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Latent Tracks in Ion-Irradiated LiTaO₃ Crystals: Damage Morphology Characterization and Thermal Spike Analysis

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Received: 2 September 2020; Accepted: 26 September 2020; Published: 28 September 2020



Abstract: Systematic research on the response of crystal materials to the deposition of irradiation energy to electrons and atomic nuclei has attracted considerable attention since it is fundamental to understanding the behavior of various materials in natural and manmade radiation environments. This work examines and compares track formation in LiTaO₃ induced by separate and combined effects of electronic excitation and nuclear collision. Under 0.71–6.17 MeV/u ion irradiation with electronic energy loss ranging from 6.0 to 13.8 keV/nm, the track damage morphologies evolve from discontinuous to continuous cylindrical zone. Based on the irradiation energy deposited via electronic energy loss, the subsequently induced energy exchange and temperature evolution processes in electron and lattice subsystems are calculated through the inelastic thermal spike model, demonstrating the formation of track damage and relevant thresholds of lattice energy and temperature. Combined with a disorder accumulation model, the damage accumulation in LiTaO₃ produced by nuclear energy loss is also experimentally determined. The damage characterizations and inelastic thermal spike calculations further demonstrate that compared to damage-free LiTaO₃, nuclear-collision-damaged LiTaO₃ presents a more intense thermal spike response to electronic energy loss owing to the decrease in thermal conductivity and increase in electron–phonon coupling, which further enhance track damage.

Keywords: latent track damage; irradiation damage model; energy loss process; coupled effect

1. Introduction

Recent research has demonstrated that perovskite-type LiTaO₃ crystals, as representative functional oxides, have extensive potential applications in various radiation environments, such as tritium breeding in fusion reactors [1,2], nuclear waste immobilization [3], and radiation detection [4]. In the research field of ion–solid interactions and irradiation effects, systematic study of the response of crystal materials to the deposition of irradiation energy to electrons and atomic nuclei has always attracted considerable attention. As a research focus, understanding and clarifying the relevant physical mechanisms is not only fundamental to predicting and evaluating the damage behaviors of

materials in natural and man-made radiation environments [5], but it is also key to the applications of ion and electron beam techniques in atomic-level defect manipulation, material modification, and micro/nanofabrication [6–16].

In this work, through appropriate choices of irradiating ion energies, velocities, and energy loss intensities, the role of irradiation energy deposited to electron and lattice subsystems and the induced lattice-defect production and structure evolution in LiTaO₃ crystals are studied comprehensively. By combining the microstructure characterizations with inelastic thermal spike model calculations, the formation of latent tracks with different damage morphologies under the action of electronic energy loss and the corresponding threshold conditions are analyzed. Based on a disorder accumulation model [17,18], the damage accumulation in LiTaO₃ produced by the nuclear energy loss process is also determined. Then, the coupled effect between the electronic excitation and nuclear collision on the thermal spike response and the induced enhanced track damage behavior are further experimentally characterized and theoretically demonstrated.

2. Materials and Methods

2.1. Energetic Ion Irradiation and Damage Characterization

Optically polished single-crystal LiTaO₃ samples with a (006) surface normal zone axis direction were used in this work. Irradiation experiments with 1 MeV (0.005 MeV/u) ¹⁹⁷Au⁺ and 20 MeV (0.71 MeV/u) ²⁸Si³⁺ at 300 K were performed utilizing a 1.7 MV tandem accelerator (NEC, Middleton, USA) and a 6 MV tandem accelerator (HVEE, Amersfoort, The Netherlands), respectively, within the State Key Laboratory of Nuclear Physics and Technology, Peking University. Irradiation experiments with 247 MeV (6.17 MeV/u) ⁴⁰Ar¹²⁺ and 358 MeV (6.17 MeV/u) ⁵⁸Ni¹⁹⁺ at 300 K were performed at the Heavy Ion Research Facility in Lanzhou (HIRFL), Institute of Modern Physics, Chinese Academy of Sciences. Relatively low ion-beam current density and ion flux ($8.3 \times 10^{10} \text{ cm}^{-2} \cdot \text{s}^{-1}$ for Au⁺, $1.9 \times 10^{10} \text{ cm}^{-2} \cdot \text{s}^{-1}$ for Si³⁺, $6.5 \times 10^{10} \text{ cm}^{-2} \cdot \text{s}^{-1}$ for Ar¹²⁺, and $8.1 \times 10^{10} \text{ cm}^{-2} \cdot \text{s}^{-1}$ for Ni¹⁹⁺) were maintained during the irradiation process to avoid undesired heating and charge accumulation effects on the LiTaO₃ samples. Rutherford backscattering (RBS)/channeling analysis was carried out with the abovementioned 1.7 MV tandem accelerator. A 2.0 MeV He⁺ beam was extracted, and a Si detector located at a scattering angle of 160° relative to the He⁺ beam was used to collect the backscattered He⁺ signal and assess the irradiation damage. Cross-sectional LiTaO₃ samples for transmission electron microscopy (TEM) observations were prepared through polishing, dimpling, and Ar-ion milling with a Gatan 695 precision polishing system (Gatan Inc., Pleasanton, USA), and the corresponding irradiation damage regions were characterized utilizing an FEI Tecnai G2 F20 transmission electron microscope (FEI Co., Hillsboro, OR, USA).

2.2. Simulations and Calculations of Irradiation Damage Models

As indicated in Table 1, the electronic/nuclear energy losses ($E_{\text{ele}}/E_{\text{nucl}}$) and displacements per atom (dpa) in LiTaO₃ induced by energetic Au⁺, Si³⁺, Ar¹²⁺, and Ni¹⁹⁺ irradiation were determined through the stopping and range of ions in matter (SRIM) 2013 simulation code with full-damage cascade mode [19,20], in which the density and default displacement energies of Li, Ta, and O atoms were set to 7.45 g·cm^{−3}, 25 eV, 25 eV, and 28 eV, respectively [21,22]. Based on the irradiation energy deposited by electronic energy loss, the induced energy exchange and diffusion and temperature evolution processes in the electron and lattice subsystems of the LiTaO₃ crystal were numerically calculated through two heat-conduction differential equations described in the inelastic thermal spike (iTS) model [23–25]. In the iTS model, the energy exchange between the electronic and lattice subsystems is described by a set of thermal diffusion equations, one for the electronic subsystem (Equation (1)) and one for the atomic subsystem (Equation (2))

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

$$C_a(T_a) \frac{\partial T_a}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left[r K_a(T_a) \frac{\partial T_a}{\partial r} \right] + g(T_e - T_a) \quad (2)$$

where the thermal conductivity K_a and specific heat coefficient C_a of the lattice subsystem versus lattice temperature were extracted from [26]; the thermal conductivity K_e and specific heat coefficient C_e of the electron subsystem were fixed to $100 \text{ W m}^{-1} \cdot \text{K}^{-1}$ and $1.0 \text{ J cm}^{-3} \cdot \text{K}^{-1}$, respectively, owing to the constant thermodynamic parameters in the electron subsystem [23,24]; the electron-phonon mean free path λ was deduced from the band gap (4.7 eV) and set to 4.2 nm [27]; the electron-phonon coupling parameter g was set to $5.7 \times 10^{18} \text{ W} \cdot \text{m}^{-3} \cdot \text{K}^{-1}$ [26]; the electron-phonon mean free time τ was $1.75 \times 10^{-13} \text{ s}$ owing to $\tau = C_e/g$ [28]; and the latent heat of fusion was set to 150 J/g . The term $A(r, t)$ describes the spatial and temporal energy deposition from the incident ion to the electrons within 10^{-17} – 10^{-15} s , and the electrons would deposit their energy close to the ion path within 10^{-15} s [24,29]. The damage accumulation in LiTaO_3 produced by nuclear energy loss originating from low-energy ion irradiation was also fitted through a disorder accumulation model.

Table 1. SRIM simulation results corresponding to different ion-irradiation conditions.

Ion-Irradiation Conditions			Surface Region				E_{ele} -Peak Region				
species	flux	fluence	energy	velocity	E_{ele}	dpa	depth	energy	velocity	E_{ele}	dpa
	($\text{cm}^{-2} \cdot \text{s}^{-1}$)	(cm^{-2})	(MeV)	(MeV/u)	(keV/nm)		(μm)	(MeV)	(MeV/u)	(keV/nm)	
$^{28}\text{Si}^{3+}$	1.9×10^{10}	1×10^{13}	20	0.71	6.2	1.6×10^{-4}	0	20	0.71	6.2	1.6×10^{-4}
$^{40}\text{Ar}^{12+}$	6.5×10^{10}	3×10^{12}	247	6.17	6.0	2.1×10^{-5}	30.0	46	1.15	8.4	6.4×10^{-5}
$^{58}\text{Ni}^{19+}$	8.1×10^{10}	3×10^{12}	358	6.17	12.0	2.9×10^{-5}	18.0	125	2.15	13.8	8.0×10^{-5}
Ion-Irradiation Conditions			Surface Region				dpa-Peak Region				
species	flux	fluence	energy	velocity	E_{nucl}	dpa	depth	energy	velocity	E_{nucl}	dpa
	($\text{cm}^{-2} \cdot \text{s}^{-1}$)	(cm^{-2})	(MeV)	(MeV/u)	(keV/nm)		(μm)	(MeV)	(MeV/u)	(keV/nm)	
$^{197}\text{Au}^{+}$	8.3×10^{10}	1×10^{14}	1	0.005	5.3	0.24	0.1	0.35	0.0018	5.4	0.71

3. Results and Discussion

3.1. Latent Ion Tracks Induced by Electronic Energy Loss

As mentioned above, SRIM 2013 simulation code is primarily used to determine the profile of electronic energy loss as a function of ion penetration path, and could not provide the radial distribution of deposited irradiation energy via electronic energy loss process at a certain penetration depth, which actually is essential for the iTS calculations so as to obtain the radial evolutions of electron and lattice temperatures and further describe the lattice melting behavior. During the ion slowing process, the interaction cross section between the irradiating ion and target electrons and the subsequently induced electron cascade in the target directly depend on the ion velocity, which would further determine the radial distribution of the energy deposited in the electron subsystem via the electronic energy loss process. Considering a cylindrical volume in which 0.66 of the total electronic energy is deposited, the corresponding cylindrical radius (radial distance from the ion trajectory) is defined as the absorption radius α_e to describe the energy distribution profile [24]. The value of α_e would increase with increasing ion velocity (velocity effect), resulting in a large energy spread region and locally low energy deposition density [24]. In this work, as shown in Figure 1a, the α_e of the LiTaO_3 crystal used for electronic energy deposition increases from 2.1 to 4.7 nm as the ion velocity increases from 0.71 to 6.17 MeV/u. Under ion irradiation with velocities of 0.71, 1.15, 2.15, and 6.17 MeV/u, the fraction of irradiation energy deposited on the electron subsystem derived from the Katz model and MC calculations [24,29,30] and the corresponding normalized energy deposition distribution $F(r)$ versus the radial distance from the ion trajectory are shown in Figure 1b. Then, considering the SRIM-simulated electronic energy loss (E_{ele}) relative to the ion velocity and ensuring that the integral of the energy deposition density over the radial area equals E_{ele} [24], the determined distribution profile of

the energy deposition density in the electron subsystem is shown in Figure 1c, illustrating that during the ion irradiation process, for a similar E_{ele} (6.0–6.2 keV/nm), the deposition density of electronic energy would accordingly increase with decreasing ion velocity from 6.17 to 0.71 MeV/u (orange and wine curves in Figure 1c). For the same ion velocity (6.17 MeV/u), the deposition density of electronic energy would accordingly increase with increasing E_{ele} from 6.0 to 12.0 keV/nm (orange and blue curves in Figure 1c). For the ion (1.15 MeV/u) with an E_{ele} of 8.4 keV/nm and the ion (6.17 MeV/u) with an E_{ele} of 12.0 keV/nm, the deposition density of electronic energy in the cylindrical core area are similar (pink and blue curves in Figure 1c). For the ion (2.15 MeV/u) with an E_{ele} of 13.8 keV/nm, the deposition density of electronic energy reaches the maximum (green curve in Figure 1c).

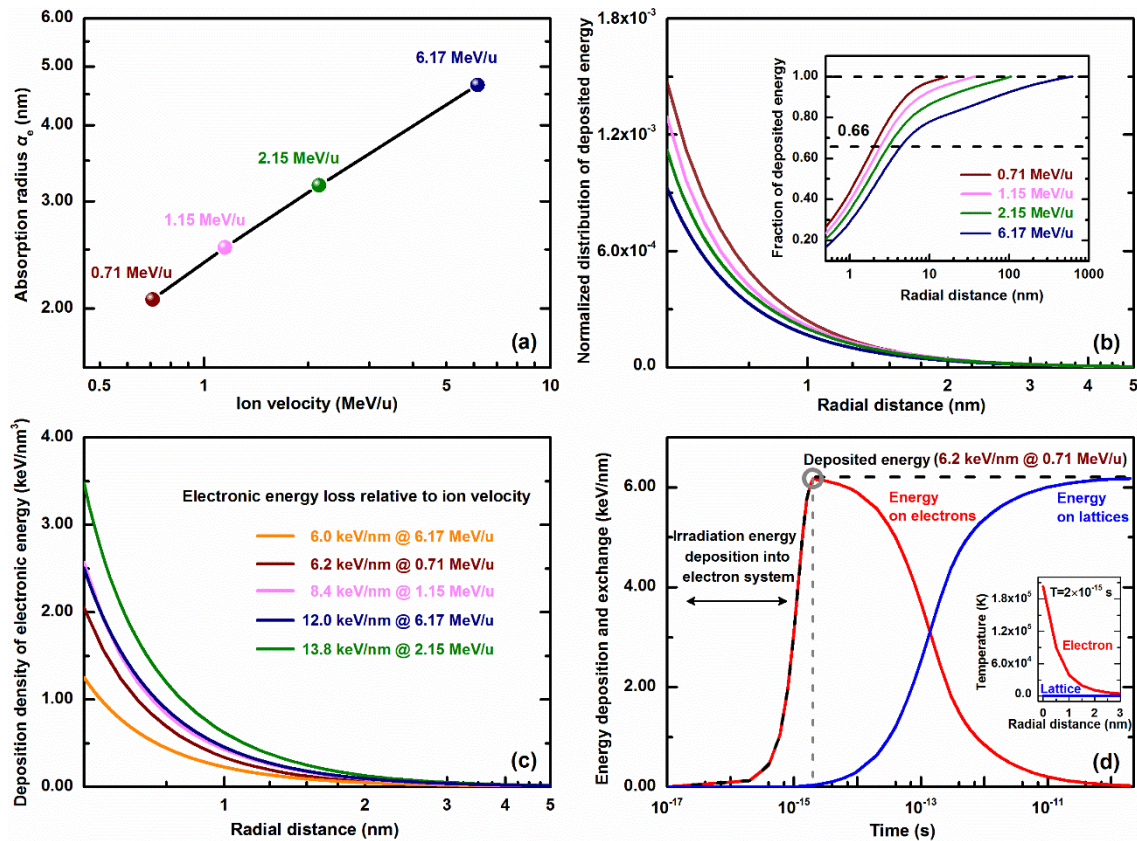


Figure 1. (a) Absorption radius α_e of LiTaO₃ crystal for energy deposition via electronic energy loss as a function of ion velocity. (b) Normalized distribution of irradiation energy deposited to electron subsystem as a function of radial distance from ion trajectory. The fraction of energy deposited to the electron subsystem versus radius is also shown in the inset. (c) Density distribution of energy deposited to the electron subsystem via ion irradiation with different ion velocities and electronic energy losses. (d) Irradiation energy deposition and subsequent energy exchange processes between electron and lattice subsystems calculated by inelastic thermal spike model. The radial temperature distributions of the electron and lattice subsystems at 2×10^{-15} s are also shown in the inset.

Under the action of electronic energy deposition, the induced energy exchange and diffusion and temperature evolution in electron and lattice subsystems are discussed utilizing the inelastic thermal spike (ITS) model [23–25]. Figure 1d illustrates the ITS-calculated energy evolution in a LiTaO₃ crystal under 0.71 MeV/u ion irradiation with an E_{ele} of 6.2 keV/nm. As shown, the kinetic energy of the incident ion is gradually deposited into the electronic subsystem via electronic energy loss, reaching a peak (6.2 keV/nm) at approximately 10^{-15} s. The radial distribution of the energy deposition density in the electron subsystem at this time is described by the wine curve in Figure 1c. Then, as shown in the inset of Figure 1d, owing to the much higher temperature in the electron subsystem than in

the lattice subsystem, the deposited electronic energy is gradually transferred from the hot electronic subsystem to the cooler lattice through electron-phonon coupling, leading to significant evolution of the electronic and lattice temperatures; once the energy transferred and deposited to lattice atoms exceeds a certain threshold, local melting occurs, and lattice damage along the ion trajectory is formed via a subsequent rapid quenching process. In this energy exchange stage, the sum of irradiation energy deposited into the electron and lattice subsystems remains constant (6.2 keV/nm). Finally, due to the decrease in electron temperature and increase in lattice temperature, the energy exchange driven by the temperature difference ceases to occur since the temperatures between the electron and lattice subsystems reach equilibrium within approximately 10^{-10} s.

As shown in Figure 2, the temporal evolution profiles of the energy deposition to atoms (Figure 2a,d,g,j,m) and the temporal evolution of the atomic temperature (Figure 2b,e,h,k,n) are shown for the different irradiation conditions at different radii (distance from the ion path), obtained by the iTS model [23–25]. The related cross-sectional TEM images in LiTaO₃ under ion irradiation with different ion velocities and electronic energy losses are also presented (Figure 2c,f,i,l,o). For 0.71 MeV/u Si³⁺ irradiation with an E_{ele} of 6.2 keV/nm, the energy deposited to the lattice atoms within a 1 nm radius exceeds the criterion E_m for melting phase formation, and the corresponding lattice temperature exceeds the melting temperature T_m . It should be pointed out that, according to the distribution of specific heat C_a as a function of the temperature T_a in LiTaO₃ crystal [26], the E_m for melting phase formation is calculated to be 0.54 eV/atom (1113 J/g) via $E_m = \int_{T_0}^{T_m} C_a(T_a) dT_a + E_l$, where T_0 is set to 300 K, and the latent heat of fusion E_l is set to 150 J/g [31]. This results in the formation of discontinuous tracks with a radius of ~1.0 nm, as shown in Figure 2c. For 6.17 MeV/u Ar¹²⁺ irradiation with an E_{ele} of 6.0 keV/nm, the energy deposited to the lattice atoms along the ion path reaches a peak at 0.47 eV/atom within $\sim 10^{-13}$ s, and the corresponding atomic temperature increases to peak at 1860 K, which is still lower than the melting temperature T_m (1923 K). Therefore, as shown in Figure 2f, no melting phase or lattice damage would occur [32]. For 1.15 MeV/u Ar¹²⁺ irradiation (E_{ele} of 8.4 keV/nm) and 6.17 MeV/u Ni¹⁹⁺ irradiation (E_{ele} of 12.0 keV/nm), accompanied by increasing energy deposition on the lattice atoms, the radii of the produced cylindrical melting zones increase to ~1.5 nm and ~2.0 nm, respectively. As shown in Figure 2i,l, this results in the formation of discontinuous tracks of larger size, with ~1.5 nm and ~2.0 nm diameter, respectively. In the case of 2.15 MeV/u Ni¹⁹⁺ irradiation with an E_{ele} of 13.8 keV/nm, the energy deposited to the lattice atoms in the cylinder center reaches 1.20 eV/atom, significantly exceeding the melting criterion, and lattice melting occurs in a cylindrical region with a radius of ~3.0 nm. Owing to the much higher distributions of atomic energy and temperature, continuous track damage with a radius of ~2.5 nm shown in Figure 2o is formed during the subsequent rapid quenching process [26].

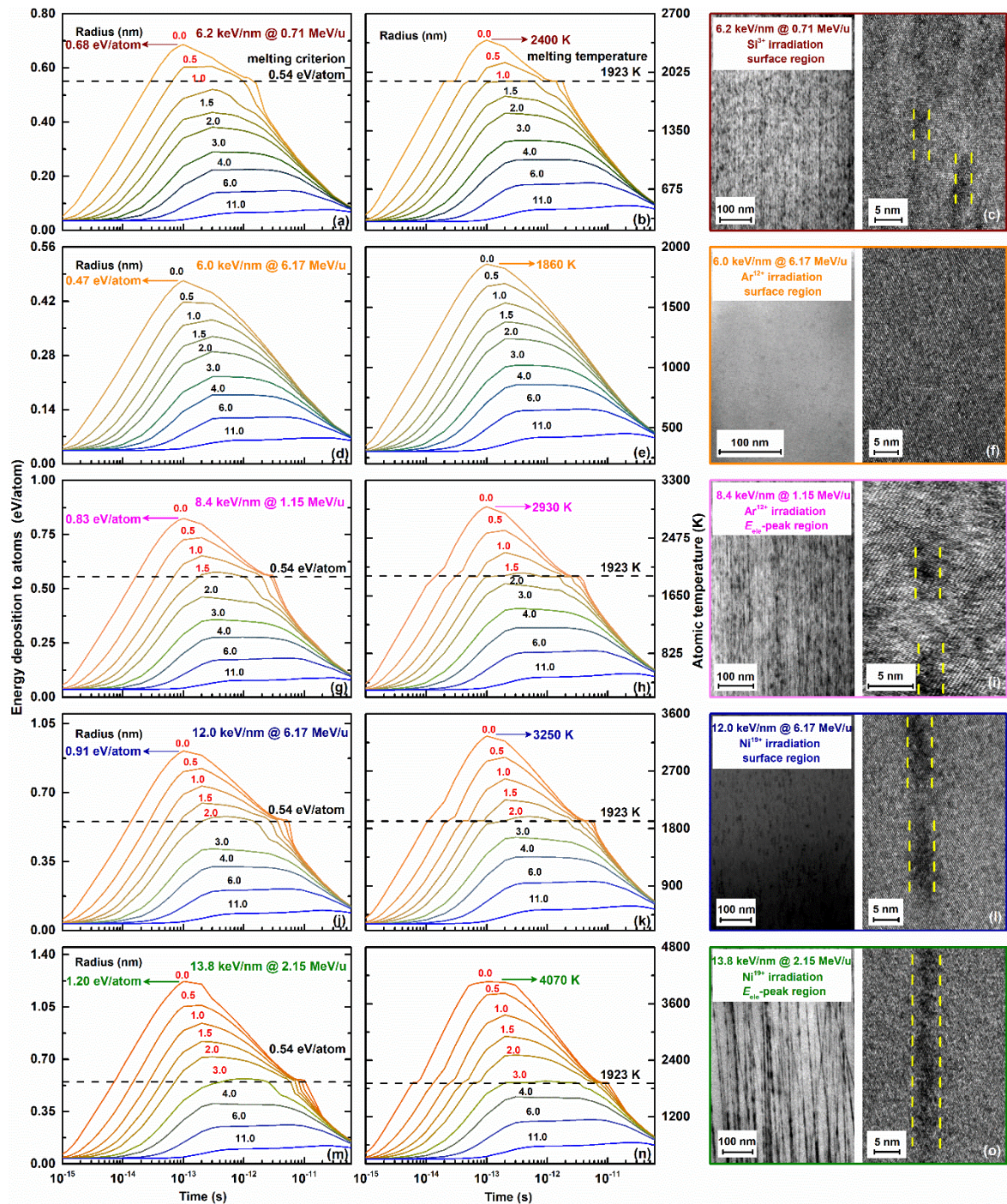


Figure 2. (a,d,g,j,m) Energy deposited to atoms and (b,e,h,k,n) corresponding evolution of atomic temperature at different radii versus time under the action of ion irradiation with different ion velocities and electronic energy losses. (c,f,i,l,o) TEM-observed latent tracks with different morphologies in LiTaO₃ under different irradiation conditions.

3.2. Damage Accumulation Induced by Nuclear Energy Loss

Utilizing 1 MeV Au⁺ irradiation, the damage production and accumulation of LiTaO₃ crystals due to nuclear energy loss only, originating from the elastic collisions between irradiating ions and target atomic nuclei, were determined. The SRIM-simulated nuclear energy loss (E_{nuc}) profile and displacements per atom (dpa) profile corresponding to the Au⁺ fluence of $1 \times 10^{14} \text{ cm}^{-2}$ are shown in Figure 3a, illustrating that the dpa peak at 0.71 is located at a depth of ~100 nm. The measured

RBS/channeling spectra of Au^+ -irradiated LiTaO_3 under different fluences are shown in Figure 3b, and then, based on an iterative procedure [33], the profiles of Ta sublattice disorder as a function of depth are calculated and shown in Figure 3c, indicating the long-range disorder in LiTaO_3 under Au^+ irradiation to the local dpa of 0.25. The Ta sublattice disorder in the damage peak region versus the local irradiation dose (dpa) is further plotted in Figure 3d, and the obtained damage accumulation curve is described by a disorder accumulation model [17,18], where the irradiation-induced disorder S (black curve) measured by the RBS/channeling technique would arise from the sum of two contributions: amorphous fraction f_a (blue curve) and interstitials and small interstitial clusters in residual crystalline regions S_d (red curve). The related fitting parameters—including the amorphization cross section σ_a , the effective cross section σ_s for defect-stimulated amorphization, the saturation value S_d^* for the defect-induced disorder, and the coefficient B proportional to an effective defect recombination volume—are also indicated.

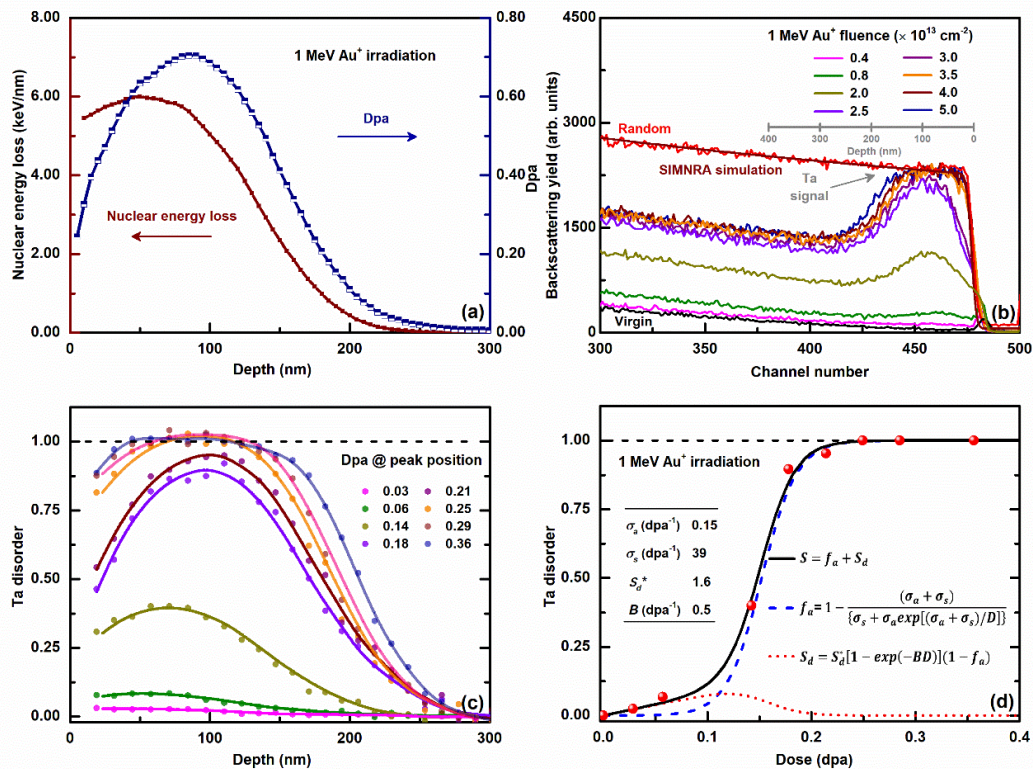


Figure 3. (a) SRIM-simulated nuclear energy loss and dpa profiles in Au^+ -irradiated LiTaO_3 corresponding to a fluence of $1 \times 10^{14} \text{ cm}^{-2}$. (b) RBS/channeling spectra of LiTaO_3 samples irradiated with Au^+ at different fluences. (c) Disorder profiles of the Ta sublattice in Au^+ -irradiated LiTaO_3 samples. (d) Relative disorder of the Ta sublattice at the damage peak versus local dose (dpa) and data fitting through the disorder accumulation model.

3.3. Enhanced Track Damage Induced by Coupled Nuclear and Electronic Energy Losses

To study the coupled effects of electronic and nuclear energy losses, the Au^+ -irradiated LiTaO_3 sample with a fluence of $2 \times 10^{13} \text{ cm}^{-2}$ was subsequently irradiated with 20 MeV (0.71 MeV/u) Si^{3+} to a fluence of $1 \times 10^{13} \text{ cm}^{-2}$. The measured RBS/channeling spectra, corresponding Ta disorder profiles, cross-sectional TEM images, and FFT patterns of LiTaO_3 crystals under different irradiation conditions are shown in Figure 4, and they are used to discuss the E_{ele} -induced track damage response in pristine and E_{nuc} -damaged LiTaO_3 . According to the RBS/channeling spectra shown in Figure 4a, the determined disorder profiles of the Ta sublattice in ion-irradiated LiTaO_3 samples shown in Figure 4b,e,h quantitatively indicated that in contrast to pristine LiTaO_3 , the same electronic energy loss induced by Si^{3+} irradiation in E_{nuc} -damaged LiTaO_3 leads to complete disorder in the region at a

depth of ~ 100 nm, demonstrating the additive effect of electronic and nuclear energy losses on damage production. Compared to pristine LiTaO_3 , the enhanced track damage response in the predamaged LiTaO_3 upon electronic energy loss is also confirmed by TEM observations. As shown in Figure 4b,c, the lattice defects and defect clusters were first produced by nuclear energy loss via Au^+ irradiation; then, upon subsequent 0.71 MeV/u Si^{3+} irradiation, the introduced E_{ele} approximatively locates at the Bragg peak position, and remains constant (6.2 keV/nm) in the near surface region; finally, the induced track damage in the corresponding E_{nuc} -damaged region is shown in Figure 4h,i. This damage is much more significant than the E_{ele} -induced damage in pristine LiTaO_3 (Figure 4e,f).

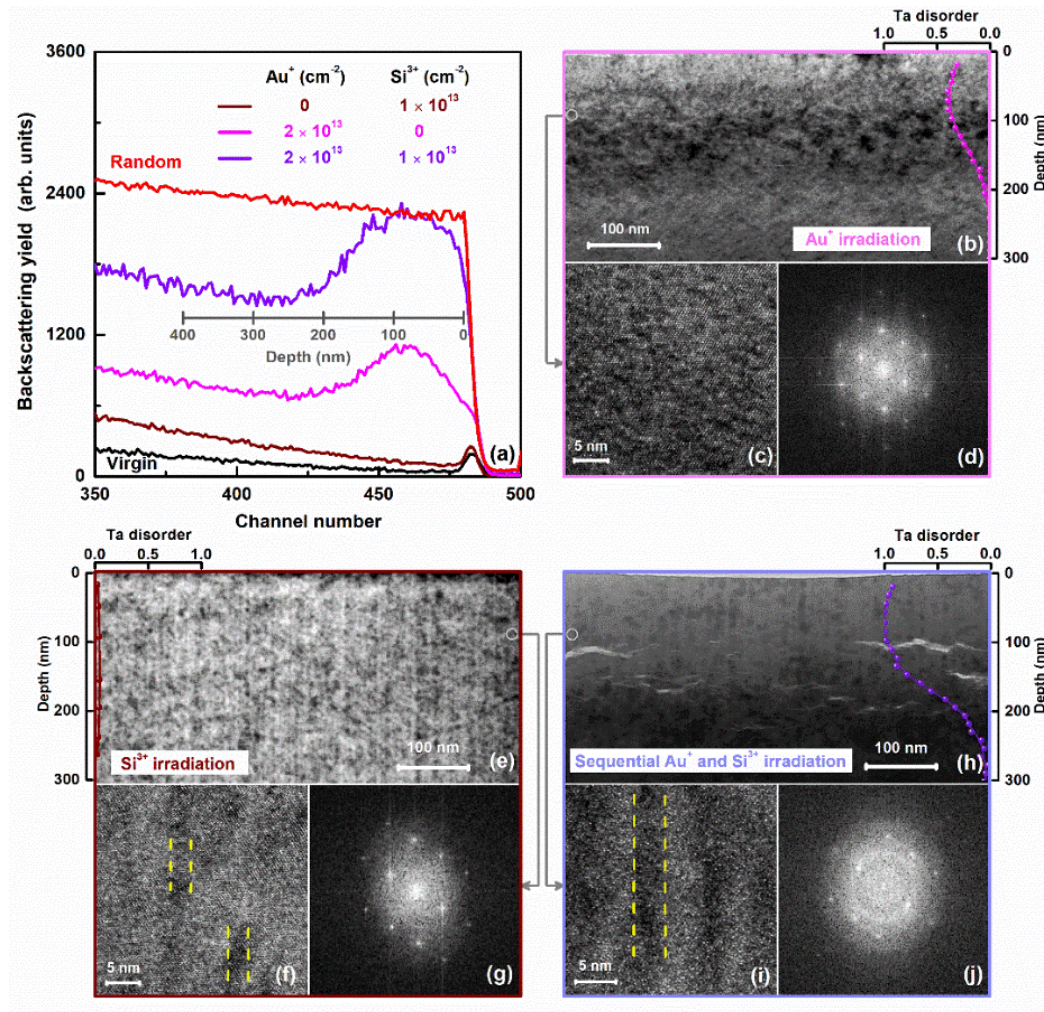


Figure 4. (a) RBS/channeling spectra of pristine and predamaged LiTaO_3 samples under subsequent 0.71 MeV/u Si^{3+} irradiation with an E_{ele} of 6.2 keV/nm. Cross-sectional TEM images and corresponding FFT patterns of LiTaO_3 under (b–d) Au^+ irradiation, (e–g) Si^{3+} irradiation, and (h–j) sequential Au^+ and Si^{3+} irradiation, respectively.

To understand the damage response of Au^+ -irradiated (E_{nuc} -damaged) LiTaO_3 upon subsequent Si^{3+} irradiation with intense electronic energy loss, the corresponding energy deposited to atoms and the consequent atomic temperature evolutions were further calculated using the iTS model [23–25]. In contrast to pristine LiTaO_3 , the lattice defects and defect clusters existing in the Au^+ -irradiated sample scatter electrons and phonons, decreasing thermal conductivities (K_e and K_a) of the electron and lattice subsystems and the electron-phonon mean free path λ , thus leading to an increasing electron-phonon coupling parameter g . Based on the previous research work [5,23,34–36], during the iTS calculation process for Au^+ -irradiated LiTaO_3 crystals, K_e and K_a are assumed to reduce to 75% and

g is assumed to be 40% larger compared to the original values of pristine LiTaO_3 . Comparing Figure 5a,c to Figure 5b,d, under 0.71 MeV/u Si^{3+} irradiation with an E_{ele} of 6.2 keV/nm, the energy deposited to the lattice along the ion path increases from 0.68 to 1.07 eV/atom within $\sim 10^{-13}$ s, and the corresponding atomic temperature increases from 2400 to 3860 K, demonstrating that the introduction of E_{nuc} -induced lattice damage would effectively enhance the response intensity of the E_{ele} -induced thermal spike. In addition, the melting temperature of the crystal decreases owing to the lattice defects and defect clusters embedded in the crystalline system, further promoting the formation of the melting phase and track damage via the subsequent rapid quenching process. Therefore, the experimentally observed additive impacts of the coupled effects of nuclear and electronic energy losses upon track damage production is theoretically demonstrated based on the evolution of energy deposition and atomic temperature in the lattice subsystem.

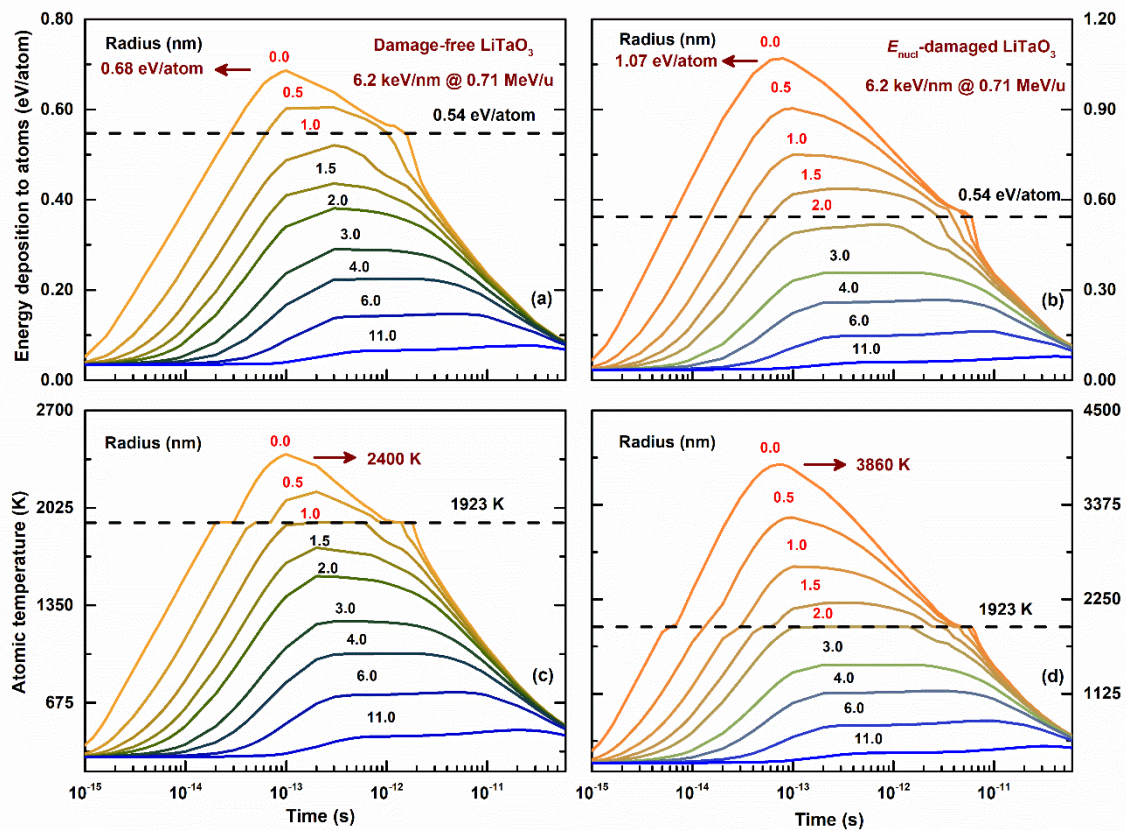


Figure 5. (a,b) Energy deposited to atoms and (c,d) corresponding evolution of atomic temperature at different radii versus time in damage-free and predamaged LiTaO_3 under subsequent electronic loss.

4. Conclusions

In the research field of ion–solid interactions and irradiation effects, a key challenge is to clarify the deposition of irradiation energy to electron and lattice subsystems and further understand the induced damage production of various materials in irradiation environments. In this work, under ion irradiation with 0.71–6.17 MeV/u with electronic energy loss ranging from 6.0 to 13.8 keV/nm, the experimentally observed latent tracks with different damage morphologies were theoretically discussed based on inelastic thermal spike analysis. The effect of electronic energy loss and absorption radius relative to ion velocity on irradiation energy deposition and exchange processes between electron and lattice subsystems and the further induced evolutions of electronic and atomic temperatures are determined, describing the formation of the lattice melting phase and latent track damage in LiTaO_3 crystals. In addition, the lattice defects and defect clusters produced via nuclear energy loss (i) decrease the

thermal conductivity and increase the electron-phonon coupling, leading to the enhanced thermal spike response under electronic energy loss and (ii) accordingly reduce the melting temperature of the crystalline system, similarly to [34,36–38], demonstrating the coupled mechanism by which electronic and nuclear energy losses promote track damage.

Author Contributions: P.L. and X.W. conceived and designed the experiments; X.H., Y.L., and Q.H. performed the irradiations, RBS/channeling, and TEM measurements; X.H. carried out the iTS-model calculations; M.L.C., E.Z., and P.L. performed the data analysis; P.L. and M.L.C. wrote the paper in consultation with all authors. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation of China (grant numbers 11875038 and 11775135), the Young Scholars Program of Shandong University and the Youth Innovation Promotion Association CAS (grant number 2019262).

Acknowledgments: P.L. acknowledges the support from the National Laboratory of Heavy Ion Accelerator in Lanzhou, and the State Key Laboratory of Nuclear Physics and Technology, Peking University; E.Z. was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

Conflicts of Interest: The authors declare no conflict interest.

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