage was observed in the LiTaO\(_3\) crystal than in the KTaO\(_3\) crystal. Electronic energy loss and simulated dpa depth profiles corresponding to 358 MeV \(^{58}\text{Ni}^{19+}\) irradiation with a fluence of \(3 \times 10^{13}\) cm\(^{-2}\) are shown in figure 1(b), as indicated by the black solid curves and red dashed curves, respectively. The peak values of dpa corresponding to \(^{58}\text{Ni}^{19+}\) irradiation at a fluence of \(3 \times 10^{13}\) cm\(^{-2}\) are 0.037 and 0.028 for the KTaO\(_3\) crystal and LiTaO\(_3\) crystal, respectively. Based on the damage accumulation curve [36, 45], the disorder levels of the Ta sublattice in KTaO\(_3\) crystal and LiTaO\(_3\) crystal corresponding to the related dpa values are 1.9% and 3.2%, respectively. Owing to the low \(^{58}\text{Ni}^{19+}\) fluences \((1 \times 10^{12}, 3 \times 10^{12} \text{ and } 3 \times 10^{13} \text{ cm}^{-2})\) and absolute dpa values in the present work, the nuclear energy loss process \((\text{Sn})\) is negligible. Thus, the irradiation damage shown in figure 1(a) is ascribed to the intense electronic energy loss process \((\text{Sn})\) and target electrons. Under the action of 358 MeV \(^{58}\text{Ni}^{19+}\) irradiation, the electronic energy loss in the KTaO\(_3\) and LiTaO\(_3\) surface regions is \(\sim 12.0\) keV nm\(^{-1}\), and the maximum electronic energy loss located at the Bragg peak \((-18.0\) µm depth\) is \(\sim 13.8\) keV nm\(^{-1}\), as shown in figure 1(b).

In this work, the prism coupling technique was first used to rapidly evaluate the in-depth damage evolution profile in the surface and its correlation with the peak electronic energy loss regions of Ni\(^{19+}\)-irradiated KTaO\(_3\) and LiTaO\(_3\) samples. Unlike other characterization experiments (RBS/channeling, TEM, etc) limited by large facilities or complex sample preparation processes, prism coupling measurements can be directly carried out on ion-irradiated samples by utilizing a miniature prism coupler. As shown in the schematic diagram in figure 1(c), utilizing the prism coupling technique the light intensity corresponding to TE or TM modes (depending on the axial orientation of the electric field vector) was measured using lasers. When the electronic energy loss located at the Bragg peak produces obvious lattice damage, the refractive index of the buried damaged layer (optical barrier) decreases owing to the damage-induced lattice swelling/disordered ion tracks embedded in the crystalline host matrix; then, the laser beam propagating in the prism can couple into the surface region via an evanescent field effect, leading to a decrease in the output laser power. The presence of sharp drops in the measured light intensity spectrum indicates the production of a buried damaged layer located at the Bragg peak. In addition, the knee point in the light intensity curve corresponds to the refractive index of the region just below the sample surface; thus, a distinguishable change in the surface refractive index measured in the ion-irradiated sample compared with the virgin sample indicates the production of surface damage. The curve shown in the schematic diagram (figure 1(d)) indicates the light intensity obtained by the prism coupling measurement, which is obtained from [46] and used to illustrate the physical mechanism of prism coupling experiments. Therefore, prism coupling measurements can quickly provide basic knowledge about the production of irradiation-induced damage and the evolution of its depth profile in transparent materials.

The prism coupling measurements were carried out utilizing a Metronic model 2010 prism coupler at a visible band (633 nm He–Ne laser) and a near-infrared wavelength band (1539 nm diode laser). The measured light intensity spectra corresponding to \(^{58}\text{Ni}^{19+}\)-irradiated KTaO\(_3\) and LiTaO\(_3\) samples are shown in figures 1(e) and (f), respectively. Based on the sharp drops in the light intensity curves, the iWKB-reconstructed refractive index profiles are shown in figures 1(g) and (h), respectively. First, focusing on the sample surface regions \((E_{\text{ion}} = 6.17\) MeV amu\(^{-1}\), \(S_r = \sim 12.0\) keV nm\(^{-1}\)), the refractive indices in the Ni\(^{19+}\)-irradiated KTaO\(_3\) samples still coincide with those in the virgin sample, indicating no significant production of surface damage in this case. However, the refractive indices in the LiTaO\(_3\) samples decrease with increasing Ni\(^{19+}\) fluence, indicating the presence of irradiation-induced surface damage. Second, concerning the peak electronic energy loss regions \((E_{\text{ion}} = 2.15\) MeV amu\(^{-1}\), \(S_r = \sim 13.8\) keV nm\(^{-1}\) at a depth of \(\sim 18.0\) µm), the refractive indices in both the KTaO\(_3\) and LiTaO\(_3\) samples decrease with increasing Ni\(^{19+}\) fluence, indicating the production of substantial irradiation damage in these regions. Third, comparing figure 1(e) with figure 1(f), for Ni\(^{19+}\)-irradiation at a fluence of \(1 \times 10^{12}\) cm\(^{-2}\) (orange lines), the guided mode (sharp drop) still cannot be supported in the KTaO\(_3\) sample owing to the relatively low level of damage (relatively high refractive index) in the buried layer, differing
from the LiTaO$_3$ sample. Upon increasing the Ni$^{19+}$ fluence to $3 \times 10^{13}$ cm$^{-2}$ (green lines), the guided mode (sharp drop) can no longer be supported in the LiTaO$_3$ sample owing to the relatively high level of damage (relatively low refractive index) in the surface region, differing from the KTaO$_3$ sample. Thus, compared with the LiTaO$_3$ crystal, the KTaO$_3$ crystal presents better damage tolerance (less sensitive) behavior under the action of the electronic energy loss induced by swift heavy ion irradiation, in good agreement with the metallographic cross-section images shown in figure 1(a).

The different damage responses in the surface regions of the Ni$^{19+}$-irradiated KTaO$_3$ and LiTaO$_3$ samples are confirmed by the RBS/channeling spectra shown in figures 2(a) and (b). With increasing Ni$^{19+}$ fluence, the channeling curves measured in the KTaO$_3$ samples remain consistent with the virgin curve, while the channeling curves measured in the LiTaO$_3$ samples visibly increase compared with the virgin curve, indicating no significant damage to the KTaO$_3$ surface regions but obvious damage to the LiTaO$_3$ surface regions. The measured HRXRD patterns corresponding to the KTaO$_3$ and LiTaO$_3$
samples under different irradiation conditions are shown in figures 2(c) and (d), respectively. For the virgin (1 0 0) KTaO₃ crystal with a cubic perovskite structure, the characteristic peak located at 2θ = 70.75° corresponds to a reflection from the (3 0 0) plane (JCPDS card no. 38-1470), and for the virgin (006) LiTaO₃ crystal with the rhombohedral ilmenite structure the characteristic peak located at 2θ = 84.33° corresponds to a reflection from the (0 0 1 2) plane (JCPDS card no. 71-0953). Upon increasing the Ni¹⁹⁺ fluence to 3 × 10¹³ cm⁻², the broadening of the main peak in the KTaO₃ sample indicates the production of damage in the buried layer located at the Bragg peak; compared with the HRXRD peak of a virgin sample, the peak obtained from the 58Ni¹⁹⁺-irradiated LiTaO₃ sample exhibited a clear shift to the lower-angle side and broadening behavior. Ion irradiation increased the lattice constant and interplanar spacing, and caused lattice swelling and tensile strain in the track damage region. The HRXRD peak shifted substantially to a lower diffraction angle with an increasing number of connected defects [47]. Thus, with the increase in irradiating 58Ni¹⁹⁺ fluence, the irradiation-induced defects and track damage inside the LiTaO₃ crystal increased, and the corresponding peak significantly shifted to a lower-angle side. The structural characterizations obtained from RBS/channeling and HRXRD measurements further confirm the above discussions based on the prism coupling results. In order to describe the accumulation of irradiation-induced damage, one LiTaO₃ sample was further calculated utilizing a classical approximate expression, \( f_d = (\chi_i - \chi_v) / (\chi_r - \chi_v) \), where \( \chi_i, \chi_v \), and \( \chi_r \) are the backscattering yields of the irradiated sample under the channeling condition, and the virgin sample along the channeling direction and with a random orientation, respectively [48]. The Ta sublattice disorders were further calculated utilizing a classical approximate expression. However, the obtained disorder levels of the Ta sublattice are not sufficient to determine the trend between the lattice disorder and irradiating ion fluence. Therefore, one LiTaO₃ sample was further irradiated with 58Ni¹⁹⁺ to a fluence of 9 × 10¹² cm⁻². The obtained relative disorder of the Ta sublattice at the LiTaO₃ surface region as a function of the irradiating 58Ni¹⁹⁺ fluence (damage accumulation curve) is shown in figure 3(a). In combination with the prism coupling measurements, the relationship between Ta sublattice disorder and refractive index corresponding to different wavelengths is fitted by a polynomial expression, as shown in figure 3(b). Swift heavy ion irradiation can modify the refractive index to facilitate a better understanding of the physical mechanisms of ion–matter interactions, which is essential for designing materials with new functionality for technological applications of novel devices. In this work, the obtained quantitative relationship between the irradiation fluence (lattice disorder) and the refractive index provides the necessary data and evidence for the optical applications of swift heavy ion irradiation.

The damage in KTaO₃ and LiTaO₃ crystals induced by swift Ni¹⁹⁺ irradiation was directly observed by utilizing the TEM technique. Cross-sectional TEM images taken of the surface and peak electronic energy loss regions in the samples
are shown in figures 4(a)–(d), for KTaO3 and figures 5(a)–(d) for LiTaO3. The corresponding selected-area electron diffraction patterns shown in the insets qualitatively reflect the irradiation-induced levels of lattice disorder. The noise-filtered images corresponding to the HRTEM images in figures 4(b) and (d) and 5(b) and (d) are shown in figures 4(e) and (g) and 5(e) and (g). Schematic diagrams of the different damage morphologies are also shown in figures 4(f) and (h) and 5(f) and (h). First, concerning the KTaO3 crystal, as shown in figures 4(a)–(f), very few individual spherical defects with diameter Φ = ~2.0 nm were produced in the surface region under the action of 358 MeV $^{58}$Ni$^{19+}$ irradiation ($E_{\text{ion}} = 6.17 \text{ MeV amu}^{-1}, S_e = \sim 12.0 \text{ keV nm}^{-1}$). As shown in figures 4(c), (d), (g) and (h), accompanying the reduced ion energy and increased electronic energy loss [49] along the ion penetration path, the number of spherical defects in the peak electronic energy loss region clearly increased, and under the action of 125 MeV $^{58}$Ni$^{19+}$ irradiation ($E_{\text{ion}} = 2.15 \text{ MeV amu}^{-1}, S_e = \sim 13.8 \text{ keV nm}^{-1}$), the neighboring spherical defects began to overlap, thus forming a region with discontinuous track damage with diameter Φ = ~2.5 nm. Second, concerning the LiTaO3 crystal [7, 50], as shown in figures 5(a)–(f), both spherical defects and discontinuous track damage regions with diameter Φ = ~3.0 nm were produced in the surface region [7, 50] under the action of 358 MeV $^{58}$Ni$^{19+}$ irradiation ($E_{\text{ion}} = 6.17 \text{ MeV amu}^{-1}, S_e = \sim 12.0 \text{ keV nm}^{-1}$). As shown in figures 5(c), (d), (g) and (h), owing to the reduced ion energy and increased electronic energy loss, the track damage in the peak electronic energy loss region became continuous and homogeneous, finally forming a continuous region of track damage with diameter Φ = ~4.5 nm under the action of 125 MeV $^{58}$Ni$^{19+}$ irradiation ($E_{\text{ion}} = 2.15 \text{ MeV amu}^{-1}, S_e = \sim 13.8 \text{ keV nm}^{-1}$). Clearly, the latent track damage behavior depends directly on the different crystals, ion energies and electronic energy losses.

3.2. Numerical calculations of track damage formation with the iTS model

The mechanism of latent track damage in ion-irradiated alkali tantalate crystals is now further discussed using the iTS model [43, 44]. Electronic energy deposition from irradiating ions onto target electrons results in an electron cascade via electron–electron interactions. Due to the difference between the electron temperature and lattice temperature, the deposited energy is then transferred from the hot electron subsystem to the cold lattice subsystem through electron–phonon coupling, leading to a local increase in temperature along the ion trajectory. Once the electronic energy loss exceeds a certain threshold, local melting appears, and via a subsequent rapid quenching process a latent track containing a region of lattice damage and an amorphous volume can be formed. The energy and temperature evolutions of the electron and lattice subsystems can be numerically calculated by utilizing the following classical heat diffusion equations:

\[
C_e \left(T_e\right) \frac{\partial T_e}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left[r K_e(T_e) \frac{\partial T_e}{\partial r}\right] - g \left(T_e - T_a\right) + A(r,t)
\]

\[
C_a \left(T_a\right) \frac{\partial T_a}{\partial t} = -\frac{1}{r} \frac{\partial}{\partial r} \left[r K_a(T_a) \frac{\partial T_a}{\partial r}\right] + g \left(T_e - T_a\right)
\]

where $T_e$ and $T_a$, $C_e$ and $C_a$, and $K_e$ and $K_a$ are the temperatures, specific heat coefficients and thermal conductivities of the electron and lattice subsystems, respectively, $g$ is the electron–phonon coupling parameter and $A(r,t)$ describes the spatio-temporal energy deposition from the irradiating ion.

![Figure 3.](image-url) Relative disorder of the Ta sublattice at the LiTaO3 surface as a function of (a) irradiating Ni$^{19+}$ fluence and (b) surface refractive index.
onto the electron subsystem. The $K_e$ and $C_e$ profiles of KTaO$_3$ and LiTaO$_3$ crystals as a function of lattice temperature $T_a$ used in the calculation process are shown in figures 6(a) and (b) [51–56], respectively. For both KTaO$_3$ and LiTaO$_3$, $C_e$ was set to 1.0 J cm$^{-3}$ K$^{-1}$ and $K_e$ was set to 100 W m$^{-1}$ K$^{-1}$, because $K_e = C_e \times D_e$ [44], where $D_e$ is the electron diffusivity (1.0 cm$^2$ s$^{-1}$) [57, 58]. In [44], based on the experimental results for different materials, the relationship between $\lambda$ and band gap was fitted. The fitting result indicates that the value of $\lambda$ appears to be directly related to the inverse of the band gap energy. Based on the band gap energies of KTaO$_3$ (3.6 eV) and LiTaO$_3$ (4.7 eV) [35], the values of the electron–phonon mean free path $\lambda$ could be deduced according to [44]: in this work these were 4.7 nm for KTaO$_3$ and 4.2 nm for LiTaO$_3$. Thus, because $\lambda^2 = C_e \times D_e / g$, the $g$ values of the KTaO$_3$ and LiTaO$_3$ crystals were set to 4.5 $\times$ 10$^{18}$ W m$^{-3}$ K$^{-1}$ and 5.7 $\times$ 10$^{18}$ W m$^{-3}$ K$^{-1}$, respectively.

The spatio-temporal evolution of the energy deposition densities and lattice temperatures induced by the action of the electronic energy losses were numerically calculated. As shown in figures 6(c)–(f), the lattice temperatures in the surface and peak electronic energy loss regions of the Ni$^{19+}$-irradiated KTaO$_3$ and LiTaO$_3$ crystals increased to 3410 K and 4140 K and 3250 K and 4070 K, respectively, substantially exceeding the melting points (1625 K for KTaO$_3$ and 1923 K for LiTaO$_3$) and leading to the formation of a molten phase. During the subsequent cooling process, due to recrystallization occurring at the cylindrical crystal–melt interfaces, the radii of the experimentally observed latent tracks (individual spherical defects and discontinuous and continuous cylindrical damage zones shown in figures 4 and 5) are smaller than the radii of the calculated molten regions shown in figures 6(e) and (f).

For different penetrating ion energies and electronic energy losses, different energy deposition densities and lattice temperature increments will be achieved, leading to different damage responses and latent track morphologies in KTaO$_3$ and LiTaO$_3$ crystals. Unlike the well-understood production of continuous tracks by relatively high electronic energy losses, the formation of individual spherical defects and discontinuous tracks in the irradiated regions is far more complicated. It can be ascribed to statistical fluctuations of the ion charge state owing to electron capture and loss processes along the ion penetration path, and, in this study, the subsequently induced non-homogeneous energy deposition [59, 60]. The statistical fluctuations of the ion charge are immediately followed by corresponding fluctuations of the momentary energy loss ($E_{mom}$): if $E_{mom}$ is only slightly higher
than the threshold value \( E_{th} \) for damage formation, then the capture of electrons by a penetrating ion would reduce \( E_{mom} \) to below \( E_{th} \), resulting in an undamaged lattice structure; if \( E_{mom} \) is only slightly lower than \( E_{th} \), then the loss of electrons by a penetrating ion would increase \( E_{mom} \) to above \( E_{th} \), leading to the production of lattice damage. In this way, when the electronic energy loss is around \( E_{th} \) for damage formation, individual spherical defects can be formed; when accompanied by increasing electronic energy loss, discontinuous and continuous latent tracks would be formed. Thus, as shown in figures 4(a) and 5(a), under \(^{58}\text{Ni}^{19+}\) irradiation with an energy of 6.17 MeV amu\(^{-1}\) (surface regions), very few individual spherical defects are produced in the surface region of the KTaO\(_3\) crystal, and an \( E_{th} \) of \( \sim 12.0 \text{ keV nm}^{-1} \) for formation of individual spherical defects in a KTaO\(_3\) crystal is determined in the present work, indicating that the corresponding lattice temperature calculated by the iTS model is the lattice temperature threshold for irradiation damage. Both spherical defects and discontinuous regions of track damage are produced in the surface region of the LiTaO\(_3\) crystal, and the related \( E_{th} \) for a LiTaO\(_3\) crystal should be slightly less than 12.0 keV nm\(^{-1}\), indicating that the corresponding lattice temperature calculated by the iTS model is slightly higher than the lattice temperature threshold. Accompanying the reduced ion energy and increased electronic energy loss along the ion penetration path, more significant track damage was produced in the peak electronic energy loss region than in other regions. The \( E_{th} \) for track formation is also dependent on ion velocity [49] and decreases upon reducing the ion velocity. Therefore, as shown in figures 4(c) and 5(c), under ion irradiation with an energy of 2.15 MeV amu\(^{-1}\) (peak electronic energy loss regions), the \( E_{th} \) for individual spherical defect formation in KTaO\(_3\) and LiTaO\(_3\) crystals should be further reduced and somewhat less than 12.0 keV nm\(^{-1}\), and the electronic energy loss of \( \sim 13.8 \text{ keV nm}^{-1} \) in the present work leads to overlapping spherical defects and the formation of discontinuous tracks in the KTaO\(_3\) crystal and continuous tracks in the LiTaO\(_3\) crystal. In the iTS model, compared with the energies of irradiating ions and electronic energy losses, the induced lattice-temperature evolution is more fundamental and essential to understanding the track damage behavior. As shown in figures 6(e) and (f), the related lattice temperature evolution demonstrates that, compared with the melting points (1625 K for KTaO\(_3\) and 1923 K for LiTaO\(_3\)), a much higher lattice temperature in the region surrounding the ion path needs to be achieved to produce stable track damage due to the non-negligible effect of melting damage caused by annealing during

\[ \text{Figure 5.} \quad \text{(a)--(d) Bright-field cross-sectional TEM images and (e)--(h) corresponding noise-filtered images and schematic diagrams of damage morphologies in the surface and peak electronic energy loss of the } \text{Ni}^{19+}-\text{irradiated LiTaO}_3 \text{ sample.} \]
the cooling process. Thus, a lattice-temperature threshold for the production of track damage is introduced: 3410 K for the KTaO$_3$ crystal and slightly less than 3250 K for the LiTaO$_3$ crystal. The different track behaviors in alkali tantalate crystals under different irradiation conditions (ion energies, velocities and electronic energy losses) could be comparatively studied by calculating and comparing the irradiation-induced spatio-temporal evolution of the lattice temperature with the lattice temperature threshold determined in this work.

4. Conclusions

In this work, employing 358 MeV $^{58}$Ni$^{19+}$ irradiation to produce intense electronic energy loss, the induced damage responses and latent track morphologies in alkali tantalate crystals (KTaO$_3$ and LiTaO$_3$) were studied. The utilized prism coupling technique has allowed us to obtain a complete refractive index profile, turning this into an effective and accurate method for studying damage behavior. The analysis of the irradiated region is rapidly conducted, providing a basic understanding of irradiation-induced damage behavior in various crystals. According to the iTS model calculations, the spatio-temporal evolution of energy deposition densities and lattice temperatures reflects the experimentally observed latent tracks in Ni$^{19+}$-irradiated alkali tantalate crystals. Compared with the melting point, a much higher lattice temperature in the region surrounding the ion path needs to be achieved to produce stable track damage due to the non-negligible effect of melting damage caused by annealing during the cooling process. More remarkable is the observation of individual and overlapped spherical defects and discontinuous
and continuous tracks induced by different electronic energy losses and ion energies; thus the threshold values for the production of track damage with different morphologies were determined. In addition, numerical calculations of the iTS model point out that the lattice temperature along the ion penetration path increases to 3410 K and 3250 K. Therefore, a lattice temperature threshold for track production, which is more fundamental and essential than the energy of irradiating ions and electronic energy loss, is introduced and assessed to be 3410 K for KTaO3 and slightly less than 3250 K for LiTaO3. Compared with the LiTaO3 crystal, a critical threshold value is found for the KTaO3 crystal, indicating relatively high damage tolerance behavior under the action of extreme electronic energy loss. Our experimental findings and numerical calculations involving the latent tracks not only facilitate a better understanding of ion–solid interactions but also contribute to a foundation for ion-irradiation applications, such as the design and fabrication of nanoscale-to-microscale structures in crystals through swift heavy ion irradiation and subsequent selective chemical etching techniques, tailoring new advantageous features for novel photonic applications.

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