As$_2$S$_3$/Sr(Ti$_{0.7}$Co$_{0.3}$)O$_3$ and As$_2$S$_3$/Sr(Ti$_{0.6}$Fe$_{0.4}$)O$_3$ strip-loaded waveguides for integrated magneto-optical isolator applications.

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ABSTRACT
Sr(Ti$_{0.6}$Fe$_{0.4}$)O$_3$ (STF) and Sr(Ti$_{0.7}$Co$_{0.3}$)O$_3$ (STC) room-temperature ferromagnetic oxides were grown epitaxially on LaAlO$_3$(001), (LaSr)(AlTa)O$_3$ (001) and Si (001) substrates. Both materials were demonstrated to be magneto-optically active, and more optically transparent at 1550nm wavelength compared with other non-garnet ferromagnetic materials. As$_2$S$_3$/STF and As$_2$S$_3$/STC strip-loaded waveguides were fabricated on epitaxial STF or STC films grown on LSAT (001) substrates using thermal evaporation and lift-off processing. The absorption of STF at 1550 nm was measured by ellipsometry and by optical transmission spectrum measurements of the As$_2$S$_3$/STF waveguides, which gave similar results. A novel design for a Non-Reciprocal Phase Shift (NRPS) strip-loaded waveguide using chalcogenide glass (ChG) as the guiding layer is proposed. The NRPS and figure of merit of these waveguides are simulated. The ChG strip-loaded waveguide structure shows advantages both in fabrication and device performance according to the simulation results.

Our study suggests the possibility of magneto-optical magneto-optical isolators monolithically integrated on a silicon platform.

KEY WORDS: Integrated magneto-optical isolator, Strip-loaded waveguides, magnetic oxides, chalcogenide glass, silicon integration

1. INTRODUCTION
Magneto-optical (MO) isolators are widely used in optical systems to increase the lifetime and stability of lasers. Conventional discrete optical isolators for communication wavelengths use the non-reciprocal magneto-optical Faraday effect. In these devices, bulk single crystals, usually bismuth doped yttrium iron garnet (Bi:YIG), are used as the magneto-optically active medium to provide non-reciprocal mode conversion of the incident laser beam for optical isolation. In order to achieve smaller device footprint, higher mechanical stability and lower fabrication cost, integration of MO isolators on a semiconductor substrate such as Si or III-V platforms is desired. However, garnet materials encounter integration difficulties due to lattice mismatch, thermal mismatch and a high thermal budget for fabrication. So far, there are no semiconductor integrated optical isolators available for near infrared communication wavelengths.

Solutions to this problem have been explored for many years and by various approaches such as optimizing the
fabrication process for garnet films on semiconductor substrates and exploring new materials which are more compatible with semiconductor substrates.\textsuperscript{5,8} The figure of merit (FOM), defined as Faraday rotation per unit length divided by optical absorption per unit length is a criterion for the usefulness of an MO material. MO materials other than garnets usually exhibit significantly inferior FOMs. For instance, at 1550nm wavelength YIG has a FOM larger than 100 deg/dB while $\gamma$-Fe$_2$O$_3$ has a FOM of 0.15 deg/dB.\textsuperscript{6} Other spinels, Co-doped CeO$_2$,\textsuperscript{7} and Co- or Fe-doped SnO$_2$\textsuperscript{8} also have FOMs much smaller than 1 deg/dB. The low FOMs of these materials are mainly due to strong material absorption in the NIR region. On the other hand, the integrated waveguide structure offers more freedom for device design than is present in bulk MO isolators. Devices such as Mach-Zehnder interferometers using the Non-Reciprocal Phase Shift (NRPS) of MO waveguides can achieve optical isolation for TM polarized light.\textsuperscript{11} Therefore, exploring new materials in combination with new device designs is very important for integrated MO isolator fabrication.

In this study, we report the properties of two novel ferromagnetic and silicon-compatible materials Sr(Ti$_{0.6}$Fe$_{0.4}$)O$_3$ (STF) and Sr(Ti$_{0.7}$Co$_{0.3}$)O$_3$ for integrated optical isolator applications. As$_2$S$_3$/STF and As$_2$S$_3$/STC strip-loaded waveguides are fabricated by thermal evaporation and lift-off and the waveguide transmission loss is measured. The NRPS property of the As$_2$S$_3$/STF waveguide is simulated. Based on the experimental and simulation results, the optimum waveguide design is also discussed.

2. MAGNETOOPTICAL FILM DEPOSITION AND WAVEGUIDE FABRICATION

2.1 MO film deposition

SrTi$_{0.6}$Fe$_{0.4}$O$_3$ (STF) and SrTi$_{0.7}$Co$_{0.3}$O$_3$ (STC) films were deposited on (La$_{0.29}$Sr$_{0.71}$)(Al$_{0.65}$Ta$_{0.35}$)O$_3$ (LSAT) (001), LaAlO$_3$ (LAO) (001), or Si (001) substrates by pulsed laser deposition (PLD) with a Coherent COMPexPro 205 KrF (248 nm) excimer laser. The targets for PLD were fabricated by standard ceramic fabrication processing. SrCO$_3$(99.99%), TiO$_2$(99.99%) and Fe$_2$O$_3$(hematite,99.945%)/CoCO$_3$(99.9%) powders were stoichiometrically mixed, ball milled for 24 hours and calcinated at 1200°C for 5 hours to form perovskite phases. The calcinated powders were pressed into 1 inch diameter pellets and sintered at 1300°C for 5 hours to form the targets. The film deposition was carried out under a chamber pressure of 2.0×10$^{-6}$ to 3.0×10$^{-6}$ torr for STF and STC film respectively. The laser energy, fluence and repetition rate were set at 550 mJ/pulse, 2.5 J/cm$^2$ and 10Hz respectively. During the deposition, the LAO and LSAT substrates were kept at 700°C and the Si substrate was kept at 800°C to allow epitaxial growth.

2.2 Waveguide fabrication

To eliminate scattering loss from substrate twin defects which are observed in LAO substrates, films grown on twin-free LSAT substrates were chosen for waveguide fabrication. A photoresist pattern with 2 $\mu$m to 8 $\mu$m waveguide structures were firstly formed on the MO film samples by photolithography. As$_2$S$_3$ glass films were then thermally evaporated on the samples which were kept at room temperature. A lift-off process was then carried out to form a strip-loaded waveguide structure with As$_2$S$_3$ channels on STF/STC films. After this step, a second layer of As$_2$S$_3$ film was deposited on the samples to form a ridge waveguide structure. SU-8 photoresist (n=1.58, MicroChem) was spin-coated onto the strip-loaded waveguides and exposed under room light for 10 min to form a top cladding layer with 2.5 $\mu$m thickness. Details about the glass deposition and lift-off process may be found in Refs. 9 and 10. The LSAT substrates were then
cleaved perpendicular to the waveguide direction to form facets for transmission spectrum characterization.

3. MAGNETOOPTICAL FILM AND WAVEGUIDE CHARACTERIZATION

Phase identification of the MO films was carried out by one dimensional X-ray diffraction (1DXRD, Rigaku RU300). To evaluate the film texture and epitaxial relationship with the substrate, two-dimensional X-ray diffraction (2DXRD, Bruker D8 with General Area Detector Diffraction System (GADDS)) and cross-sectional HRTEM characterization were also used. The film roughness (RMS) and magnetic domain structure were characterized by atomic force microscopy (AFM) and magnetic force microscopy (MFM) respectively on a Digital Instruments Nanoscope IIIa Atomic Force Microscope. The magnetic hysteresis of the films were measured by vibrating sample magnetometry (VSM) on an ADE Technologies VSM Model 1660. Room temperature Faraday rotation hysteresis of the films was characterized at 1550nm wavelength on a custom-built apparatus with the laser beam propagating perpendicular to the film normal direction. Optical constants of the films were characterized from 400nm to 1700nm wavelengths for both films on a WVASE32 ellipsometer. General oscillator models were applied to fit the optical constants, and the fitting root mean square error (MSE) was smaller than 1. The validity of the optical measurements was evaluated by measuring the transmission spectrum of both films on a Cary 500i UV-Vis-NIR Dual-Beam Spectrophotometer from 175 nm to 1700 nm wavelength.

The waveguide cross section morphology was characterized on a Zeiss/Leo Gemini 982 SEM. Waveguide transmission measurements were performed on a Newport AutoAlign workstation in combination with a LUNA tunable laser (optical vector analyzer, LUNA Technologies, Inc.). Lens-tip fibers are used to couple light from the laser into and out of the devices. Reproducible coupling is achieved via an automatic alignment system with a spatial resolution of 50 nm. The sample is mounted on a thermostat stage and kept at 25 °C for all measurements.

4. RESULTS AND DISCUSSION

4.1 Structure and properties of STF and STC films

Figure 1 (a) shows the 1D and 2D XRD spectra for STF and STC films grown on LAO (001) substrates. Similar XRD spectra were also observed for films grown on LSAT(001) substrates. Both films show (00k) diffraction peaks of single perovkite phases in the 1DXRD spectra, suggesting a “cube-on-cube” crystal orientation relation between the film and the substrate. The well defined spot-like 2DXRD diffraction patterns for STF(002) and STC(001) peaks indicate that both films were epitaxially grown on the LAO substrates. STF and STC show similar out-of-plane lattice constants of 3.992Å and 3.993Å respectively. These values are larger than those of the LAO (3.78Å) and LSAT (3.87Å) substrates suggesting the films are under in-plane compressive stress. A detailed study of the reciprocal space map of an STC film grown on an LAO substrate results an in-plane constant of 3.914Å (data not shown). Considering that bulk STC and STF samples show cubic symmetry, the films are tetragonalized along the out-of-plane direction due to in-plane biaxial stress. The epitaxial growth of the films can also be seen in their cross-sectional TEM images. Figures 1 (b) and (c) show cross-sectional TEM image of the STF and STC films grown on LAO substrates, and the insets show the Fast Fourier
Transform (FFT) pattern for the STF film and the interface diffraction pattern for the STC film respectively. Both films show coherent interfaces with the substrates and no secondary phases can be observed. The surface RMS roughness of the films was characterized by AFM. By averaging more than three 5\( \mu \)m by 5\( \mu \)m area scans at different regions, the RMS roughness of both films was estimated to be less than 3nm. These results indicate that epitaxial STF and STC films are single phase and structurally uniform.

![1DXRD spectra of epitaxial SrTi\textsubscript{0.6}Fe\textsubscript{0.4}O\textsubscript{3} (STF) and SrTi\textsubscript{0.7}Co\textsubscript{0.3}O\textsubscript{3} (STC) on LaAlO\textsubscript{3} (001) (LAO) substrates. Also shown is the 2DXRD pattern of the STF(001) and STC(002) peaks. (b) Cross-sectional TEM image of a STF film on LAO substrate. The inset shows the FFT pattern. (c) Cross-sectional TEM image of STC film on LAO substrate. The inset shows the electron diffraction pattern at the interface.](image)

To explore the possibility for silicon integration, we also deposited STF film on Si (001) substrates with YSZ (yttrium stabilized zirconia)/CeO\textsubscript{2} double buffer layers. The YSZ(100nm)/CeO\textsubscript{2}(150nm) buffer layers were firstly grown by PLD on Si(001), then the STF film with 220nm thickness was in-situ deposited on the buffer layers. Epitaxial growth of all three layers can be demonstrated by 1D and 2D XRD spectra as shown in figure 2. Both YSZ and CeO\textsubscript{2} layers show “cube-on-cube” orientation with respect to the Si substrate, while the STF film shows a 45° in-plane rotation of the lattice with respect to CeO\textsubscript{2} to allow a better lattice match.\textsuperscript{14} The RMS roughness of this sample is also below 3nm as measured by AFM.

![1DXRD and 2DXRD spectra for an STF film epitaxially grown on a Si(001) substrate using YSZ/CeO\textsubscript{2} buffer layers.](image)

Figure 3 (a) and (b) show the room temperature magnetic hysteresis loops of STF and STC films. Both films are room temperature ferromagnets with an out-of-plane easy axis. The saturation magnetization \( M_s \) for the STF and STC films are

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Proc. of SPIE Vol. 7218  721803-4
The room temperature ferromagnetism observed in these thin films is quite different from the magnetic behavior of STF and STC bulk samples, which are paramagnetic at room temperature. This is because the STF and STC films are fabricated in high vacuum, and the valence state of Fe and Co atoms, oxygen vacancy concentration, and stress in these epitaxial films are different from bulk samples. X-ray photoelectron spectroscopy (XPS) analysis indicates that Fe has Fe\(^{2+}\), Fe\(^{3+}\) and Fe\(^{4+}\) valence states in STF while Co has Co\(^{2+}\), Co\(^{3+}\) and Co\(^{4+}\) in STC. However in bulk STF and STC samples, Fe\(^{3+}\) and Co\(^{3+}\) are the majority ions coexisting with small amount of Fe\(^{4+}\) and Co\(^{4+}\) ions. High vacuum deposition also creates many oxygen vacancies which may be beneficial to ferromagnetic properties. It should be noted that this magnetic behavior of both films were observed in more than 10 samples fabricated under similar conditions. To demonstrate the magnetic structure of the film, we also carried out MFM measurement on the STF sample. Figure 3 (c) shows the MFM image of a 5 \(\mu\)m by 5 \(\mu\)m area for the STF film after out-of-plane AC demagnetization. Clear magnetic domains can be observed in the film, which indicates the intrinsic ferromagnetic behavior from the crystal lattice rather than any secondary phases or clusters.

Figure 4 (a) shows the out-of-plane Faraday rotation hysteresis loops of the STF and STC films. The STF and STC films show saturation Faraday rotations of 780 deg/cm and 350 deg/cm respectively, and saturate below 3000 Oe. Figures 4 (b) and (c) show the refractive index \(n\) and extinction coefficient \(k\) of both films measured by ellipsometry. STF shows a refractive index of 2.2 at 1550nm, while STC has a slightly higher index of 2.34. The extinction coefficients of both films are in the low \(10^{-3}\) range at 1550nm wavelength. The validity of the ellipsometry results is also confirmed by UV-VIS-NIR transmittance spectrum (data not shown here) which shows high transmittance of both films at NIR wavelengths. The figure of merit (Faraday rotation per dB optical absorption) is estimated to be 1.1 deg/dB and 0.66 deg/dB for STF and STC films respectively.
The relatively low optical absorption of both films in the near infrared range can be understood from defect chemistry. Free carrier absorption is believed to be the main absorption mechanism in SrTiO$_3$ films deposited in vacuum or STF/STC films deposited in oxygen atmosphere. However, in the vacuum-deposited films used here, Fe$^{2+}$, Fe$^{3+}$, Co$^{2+}$ and Co$^{3+}$ at Ti$^{4+}$ sites act as acceptors while oxygen vacancies formed during deposition are donors. The defect balance between the acceptors and donors yields a low free carrier concentration in the STF and STC films making them more transparent, although the extinction coefficient of $\sim 10^{-3}$ indicates that significant absorption still exists. This is attributed to optical absorption of octahedrally-coordinated Fe$^{2+}$, Fe$^{4+}$ and Co$^{3+}$ ions, of which cause optical absorption in the near infrared in garnets. Meanwhile, Verwey conduction between multi-valence state transition metal ions can also cause absorption tails in the infrared range.

4.2 Strip-loaded waveguide fabrication and Non-Reciprocal Phase Shift (NRPS) waveguide design

As$_2$S$_3$/STF and As$_2$S$_3$/STC strip-loaded waveguides with waveguide width ranging from 2 µm to 8 µm were fabricated on LSAT substrates. Figure 5 shows the cross-section SEM image of a 4 µm wide As$_2$S$_3$/STF strip-loaded waveguide. The waveguide consists of a LSAT substrate, 100 nm STF film, an As$_2$S$_3$ layer with 500nm thick slab and 400nm thick rib layers, and a 2.5 µm thick SU-8 top-cladding layer. The interface between the As$_2$S$_3$ and STF film maintained its integrity during processing. The As$_2$S$_3$ rib shows slanted edges and round corners due to the lift-off process. The sidewall angle measured from the SEM image is 55°~60°, which is slightly smaller than that of ChG waveguides fabricated on silicon substrates by lift-off processing. As$_2$S$_3$/STC waveguides showed similar morphology. The
transmission loss of As$_2$S$_3$ ridge waveguides, with similar dimensions but without the STF, on LSAT substrates were characterized, and the coupling loss and transmission loss of the As$_2$S$_3$ waveguide were estimated from these measurements. Using the measured total transmission loss of the strip-loaded waveguide and simulated confinement factor of the STF film layer, the optical absorption coefficient of the STF film can be estimated.

![Cross-sectional SEM image of an As$_2$S$_3$/STF strip-loaded waveguide with an SU-8 top-cladding layer fabricated on an LSAT(001) substrate.](image)

Fig. 5 (a) Cross-sectional SEM image of an As$_2$S$_3$/STF strip-loaded waveguide with an SU-8 top-cladding layer fabricated on an LSAT(001) substrate. (b) Transmission spectra of an As$_2$S$_3$ ridge waveguide and an As$_2$S$_3$/STF strip-loaded waveguide with the same dimensions of the As$_2$S$_3$ layer on LSAT substrates. The inset shows the measured mode profile of the As$_2$S$_3$ ridge waveguide.

Optical transmission was observed in the As$_2$S$_3$/STF waveguides at 1550 nm wavelength. The transmission spectra of both waveguides are shown in figure 5(b). The optical absorption coefficient of the STF film at 1550 nm wavelength is estimated to be $(5.0 \pm 1.6) \times 10^2$ cm$^{-1}$. Multi-mode beating effects in As$_2$S$_3$ ridge waveguides on bare LSAT substrates and the consequent uncertainty in insertion loss estimation leads to the measurement error bar. These absorption values are consistent with our ellipsometry measurement results.

The magneto-optical performance, specifically the Non-Reciprocal Phase Shift (NRPS) of the strip-loaded waveguides, is simulated based on ChG/STF strip-loaded waveguides. The NRPS in a magneto-optical waveguide structure shown in the inset of figure 6(a) can be calculated by perturbation theory, which yields an expression for NRPS to be:\(^1^8\)

$$\Delta \beta_{TM} = - \frac{2 \beta_{TM}^{TM}}{\omega \varepsilon_0 N} \iint \frac{K^* M_y}{n^4} H_y \partial_x H_y \, dx \, dy$$

where $\beta_{TM}$ is the propagation constant of a TM mode wave, $\Delta \beta_{TM} = \beta_{forward} - \beta_{backward}$ is the difference between forward and backward propagation constants (NRPS), $\omega$ is the frequency, $\varepsilon_0$ is the vacuum dielectric constant, $N = (\iint E \times H^* + E^* \times H)_z \, dx \, dy$ is the power flow along the z direction, $K^* M_y$ is the off-diagonal component of the dielectric constant matrix under an applied field $M_y$ along the y direction and $H_y$ is the magnetic field component of the electromagnetic wave along the y direction. The integral is over all the magneto-optically active
regions in the waveguide. To calculate the NRPS in our strip-loaded waveguide, the TM mode profile is firstly simulated by FIMMWAVE software using a wave matching method (WMM). From the mode profile the NRPS can be calculated using equation (1). The insertion loss of the waveguide can be also simulated by considering the confinement factor and optical absorption in the STF layer. The results are shown in figure 6(a), (b) and (c). Firstly we note that for a MZ interferometer isolator using NRPS, ± 90° NRPS has to be achieved in its either arm. The result in fig. 6(a) indicates that isolator device within millimeter to low centimeter scales can be fabricated based on most of the proposed WG structures. Secondly, in figure 6(a) and (b) the NRPS and insertion loss of the waveguides both decrease with increasing ChG layer thickness. This can be easily understood because the confinement factor in the magneto-optical film decreases with increasing ChG layer thickness. For the same reason, the NRPS and insertion loss also decrease with increasing refractive index of the ChG layer. However in figure 6(c), the figure of merit (FOM, defined as NRPS divided by insertion loss) shows a maximum value for a ChG layer thickness of 1.2 μm. This is due to the design trade-off between maximizing the mode asymmetry (NRPS) and minimizing the optical confinement factor (absorption loss) in the STF layer. Meanwhile, waveguides with an As2Se3 top layer show the highest FOM in all waveguide dimensions. This result can be interpreted as due to the $\partial_y H_y$ term in equation (1). Higher index contrast between the top and bottom cladding yields higher field gradient and NRPS in the STF layer, therefore the FOM is also enhanced. Experimental characterization of the NRPS in these waveguides is currently under investigation.

Fig. 6 (a) Simulated Non-Reciprocal Phase Shift (NRPS) in ChG/STF strip-loaded waveguides. The inset shows the waveguide dimensions used for mode profile simulation. (b) Simulated waveguide insertion loss of ChG/STF strip-loaded waveguides. (c) Figure of merit (FOM) defined as NRPS divided by insertion loss in ChG/STF strip-loaded waveguides.
From the waveguide characterization and simulation results above, we can conclude that the high index of ChG glass is beneficial to the performance of magneto-optical waveguides, and the lift-off method allows non-composition-specific rapid prototyping and prevents etching-induced waveguide sidewall roughness. These analyses indicate that high index ChG is an ideal material for magneto-optical waveguide fabrication. However the FOM of current strip-loaded waveguides is still too low for optical isolator fabrication. Improvement of the FOM may be achieved by either decreasing the Fe\(^{2+}\), Fe\(^{4+}\) or Co\(^{2+}\) concentration in the STF or STC films to decrease the optical absorption, or doping Ce\(^{3+}\) or Bi\(^{3+}\) into the STF or STC lattice to enhance the magneto-optical properties, as is done in garnets.

5. SUMMARY

Sr(Ti\(_{0.6}\)Fe\(_{0.4}\))O\(_3\) and Sr(Ti\(_{0.7}\)Co\(_{0.3}\))O\(_3\) films were epitaxially grown on LaAlO\(_3\) (001), LSAT (001) and Si (001) substrates by pulsed laser deposition. Both films are single phase with low surface roughness, and are room temperature ferromagnets with out-of-plane magnetization easy axis. The room temperature saturation Faraday rotations for STF and STC films at 1550nm wavelength are 780 deg/cm and 350 deg/cm respectively, while the extinction coefficient coefficients for both films are in the low 10\(^{-3}\) range at this wavelength. As\(_2\)S\(_3\)/STF and As\(_2\)S\(_3\)/STC strip-loaded waveguide were fabricated by thermal evaporation and lift-off process. The absorption coefficient of the STF film is estimated to be \((5.0 \pm 1.6) \times 10^2 \text{ cm}^{-1}\) by measuring the transmission spectrum of the strip-loaded waveguides, which is consistent with our ellipsometry measurement results. The magneto-optical properties of the strip-loaded waveguides were simulated by analyzing their mode profile and calculating the non-reciprocal phase shift. It is found that the best figure of merit can be achieved by control of the waveguide geometry and the refractive index in the chalcogenide glass layer. These results suggest that by incorporating magneto-optical materials into chalcogenide glass-based strip-loaded waveguides, silicon-compatible magneto-optical isolators can be fabricated.

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