



Investigation of irradiation effects induced by 6.0 MeV O-ions in SrWO₄ crystals



Chuan-Lei Jia^{a,*}, Zhi-Ning Wei^a, Yao Lu^b

^a Department of Physics, China University of Mining and Technology, Xuzhou 221116, Jiangsu, People's Republic of China

^b Advanced Analysis and Computation Center, China University of Mining and Technology, Xuzhou 221116, Jiangsu, People's Republic of China

ARTICLE INFO

Article history:

Received 20 April 2014

Received in revised form

24 June 2014

Accepted 6 July 2014

Available online 19 July 2014

Keywords:

Laser materials

Ion radiation effects

Optical constants

ABSTRACT

Single crystals of z-cut SrWO₄ are irradiated at room temperature with 6.0 MeV O ions at a fluence of 1.6×10^{14} ions/cm². The effects of irradiation on the structural properties are studied by a variety of techniques, including X-ray diffraction (XRD), Raman spectroscopy and Rutherford backscattering spectrometry in channeling geometry (RBS/C) along the (001) axial direction. The optical properties are investigated by prism coupling technique with a wavelength of 633 nm; the results indicate that both the ordinary (n_o) and extraordinary indices (n_e) of SrWO₄ crystals are raised together after implantation.

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1. Introduction

Pure and rare-earth (RE) doped tungstate crystals, as matrices for laser-active elements with nonlinear self-conversion of radiation to a new spectral range, have received a great deal of interest in the field of solid-state stimulated Raman scattering (SRS) laser system in recent years [1–3]. Among those, strontium tungstate (SrWO₄) is the most promising material in SRS lasers and various technological applications due to the remarkable properties, e.g. high Raman gain coefficient and narrow Raman shifted lines [4–7]. SrWO₄ belongs to the tetragonal scheelite-type structure with space group *I*4₁/a (88), and the unit cell parameters are $a=b=0.5417$ nm and $c=1.1951$ nm. In the scheelite structure, the W⁶⁺ ions are surrounded by four O²⁻ anions in a tetrahedral configuration WO₄ and the Sr²⁺ ions occupy a distorted dodecahedral position with an eightfold O²⁻ coordination by forming SrO₈ dodecahedra.

The miniaturization of lasers into monolithic devices has been a subject of considerable interest from the point of view of both scientific investigations and technological applications. Optical waveguides act as the gemstone of integrated optics and photonics devices, and the applications rang from waveguide laser, to modulator and amplifier *etc.* [8–10]. Such compact geometry allows for the intense optical signal with respect to the bulk materials owing to the better light confinement. Up to now ion beam technique has been demonstrated to be an effective way to produce waveguide in most optical materials for its features of controllability and

reproducibility [11,12]. More recently, moderate swift ions irradiation has attracted much attention to produce waveguides in a variety of materials because it may introduce larger refractive index changes for the waveguide structure in comparison to the light ion implantation [13–15]. In addition, exposure to ion bombardment is also a consideration in the performance of practical application of optical and photonic devices. However no results were reported for irradiation effect on SrWO₄ crystals yet. Thus it is necessary to achieve a clear and specific understanding of the optical and structural response of SrWO₄ to ion irradiation. In this work, the energetic O ions implantation in SrWO₄ crystals was carried out at the 1.7 MV tandem accelerator of Peking University. The effects of irradiation on the structural and optical properties are discussed in detail.

2. Experimental details

The z-cut SrWO₄ crystals are optically polished and cleaned; the refractive indices are measured at wavelength of 633 nm with transverse electric (TE) polarization and transverse magnetic (TM) polarization. All specimens are implanted with 6.0 MeV O ions at a fluence of 1.6×10^{14} ions/cm². During the implantation, all samples are tilted at 7° from the z axis in order to minimize the channeling effect and the beam is raster scanned over the sample holder to generate uniform doses. Both the XRD patterns and Raman spectrum are collected to analyze the crystalline structures of the implanted samples; the RBS/C measurement is performed to investigate the damage properties by irradiation. The waveguide properties are also investigated by *m*-lines arrangement using Model 2010 Prism Coupler (Metricon, USA).

* Corresponding author.

E-mail address: jjchl@cumt.edu.cn (C.-L. Jia).

3. Results and discussion

XRD is powerful nondestructive technique for characterizing crystalline materials, which has been intensively performed for identifying crystalline phase, detecting crystallinity, determining composition, measuring residual stress in thin films and bulk materials. It is also an effective method to supply information about deformation of a crystalline sample. Usually, deformation of a crystal lattice will result in a change in the interatomic distances; in addition, the variation of the crystallite will influence the diffraction peaks, leading peak broadening and shifts. The virgin and as-implanted SrWO_4 crystals are structurally characterized by X-ray diffraction (XRD) using $\text{Cu-K}\alpha$ radiation ($\lambda=0.15406\text{ nm}$). The diffraction patterns are recorded in a 2θ range of 20° – 70° . Fig. 1(a) and (b) shows the XRD $\theta-2\theta$ scans patterns of virgin and as-implanted samples, respectively. For the virgin case, a strong SrWO_4 (004) diffraction peak is detected at 29.93° and (008) diffraction peaks at 62.18° , which result is in good accordance with the standard XRD diffraction card of SrWO_4 . The corresponding full width at half maximum (FWHM) of both peaks are around 0.165° , which small value indicates the good crystallinity of SrWO_4 single crystal. After implantation, the SrWO_4 (004) diffraction peak shifts from 29.93° for virgin crystal to 29.82° with the FWHM of 0.18, and the (008) diffraction peak shifts from 62.18° to 61.91° with the FWHM of 0.26, respectively. It is clearly that the (008) diffraction peak intensity degrades after implantation. As is known, the intensity of a given diffraction peak is related to a set of factors, such as the degree of crystallinity, surface nucleation, preferred orientation and residual strain, etc. Normally, much larger scale will be covered during the high-angle diffraction than the low-angle diffraction by performing XRD measurement. In Fig. 1, the (008) peak produced by high-angle diffraction shows more information about the crystallites in a larger scale than that from low-order (004) peak. By comparison between Fig. 1(a) and (b), the effects of irradiation on the crystallographic structure could be found. Clearly, there are only some tiny changes on (004) peak produced by low-angle diffraction, which shows that the crystallites have been well retained in the top surface region after ion irradiation. Differently, in case of high-angle diffraction, the deviation in (008) diffraction peaks are visible between the irradiated sample and the pure one, which reflects the local variation in a sample. It can reasonably deduce that the shift is from the deformation of crystallites in the heavy damaged region at the end of the ion irradiation.

During irradiation process, damages can be introduced into the crystals, including point and extended defects, vacancy, lattice distortion, and so on. Raman spectroscopy has been demonstrated

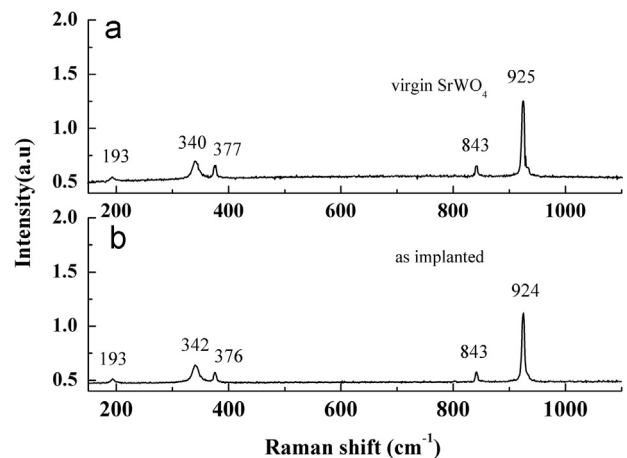


Fig. 2. Raman spectra obtained from (a) virgin crystal and (b) as-implanted sample.

to be an effective way for probing energetic ion damage and material microstructure changes [16]; it provides a direct approach for sampling over the set of crystal vibrational normal modes which are sensitive to the crystallinity. In this work, a wavelength of 532 nm at 5.0 mw power is used as the excitation source, along with a computer controlled three-dimensional stage. Raman spectroscopy is performed on the specimens with the incident light parallel to the z-axis; in this case the selection rules allow backscattering of the multiple phonon modes. Fig. 2 show the Raman spectra of (a) virgin and (b) as-implanted samples recorded at room temperature in the frequency ranging from 150 to 1100 cm^{-1} with labeled sharp peaks. It is known that SrWO_4 presents good nonlinear properties for SRS, and the most intense lines corresponding to the shift produced by the coupling with the totally symmetric vibration Ag of the $(\text{WO}_4)^{2-}$ groups. The observation of Raman peaks mainly arises from the stretching and bending vibration of the metal-oxygen bonds with the anionic groups. In Fig. 2(a), considering the report in Ref. [17], the vibrational mode at 925 cm^{-1} may be attributed to W–O symmetric stretching, peak 843 cm^{-1} from W–O anti-symmetric stretching, peak 377 cm^{-1} from O–W–O anti-symmetric bending, peak 340 cm^{-1} from O–W–O symmetric bending, and peak 193 cm^{-1} from Sr–O stretching, respectively. Comparison of the Raman spectra made on the virgin and irradiated samples, the results reveal that some relative positions of Raman modes have small shifts, which may be caused by distortions on the (O–W–O)/(O–Sr–O) bonds. In general, the penetration depth is typically ranging from 1–5 microns by Raman spectroscopy at confocal mode. In Fig. 2, the Raman results indicate that the effective depth for Raman scattering inside SrWO_4 crystal should be less than the range of irradiation by 6.0 MeV O ions in which the heavy damage peak is produced in depth approximately of $3.2\text{ }\mu\text{m}$ simulated by using program SRIM code, since the observed features are very similar between the implanted sample and the pure one. In addition to this, from the figure, it can be noted that there is no significant change in crystallite microstructure in the near-surface ion-implanted region.

Despite of the unique advantages of ion implantation, there are still some undesired effects, consisting of defects, interstitials, vacancies and lattice disorders by irradiation. Damage due to energetic ion irradiation can be analyzed using well-developed RBS/C spectrometry, typically employing H ions or He ions. This method has been extensively and successfully used for material characterization of radiation damage for its features such as, nondestructiveness, high depth resolution and good sensitivity for heavy elements. In order to evaluate how implantation induced considerable damage into the surface layers, RBS/C experiments

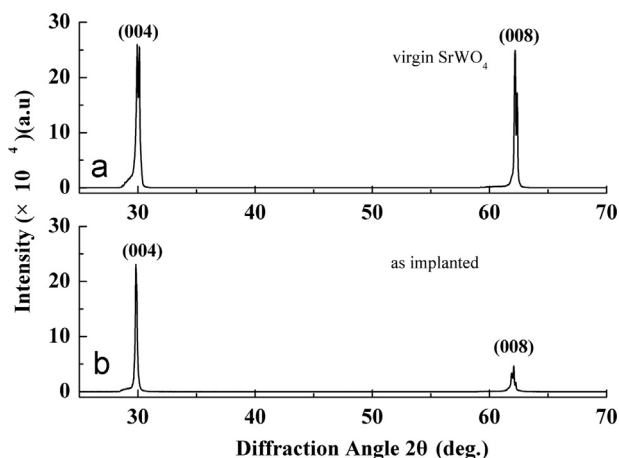


Fig. 1. XRD patterns: (a) virgin crystal and (b) as-implanted sample.

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