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ABSTRACT
Bismuth-substituted yttrium iron garnet (Bi:YIG) films were prepared by using spin coating processes with metal-organic-decomposition-method-based solutions on crystalline silicon (Si) substrates, and their magneto-optic properties at the 1550-nm wavelength region were investigated by performing various thermal treatments. The maximum Verdet constant of the Bi$_1$Y$_2$Fe$_5$O$_{12}$ film on the Si substrate with a middle buffer layer of Bi$_2$Y$_1$Fe$_5$O$_{12}$ was measured to be 1 072 038 ○/T/m at 1550-nm wavelength in the unsaturated linear magnetization region by accounting for the negative Verdet constant of the silicon substrate. The optimum thermal treatment condition was observed at the maximum annealing temperature of 700 ○C and the annealing speed of 3 ○C/min. These spin coating enabled processes may be included to the conventional complementary metal-oxide semiconductor fabrication processes to demonstrate integrated optical waveguide-type isolators on silicon-on-insulator wafers.

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I. INTRODUCTION
A good magneto-optic (MO) material applicable to the complementary metal-oxide semiconductor (CMOS) processes is needed to demonstrate integrated optical isolators (IOIs) on silicon (Si) wafers, which are critical parts in photonic integrated circuits (PICs). Various MO materials based on organic and inorganic materials have been studied by many researchers. The organic MO materials are good for thin-film coating, but they are limited to relatively low MO properties and low thermal budgets which are major obstacles in their applications as integrated optical isolators. The inorganic magneto-optic metal-oxides (MOMOs), such as yttrium iron garnet (YIG), orthoferrites (YFeO$_3$, LaFeO$_3$, and BiFeO$_3$), and nanocrystals (γ-Fe$_2$O$_3$ and CoFe$_2$O$_4$), have been recognized for their high thermal durability and good magneto-optic properties as potential materials in applications to integrated optical isolators.

Recently, metal-substituted YIG crystals are most frequently chosen as the magneto-optic materials for optical isolator applications because of their low optical absorption properties at the near-infrared wavelength region. Bismuth-substituted yttrium iron garnet (Bi:YIG) films grown by the liquid phase epitaxy (LPE) method on gadolinium gallium garnet (GGG) substrates were used to be patterned into direct planar waveguides and to demonstrate the integrated optical waveguide-type isolator (IOWTI) with an external magnetic field applied and with sputter-deposited thin-film magnets. A thin layer of cerium-substituted YIG (Ce:YIG) fabricated by the sputtering method on the GGG substrate was bonded on the top of a waveguide-type Si Mach–Zehnder interferometer (MZI) or of a waveguide-type Si microring interferometer (MRI) as an upper cladding to form the IOWTIs. These approaches to IOWTI implementation using the direct Bi:YIG waveguide or the bonded Ce:YIG cladding layer are not suitable for mass production of PIC chips under the CMOS processes. A monolithic integration of the MO material layer directly on the Si waveguides has been pursued to demonstrate the IOWTI by using pulsed laser deposition (PLD). This PLD method is still a challenging approach to be adapted to the CMOS process for a full wafer scale PIC production. In addition, further improvement is...
needed to reduce the large optical insertion loss caused by this approach.

A solution-based spin coating approach to form the MO film layer can relatively be easy to be adapted to the CMOS process. Recently, the metal-organic decomposition (MOD) method was demonstrated to form the spin-coated MO film layers directly on a substrate. The spin-coated Bi:YIG films on glass substrates were prepared from the MOD solutions, and their MO properties such as Faraday rotation, magnetization, and optical absorption at the visible wavelength region were reported. The MO properties of the Bi:YIG films spin-coated on glass substrates with the MOD solutions at the 1310-nm and 1550-nm wavelengths have also been reported previously. However, the MO properties of the Bi:YIG films spin-coated on Si substrates are very important for their applications to the IOWTI in Si-based PICs.

In this paper, we have prepared spin-coated Bi:YIG films on the Si substrates using the MOD solutions and measured the MO properties of the films by performing thermal treatments at various conditions. The maximum Verdet constant of the films at 1550-nm wavelength was determined at unsaturated linear magnetization conditions. The maximum Verdet constant of the films at 1550-nm wavelength region were reported. However, the MO properties of the Bi:YIG films spin-coated on Si substrates is approximately about 0.25 μm. A reasonable thickness (about 60–80 nm range) of the buffer layers was obtained by repeating only 3 times the above spin coating process and the pre-annealing process. The main annealing process was performed by heating, baking, and cooling the samples using a computer controlled electric furnace for various conditions of heating and cooling speeds (annealing speeds) and baking temperatures. The optimum baking temperature for the crystallization of the MOD films is reported to be around 750 °C.

A spin-coated and thermally treated Bi$_2$:YIG film sample was prepared on the Si substrate and a Bi$_2$:NIGG buffer layer, and its crystal structure and grain sizes were measured with an x-ray diffractometer (XRD) and an atomic force microscope (AFM), respectively, whose measured results are shown in Figs. 1(a) and 1(b), respectively. The measured XRD pattern in Fig. 1(a) shows the strong peaks corresponding to the garnet structure along with weak peaks corresponding to the partially formed orthoferrites, such as BiFeO$_3$ and YFeO$_3$, which are intermediate products of the thermally treated Bi$_2$:YIG films. Based on the measured AFM image in Fig. 1(b), the maximum size of the crystal grains in the Bi$_2$:YIG film is approximately about 0.25 μm.

The thickness of the Bi$_2$:YIG film on the Si substrate was determined directly from the cross-sectional view of the scanning electron microscope (SEM) image of the film sample. The SEM image taken at the cross-sectional edge of the fabricated Bi$_2$:YIG film on the Si substrate is shown in Fig. 1(c). Since the boundaries of the buffer layer were not easily distinguishable from the main Bi$_2$:YIG layer can relatively be easy to be adapted to the CMOS process. Recently, the metal-organic decomposition (MOD) method was demonstrated to form the spin-coated MO film layers directly on a substrate. The spin-coated Bi:YIG films on glass substrates were prepared from the MOD solutions, and their MO properties such as Faraday rotation, magnetization, and optical absorption at the visible wavelength region were reported. The MO properties of the Bi:YIG films spin-coated on glass substrates with the MOD solutions at the 1310-nm and 1550-nm wavelengths have also been reported previously. However, the MO properties of the Bi:YIG films spin-coated on Si substrates are very important for their applications to the IOWTI in Si-based PICs.

In this paper, we have prepared spin-coated Bi:YIG films on the Si substrates using the MOD solutions and measured the MO properties of the films by performing thermal treatments at various conditions. The maximum Verdet constant of the films at 1550-nm wavelength was determined at unsaturated linear magnetization conditions. The maximum Verdet constant of the films at 1550-nm wavelength region were reported. However, the MO properties of the Bi:YIG films spin-coated on Si substrates is approximately about 0.25 μm. A reasonable thickness (about 60–80 nm range) of the buffer layers was obtained by repeating only 3 times the above spin coating process and the pre-annealing process. The main annealing process was performed by heating, baking, and cooling the samples using a computer controlled electric furnace for various conditions of heating and cooling speeds (annealing speeds) and baking temperatures. The optimum baking temperature for the crystallization of the MOD films is reported to be around 750 °C.

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layer and the substrate layer, the thickness of the buffer layer was calculated by measuring the thickness of the Bi₂:YIG film sample of the same material compositions with no buffer layer and with a buffer layer with a desired thickness (see Ref. 15 for details). The measured thicknesses of the films on the Si substrates were in the range of 0.642–1.517 μm depending on the material composition, and their error range was within ±0.040 μm. This thickness error was accounted to estimate the error of the measured Verdet constants of the samples and denoted as an error bar in the plotted data. In this experiment, we have fabricated the main film samples of Bi₁₋₂:YIG and Bi₁:YIG material compositions on the Si substrate with no buffer layer and with one of the Bi₁:YIG, Bi₂:YIG, and Bi₃:NiGG buffer layers.

B. Measurement of the Verdet constants of Bi:YIG films

The Verdet constants of the fabricated Bi:YIG film samples on the Si substrates were measured by using a sensitive Faraday rotation measurement system with a lock-in amplifier (LIA) and an auto-balanced photoreceiver (ABPR) under an alternating magnetic field, as described in Refs. 15 and 16. When the light beam passes through a Bi:YIG film sample, it suffers a Faraday rotation whose polarization rotation angle \( \theta \) is proportional to the product of the Verdet constant \( V \) and thickness \( L \) of the sample and the applied magnetic field \( B \):\(^\text{17}\)

\[
\theta = VBL. \tag{1}
\]

The total Faraday rotation angle per unit magnetic field corresponds to \( V_{\text{total}}L_{\text{total}} \) which is the sum of the Faraday rotation angles caused by the Bi:YIG film sample layer, the buffer layer, and the silicon substrate. It can be expressed as

\[
\frac{\theta}{B} = V_{\text{total}}L_{\text{total}} = V_{\text{Bi:YIG}}L_{\text{Bi:YIG}} + V_{\text{Buffer}}L_{\text{Buffer}} + V_{\text{Subs}}L_{\text{Subs}}. \tag{2}
\]

where \( V_{\text{Bi:YIG}}, L_{\text{Bi:YIG}}; V_{\text{Buffer}}, L_{\text{Buffer}}; \) and \( V_{\text{Subs}}, L_{\text{Subs}} \) indicate the Verdet constants and thicknesses of the Bi:YIG film, the buffer layer, and the substrate, respectively. One technical point that we have to consider is the opposite signs of the Verdet constants of the Si substrate and the YIG film layer. The crystalline silicon has a negative Verdet constant due to its diamagnetic property, while the YIG garnet has a positive Verdet constant due to its ferrimagnetic property.\(^\text{15,16}\) The Faraday rotation angle of the incoming light beam in the sample per unit magnetic field \( \frac{\theta}{B} = V_{\text{total}}L_{\text{total}} \) varies from a negative value for the Si substrate only case to a positive value for the case of a sufficiently thick Bi:YIG film deposited on the Si substrate. With the Faraday rotation measurement setup shown in Fig. 2, the Faraday rotation angle per unit magnetic field \( \frac{\theta}{B} = V_{\text{total}}L_{\text{total}} \) is determined from the measured output voltage \( R_{\text{LIA}} \) of the LIA and the time-averaged output voltage \( V_{\text{SM}} \) measured at the signal monitoring mode of a single photodiode in the ABPR under the measured root mean square (rms) magnetic field \( B_{\text{RMS}} \) as follows:\(^\text{15,16}\)

\[
\frac{\theta}{B} = V_{\text{total}}L_{\text{total}} \approx \frac{0.5}{B_{\text{RMS}}} \sin^{-1} \left( \frac{R_{\text{LIA}}}{20V_{\text{SM}}} \right). \tag{3}
\]

Since the LIA output \( R_{\text{LIA}} \) only reflects the absolute value of the rms output of the ABPR depending on the polarization rotation that had taken place in the sample under the AC modulated magnetic field, it is not easy to account for the opposite Faraday rotations of the diamagnetic substrate and the ferrimagnetic main Bi:YIG and buffer layers. It is known that the silicon has a relatively large negative Verdet constant of about \( \sim 1500 \mu \text{T/m} \) at the 1550-nm wavelength due to its diamagnetic responses compared to the positive Verdet constant of the silica glass of 6.6 \( \mu \text{T/m} \).\(^\text{20,21}\) Unlike in the case of measuring the Verdet constant of metal-substituted YIG crystal films on the silica glass substrate,\(^\text{22}\) we must consider the opposite Faraday rotation of the light beam passing through the Si substrate of the large diamagnetic property to achieve the net Faraday effect within the main Bi:YIG and buffer layers. In order to eliminate the opposite Faraday rotation due to the Si substrate from the measured LIA output \( R_{\text{LIA}} \), we rotated the sample to two different angles of \( 0^\circ \) and \( 60^\circ \) with respect to the light beam path and measured the \( R_{\text{LIA}} \) values. Then, we compared the measured \( R_{\text{LIA}} \) values with analytically calculated \( R_{\text{LIA}} \) values for the two optical path length cases with the known Verdet constant of the Si substrate and the known refractive indices of the main Bi:YIG film, the Bi:YIG buffer layer, and the Si substrate. The refractive indices of the Bi:YIG, Bi:YIG, and silicon are known to be 2.32, 2.44, and 3.47, respectively, at the 1.55-μm wavelength.\(^\text{22,23}\) The refractive index of the Bi₀.₃:NiGG buffer layer was estimated to be similar to that of a film-type ferrite garnet [Bi₁₋ₓ, Lu, Y]₁₋ₓ(Fe, Ga)₃O₁₂] whose refractive index was reported as 2.15 ± 0.1 at the 1550-nm wavelength.\(^\text{24}\) In the calculation of the \( R_{\text{LIA}} \) values, the direction of the applied magnetic field, the Fresnel reflections at the layer interfaces, the Faraday rotation at each layer, and the interference effects among the boundaries between layers were also considered for both the beam incidence angles at the 1550-nm wavelength.

The calculated Verdet constants (horizontal axis) corresponding to the outputs of the LIA (\( R_{\text{LIA}} \), vertical axis) for the cases of the Bi₁₋₂:YIG and Bi₁:YIG film samples on the Si substrate with the Bi₁:YIG and Bi₂:YIG buffer layers, respectively, are shown in Figs. 3(a) and 3(b). In Fig. 3(a), the LIA output \( R_{\text{LIA}} \) is originally about 0.048 mV when the Verdet constant of the Bi₂:YIG films is zero. As the Verdet constant of the Bi₂:YIG film increases, the \( R_{\text{LIA}} \) value decreases due to the opposite Faraday rotation of the Bi₂:YIG film with respect to that of the Si substrate until both are the same.
When the Faraday rotation angle of the Bi$_2$:YIG film is larger than that of the Si substrate, the $R_{LIA}$ value starts to increase. If the measured $R_{LIA}$ value is 0.01 mV at the 0° degree beam incident case, as shown in Fig. 3(a), the $R_{LIA}$ value can either decrease or increase when the Bi$_2$:YIG film on the Si substrate is rotated to 60°. Once the $R_{LIA}$ value is decreased when the Bi$_2$:YIG film on the Si substrate is rotated to 60° from 0°, the Verdet constant of the Bi$_2$:YIG film is determined as the value of $V_1$. On the other hand, if the $R_{LIA}$ value...
is increased during the sample rotation, it becomes \( V_{2} \). The hatched and filled areas in Fig. 3 indicate the error ranges calculated from the thickness errors of the layers, as mentioned above.

The Faraday rotation angle per unit magnetic field within the samples with or without a buffer layer consists of the sum of \( V_{\text{Buffer}} \) and \( V_{\text{Subs}} \), as shown in Eq. (2). When the sample is rotated from 0° to 60°, the measured \( R_{\text{LIA}} \) value either decreases or increases depending on the Faraday rotation \([V_{\text{Buffer}} + V_{\text{Subs}}]\) within the Bi:YIG layer compared to that \((V_{\text{Subs}})\) within the Si substrate, as illustrated with arrows in Figs. 3(a) and 3(b). With the known \( V_{\text{Subs}} \) and the measured \( L_{\text{Buffer}} \), we can determine the Verdet constant of the Bi:YIG film. For the samples with a buffer layer, the Faraday rotation within the buffer layer \((V_{\text{Buffer}})\) was calculated by taking the average value of the Verdet constant of each buffer layer treated at each thermal condition which was obtained from the measurement of each of the Bi:YIG films, as shown in Fig. 4. However, there is an uncertain region where the variation of the measured \( R_{\text{LIA}} \) value is not well noticeable during the sample rotation possibly depending on the Bi:YIG film thickness. These uncertainties with the thickness errors are marked as error bars in Fig. 4.

III. MEASURED RESULTS

The measured Verdet constants of the Bi:YIG films [Bi:YIG and Bi:YIG for Figs. 4(a) and 4(c), and 4(b) and 4(d), respectively] as functions of the annealing temperature corresponding to the maximum temperatures of the main annealing processes are shown in Fig. 4 for various conditions of the buffer layer and for two different cooling speeds of the main annealing process (1°C/min and 3°C/min for Figs. 4(a) and 4(b), and 4(c) and 4(d), respectively). The enhanced Verdet constants of the Bi:YIG films are measured at the annealing temperatures from 650°C to 700°C but observed to be dependent on the buffer layer and the annealing speed. The maximum Verdet constant of the Bi:YIG film measured in this experiment is 1.072 \times 10^{38} T/m for the Bi:YIG film on a Bi:YIG buffer layer treated at an annealing temperature of 700°C and at an annealing speed of 3°C/min. Compared to the same Bi:YIG film on a glass substrate fabricated by the same MOD method, the measured Verdet constant for the silicon substrate shows 15.8% improvement. This improvement may be attributed to the crystallized and periodic structure of the surface of the Si substrate and to the smaller difference in the thermal expansion coefficient of the Si substrate \([-3.5 \times 10^{-6}/°C\) (silicon, single crystal)] compared to that of the YIG crystal \([-10 \times 10^{-6}/°C\) (YIG, single crystal)] and that of the glass substrate \([-1.3 \times 10^{-6}/°C\) (glass, fused silica)]. For the Si substrate, the Bi:YIG films show relatively low Verdet constants compared to those of the Bi:YIG films. The maximum Verdet constant of the Bi:YIG film is measured to be only 451 \times 10^{38} T/m for the condition of the annealing temperature of 700°C and the annealing speed of 1°C/min. with no buffer layer and plotted as a black dotted line with open squares in Fig. 4(b). However, the Bi:YIG films with a buffer layer on the Si substrate show significantly improved Verdet constants and are plotted as red and blue lines with closed circles and with open triangles, respectively, in Figs. 4(a) and 4(c). This indicates that the formation of the buffer layer between the films and the Si substrate dramatically improves the crystallinity of the Bi:YIG films. For the case of the Bi:YIG films, as shown in Figs. 4(b) and 4(d), the existence of the buffer layer is not so effective to enhance the MO quality of the films probably due to an improper composition of the metal components.

To analyze the magnetization characteristics of the Bi:YIG films, we measured magnetic moments of the films as functions of the induced external magnetic field using a vibrating sample magnetometer (VSM). Figures 5(a) and 5(b) show the measured results for both Bi:YIG and Bi:YIG films on the Si substrates with Bi:YIG and Bi:NIGG buffer layers, respectively. The saturated magnetic moments \((M_{\text{SAT}})\) of both films, which correspond to the maximum Faraday rotation conditions, are clearly larger at the optimum annealing temperature condition than at the non-optimized annealing conditions. The Bi:YIG film treated at the optimum annealing condition also shows a clear hysteresis loop, which originated

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**FIG. 5.** Measured magnetic moments of the (a) Bi:YIG and (b) Bi:YIG films on the Si substrates with Bi:YIG and Bi:NIGG buffer layers, respectively, as functions of the magnetic field of a vibrating sample magnetometer. The Verdet constant measurements for both samples were performed in the unsaturated linear magnetic field region which is marked with the hatched area between the two dotted lines.
from the ferrimagnetic characteristics of the crystallized YIG, and the hysteresis loop width is much broader than that of the Bi$_2$YIG film. This indicates that significantly improved MO properties of the Bi$_2$YIG film are expected compared to the Bi$_2$YIG film. The unsaturated MO properties, i.e., Verdet constants, of the film samples were measured within the linear magnetization region marked with the hatched areas in Figs. 5(a) and 5(b). Although a large Verdet constant was measured for the Bi$_2$YIG film compared to the Bi$_2$YIG film, as shown in Fig. 4, the saturated magnetization levels $M_{\text{sat}}$ of both the film samples shown in Fig. 5 are similar. This feature can be explained by the relationship between the magnetization and the Faraday rotation as follows:

$$\theta_F = \frac{\pi N M}{\lambda \varepsilon_0 M_{\text{sat}}} \left(\alpha_+ - \alpha_-\right),$$  

(4)

where $\theta_F$ is the angle of Faraday rotation; $n_0$ is the refractive index of the magneto-optic material; $\lambda$ is the wavelength of the propagating beam; $\varepsilon_0$ is the permeability of free space; $M$ and $M_{\text{sat}}$ are the projected amount of the non-saturated and saturated magnetization along the direction of the external magnetic field, respectively; and $\alpha_+$ and $\alpha_-$ are the right (+) and left (−) circular electronic polarizability of the material, respectively. The Faraday rotation angle $\theta_F$ of the beam propagating through the magneto-optic material under an applied magnetic field obviously increases linearly as a function of the magnetization projection ($M$) along the magnetic field which corresponds to the measured magnetic moment in Fig. 5. However, $M$ and $M_{\text{sat}}$ affect not only the amount of Faraday rotation but also the difference of the circular electronic polarizability ($\alpha_+ - \alpha_-$) which originated from the difference between the resonance energies of the electrons in the crystal structure of the Bi$_2$YIG films for both right and left circular polarized electromagnetic waves. Thus, similar levels of saturated maximum magnetization, $M_{\text{SAT}s}$, of the Bi$_2$YIG and the Bi$_2$YIG films, as shown in Fig. 5, do not always mean similar levels of magneto-optic performances for both the materials but may result in a different birefringence depending on the different MO performance. The circular polarization birefringence of the Bi$_2$YIG film which is proportional to ($\alpha_+ - \alpha_-$) in Eq. (4) is much larger than that of the Bi$_2$YIG film for optical input beams having both circular polarization components.

IV. CONCLUSIONS

Spin-coated Bi$_2$YIG film samples were prepared on Si wafers by a MOD method which can be considered to be a potential CMOS-compatible process, and their MO properties were investigated after various thermal treatments. The Verdet constants of the films were measured under an unsaturated linear magnetic field range when the films were formed with various buffer layer conditions and treated at various annealing temperatures and annealing speeds. The maximum Verdet constant of a Bi$_2$YIG film formed on the Si substrate with a Bi$_2$YIG buffer layer was obtained to be 1.072 038°/T/m after a thermal treatment at a maximum annealing temperature of 700 °C and annealing speed of 3 °C/min. The measured result indicates that the magneto-optic characteristics of the Bi$_2$YIG films on the silicon substrate are enhanced significantly compared to those formed on the glass substrate. The enhanced MO characteristics of the Bi$_2$YIG films on the silicon substrates will be useful for demonstrating integrated optical waveguide-type isolators.

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