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Ion beam damage assessment and waveguide formation induced by energetic Si-ion irradiation in lanthanum aluminate crystal

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ABSTRACT

Lanthanum aluminate ($LaAlO_3$) crystal has emerged as one of the most valuable functional-materials, and its physical, electronic and optical properties strongly depend on the crystal structure, which can be easily altered in an irradiation environment and therefore affect the performance of $LaAlO_3$-based devices. On the other hand, the preparation of $LaAlO_3$ waveguide is also a scientific challenge for its potential application prospects in optoelectronics field. In this work, the damage evolution behavior of $LaAlO_3$ crystal under Si-ion irradiation has been discussed in detail utilizing complementary characterization techniques, and then, single-mode waveguide of $LaAlO_3$ crystal in the visible band can be obtained based on ion-irradiation-induced lattice damage behavior. Waveguide optical-coupling techniques are used to show its competitive features. Thus, novel optical waveguides with optimized features in $LaAlO_3$ crystals can be tailored by a proper selection of ion mass, energy and fluence using the modification of the target material during ion irradiation process.

1. Introduction

ABO_3 perovskite-structured oxides have attracted considerable attention in recent years owing to their various chemical compounds and crystal structures, and therefore induced diverse physicochemical properties [1–3]. Lanthanum aluminate ($LaAlO_3$) crystal, as one of the representative ABO_3 crystals, has emerged as one of the most scientifically and technologically valuable functional-materials, and in multitudinous fields, it has attracted plenty of fundamental researches, such as two-dimensional electron gas (2-DEG) [4–6], high-temperature superconductivity (HTS) [7–9] and giant magnetoresistance (GMR) [10,11], and also many promising applications, such as gate dielectric layer [12], laser host [13] and microwave resonator [14]. The physical, electronic and optical properties of $LaAlO_3$ strongly depend on its crystal structure, which can be easily altered in an irradiation environment. Therefore, the damage behavior of $LaAlO_3$ crystal could significantly affect the performance of $LaAlO_3$-based integrated-optoelectronic devices [15,16]. On the other hand, considering the potential application prospects of $LaAlO_3$ crystal in optoelectronics field [17–19], the fabrication of $LaAlO_3$ waveguide is a scientific challenge in order to confine the light propagation in small dimensions and further improve the linear and nonlinear optical performance of the device.

In this work, focusing on the above-mentioned two points, firstly, the damage evolution behavior of $LaAlO_3$ crystal under energetic Si-ion irradiation has been discussed in detail utilizing complementary irradiation-damage characterization techniques. Then, based on the dispersion relation of optical guided mode, a waveguide of ion-irradiated $LaAlO_3$ crystal has been prepared. Its properties can be properly tailored through the fine selection of ion mass, energy and fluence [20]. An increase in the ion energy and irradiation fluence will allow an increase of the waveguide thickness and a more abrupt change in the refractive index jump (Δn) of the optical barrier. The related optical properties of the $LaAlO_3$ waveguide using several waveguide coupling techniques are characterized and presented.

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2. Experiment and simulation

2.1. Experiment details

2.1.1. $LaAlO_3$ crystal

$LaAlO_3$ crystal exhibits well-known temperature-induced phase transition behavior. When $LaAlO_3$ is cooled below T_C (813 K, given by calorimetry measurement), the crystal structure will transform from cubic (*PM*-3*M*, space group No. 221) to rhombohedral (*R*3*C*, space group No.161) symmetry due to the rotation of AlO_6 octahedra [21–23]. The rhombohedral structure of $LaAlO_3$ at 300 K is commonly described in hexagonal system with $a = 5.36$ Å and $c = 13.11$ Å, and then, the $(012)_{hex}$ plane (called *R*-plane in rhombohedral system) defines a pseudocubic symmetry with a lattice constant of $a = 3.79$ Å and a lattice angle of 90.087° [24,25]. The $(012)_{hex}$ (hexagonal index) surface normal zone axis direction of $LaAlO_3$ crystal corresponds to $(001)_{pc}$ in pseudo-cubic notation (the subscript *pc* is used to differentiate between the pseudo-cubic *pc* and cubic *c* symmetry) [26,27]. In this work, $LaAlO_3$ single crystal was bought commercially from Shanghai Daheng Optics and Fine Mechanics Co., Ltd, [28], which was grown along c-axis through a Czochralski process utilizing La_2O_3 (4N) and Al_2O_3 (4N) as starting materials [29]. $LaAlO_3$ wafer ($\Phi = 50$ mm × 0.5 mm) with $(012)_{hex}$ surface normal zone axis direction was cut into samples with dimensions of 10 × 10 × 0.5 mm^3 utilizing a diamond saw.

2.1.2. Si-ion irradiation

Si-ion irradiation process and subsequent Rutherford Backscattering Spectroscopy in channeling configuration (RBS/C) analysis were carried out utilizing 2 × 1.7 MV tandem accelerator within State Key Laboratory of Nuclear Physics and Technology, Peking University. During Si-ion irradiation process, ion beam was rastered over the sample surface at 7° off surface normal direction with fixed horizontal and vertical scan frequencies to avoid channeling effect and ensure uniform irradiation. The optically polished 10 × 10 mm^2 surfaces of three $LaAlO_3$ samples were subjected to Si-ion irradiation at 300 K varying ion energies and fluences. As shown in Table 1, sample 1 (S1), sample 2 (S2) and sample 3 (S3) correspond to the specific irradiation conditions of 1.0 MeV Si^+ with beam flux of 6.5×10^{10} cm^{-2} S^{-1} to the fluence of 3.7×10^{14} cm^{-2}, 6.0 MeV Si^{3+} with beam flux of 5.0×10^{10} cm^{-2} S^{-1} to the fluence of 4.4×10^{14} cm^{-2} and 6.0 MeV Si^{3+} with beam flux of 5.0×10^{10} cm^{-2} S^{-1} to the fluence of 6.6×10^{14} cm^{-2}, respectively.

2.1.3. Irradiation-induced damage characterization

The damage behavior of $LaAlO_3$ sample 1 (S1) induced by 1.0 MeV Si^+ irradiation was characterized using complementary techniques of Rutherford backscattering/channeling (RBS/C) spectroscopy, X-ray diffraction (XRD), high-resolution X-ray diffraction (HRXRD), Raman spectroscopy and nano-indentation test. In RBS/C analysis, 2.0 MeV He^+ beam extracted from the accelerator was used to assess the irradiation-induced damage with probe size of 2 × 2 mm^2. A Si detector located at a scattering angle of 160° relative to the incoming beam was used to collect the signal of backscattered He^+ yield from pristine and irradiated $LaAlO_3$ sample. Through rotating sample holder, the main axial channeling orientation and He beam can be aligned, which corresponds to the minimum He^+ yield. The irradiation-induced damage profile in depth in $LaAlO_3$ was determined utilizing an iterative procedure to analyze RBS/C spectra, which is described in more detail below. XRD experiments were performed using a Bruker D8 Advance diffractometer equipped with copper anticathode, and the diffraction patterns were recorded between 15° and 85° in 2θ scale with a step size of 0.02° and a counting time of 0.15 s per step. HRXRD measurements were performed on a Bruker AXS HRXRD D5005 system, which used a Cu-$K_{\alpha 1}$ anticathode with a four-crystal Ge (022) monochromator so as to provide a parallel and monochromatic X-ray beam ($\lambda = 1.54056$ Å). The ω scan (rocking curve) and ω-2θ scan were recorded in the vicinity of $(024)_{hex}$ Bragg reflection with a step size of 0.002°. Raman spectra of $LaAlO_3$ samples at 300 K were acquired through a confocal Raman system (Bruker SENTERRA dispersive Raman microscope) operating at the excitation wavelengths of 532 nm and 785 nm, respectively. The excitation beam from the light source was focused into a spot with 1-μm-diameter on $LaAlO_3$ sample surface, and then the backscattered Raman signals were collected for analysis. After the above-mentioned microstructure characterizations, nano-indentation test was performed utilizing an Agilent Technologies G200 Nano Indenter. A Berkovich diamond indenter tip with a radius of 20 nm was used to perform the Continuous stiffness measurement (CSM) on sample surface, and the profiles of hardness and elastic (Young's) modulus as a function of depth were further obtained. The modifications of surface topography and chemical composition induced by Si-ion irradiation have been studied through atomic force microscopy (AFM), scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS), which were performed on NT-MDT model BL222 RNTE, HITACHI SU8010 and ESCALAB 250, respectively.

2.1.4. Optical-waveguide performance characterization

The optical-waveguide properties of $LaAlO_3$ samples (S1, S2 and S3) irradiated with energetic Si ions were characterized using prism coupling and end-facing coupling techniques [30]. The dark-mode spectra of irradiated samples, gained through m-line technique via a prism coupler (Metricon Model 2010), were measured to study the waveguide performance at wavelengths of visible (633 nm, He-Ne laser) and near-infrared telecommunication band (1539 nm, diode laser). In prism coupling measurements, a lack of reflected light would result in a dip of the dark mode spectrum when laser beam is coupled into waveguide region, indicating the supported waveguide mode and its corresponding effective refractive index. In end-facing coupling measurement, light from a 633 nm He-Ne laser is coupled into the polished end-face via a 25 × microscope objective lens, and then the light coupled out from the opposite polished end face is re-collimated by a 25 × objective lens and imaged onto a charge-coupled device (CCD), which shows the optical near-field intensity distribution of waveguide mode.

Table 1
Irradiation conditions of $LaAlO_3$ samples.

Sample	Irradiated ions	Ion energy (MeV)	Ion fluence (× 10^{14} cm^{-2})	Depth of damage peak (μm)	dpa at damage peak	Characterization
S0	virgin					
S1	Si^+	1.0	3.7	0.54	0.26	irradiation damage assessment
S2	Si^{3+}	6.0	4.4	1.80	0.26	optical waveguide properties
S3	Si^{3+}	6.0	6.6	1.80	0.40	optical waveguide properties