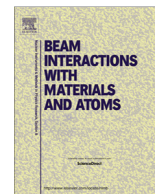




Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Nuclear collision induced lattice swelling and refractive-index modification in ion-irradiated yttrium orthoaluminate crystal

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ARTICLE INFO

Article history:

Received 8 December 2016
 Received in revised form 21 March 2017
 Accepted 3 April 2017
 Available online xxx

Keywords:

Ion irradiation
 Nuclear collision
 Lattice swelling
 Refractive-index modification

ABSTRACT

This work reports the study of lattice damage behavior in yttrium orthoaluminate (YAlO₃) crystal irradiated with medium-energy (6.0 MeV) and relatively high-energy (20.0 MeV) Si ions through complementary characterization techniques including Rutherford backscattering/channeling spectroscopy, transmission electron microscopy and X-ray diffraction. The results clearly demonstrate that under Si-ion irradiation over the energy range from a few MeV up to tens of MeV, the nuclear energy loss (elastic collisions between injected ions and target atoms) along ion trajectory would play a dominant role in lattice damage and swelling, which leads to the decrease of refractive index in the nuclear energy deposition region and the waveguide formation in YAlO₃ crystal. By contrast, the electronic energy loss (ionization and electronic excitation) over the corresponding ion energy range would not produce obvious lattice damage, and therefore could not significantly modify the refractive index in YAlO₃ crystal. Utilizing optical-coupling measurements and iWKB-procedure simulation, the modified refractive-index profile in ion irradiation region has been reconstructed, and the obtained corresponding relationship between the refractive-index profile and SRIM-simulated dpa profile further confirms the nuclear-energy-loss induced lattice swelling and refractive-index decrease behaviors in ion-irradiated YAlO₃ crystal, consisting with the microstructure characterization results.

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1. Introduction

During ion irradiation process, irradiating ion with relatively-low energy will mainly interact with target nuclei and lose energy via nuclear energy deposition (elastic collision) process, which could create a cascade of atomic collision events, displace atoms from initial sites and therefore produce permanent atomic-scale defects in crystal materials [1]; irradiating ion with relatively-high energy will primarily interact with target electrons and lose energy via electronic energy deposition (ionization and electronic excitation) process, which could induce the temperature rise along ion trajectory, and further produce the track containing partially or completely amorphous volume (lattice damage) in crystal materials [2–5]. Irradiation damage induced by nuclear or electronic energy losses could significantly change the physicochemical properties of crystal materials, and has been widely used to modify the optical properties of crystals and fabricate the micro- and nano-scale devices in integrated optics field [6,7]. Recently, for

some functional crystals, the physical mechanism of refractive-index modification induced by the nuclear or electronic energy losses corresponding to different ion energies has been well understood [8,9]. In this work, yttrium orthoaluminate (YAlO₃) crystal has been irradiated with medium-energy (6.0 MeV) and relatively high-energy (20.0 MeV) medium-mass (Si³⁺) ions to different fluences. Lattice damage production and refractive-index modification in YAlO₃ crystal induced by ion irradiation over the energy range from a few MeV up to tens of MeV have been studied, and the effects of nuclear and electronic energy losses on irradiation damage have been discussed comparatively.

2. Experiment and simulation details

Optically-polished YAlO₃ crystal samples with (100) surface normal zone axis direction and dimensions of 10 × 10 × 0.5 mm³ were irradiated with 6.0 MeV and 20.0 MeV Si³⁺ at 300 K to different fluences using 2 × 1.7 MV and 2 × 6 MV tandem accelerators within the State Key Laboratory of Nuclear Physics and Technology at Peking University, respectively. The specific irradiation conditions of sample 1 (S1), sample 2 (S2), sample 3 (S3) and sample 4

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Table 1
SRIM-simulated electronic energy loss, nuclear energy loss and dpa values, and RBS/channeling-measured disorder level corresponding to different irradiation conditions.

Sample	Si ³⁺ energy (MeV)	Si ³⁺ fluence (cm ⁻²)	Damage peak region dpa	Near surface region			
				Nuclear energy loss (keV/nm)	Electronic energy loss (keV/nm)	dpa	Disorder level
S1	20.0	1.0×10^{13}	0.003	0.007	6.6	0.0001	0
S2	6.0	4.4×10^{14}	0.15	0.04	5.2	0.011	0.06 ± 0.01
S3	6.0	6.6×10^{14}	0.22	0.04	5.2	0.017	0.08 ± 0.01
S4	20.0	1.0×10^{15}	0.33	0.007	6.6	0.01	0.14 ± 0.02

(S4) are indicated in Table 1. Lattice damage and swelling in YAlO₃ crystal induced by ion irradiation were characterized through Rutherford backscattering/channeling spectroscopy, transmission electron microscopy and X-ray diffraction using 2×1.7 MV tandem accelerator, 200 kV Tecnai G2 F20 transmission electron microscope and Bruker D8 Advance diffractometer, respectively. Dark-mode spectra of ion irradiation region and near-field intensity distribution of guided mode were measured by prism and end-face coupling techniques, and used to discuss the refractive-index modification behavior induced by ion irradiation process. SRIM 2013 code [10], SIMNRA code [11] and inverse Wentzel-Kramers-Brillouin (iWKB) procedure [12,13] were used to determine the nuclear and electronic energy losses along ion trajectory, fit the measured RBS spectrum and reconstruct the refractive-index profile in ion irradiation region, respectively.

3. Results and discussion

3.1. Lattice damage and swelling

The nuclear and electronic energy losses induced by 6.0 MeV and 20.0 MeV Si-ion in YAlO₃ (density: 5.35 g cm^{-3}) have been determined utilizing SRIM 2013 full-cascade simulation code. As indicated in Table 1, the electronic energy loss is dominant (5.2 keV/nm for 6.0 MeV Si³⁺ and 6.6 keV/nm for 20.0 MeV Si³⁺) in the near surface region. The nuclear energy loss would produce lattice defects through the cascades of atomic collision events, which could be characterized by displacement per atom (dpa). For Si³⁺-irradiated S1, S2, S3 and S4, the dpa values are 0.0001,

0.011, 0.017 and 0.01 in the surface region, and 0.003, 0.15, 0.22 and 0.33 in the heavily damaged dpa-peak region, respectively.

Utilizing 2.0 MeV He⁺ beam, the measured RBS/channeling spectra of YAlO₃ samples are shown in Fig. 1, and SIMNRA-fitting curve is also indicated. The disorder levels on Y sublattice at the surface of S1, S2, S3 and S4 have been determined through a classical approximate expression [14], which are 0, 0.06 ± 0.01 , 0.08 ± 0.01 and 0.14 ± 0.02 , respectively, and have been summarized in Table 1. The results indicate that in the near surface region, the dominant electronic energy loss would not produce obvious irradiation damage, and the measured lattice damage should be attributed to the nuclear collision process. Compared to S2 and S3, the surface region in S4 has lower dpa, and the measured relatively-high disorder level would be ascribed to the measurement error (slight inaccuracy of channel direction), which could be supported by the change of refractive index at the sample surface (Fig. 4).

TEM observations have been performed on the cross section of S4, which has relatively high disorder level in the damage peak region owing to the highest ion fluence. Compared to the un-irradiated region (Fig. 2(a)), high resolution TEM image and electron diffraction pattern taken from the surface region (Fig. 2(b)) indicate that the near-surface region still remains relatively complete crystal structure, and the dominant electronic energy loss would not produce obvious lattice damage. Fig. 2(c)–(e) show the TEM images taken from the heavily damaged dpa-peak region under different magnifications, confirming the highly-disordered and amorphous domains. As shown in the electron diffraction pattern (Fig. 2(f)) taken from the dpa-peak region, the disappearance and deformation of diffraction spots, and the appearance of ring

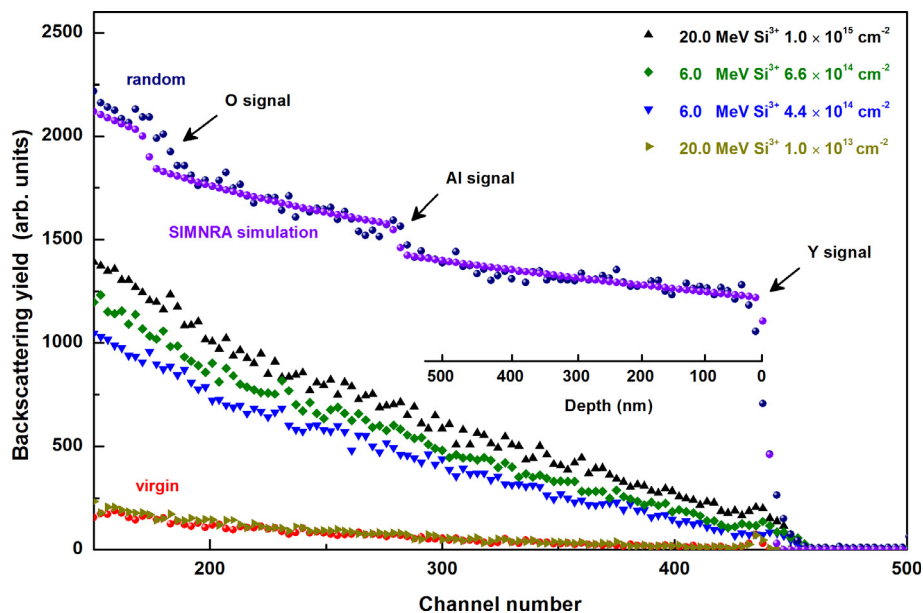


Fig. 1. RBS/channeling spectra of YAlO₃ crystal samples corresponding to different Si³⁺-irradiation conditions.

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