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Growth of crystalline $Gd_3Ga_5O_{12}$ thin-film optical waveguides by pulsed laser deposition

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Abstract

Crystalline thin-film optical waveguides of gadolinium gallium garnet ($Gd_3Ga_5O_{12}$) have been deposited onto heated yttrium aluminium garnet ($Y_3Al_5O_{12}$) substrates by a pulsed laser deposition technique. The refractive indices of the films have been inferred from dark-mode prism coupling measurements and are in excellent agreement with the bulk crystal.

Optical waveguides of laser gain media are highly desirable because the high intensity-length product and good pump–signal mode overlap, which can be achieved in the waveguide geometry, leads to a reduced threshold pump power as compared to bulk lasers. GGG is an attractive laser host crystal with a broad tunability since laser action has been observed in Cr^{+} doped GGG [1]. Although optical waveguides of GGG have been fabricated by ion-implantation of the GGG crystal [2,3], the technique is very expensive with the need for an ion generator, accelerator, ion separator and a raster-scan deflector. Cheaper methods to produce waveguides involve the deposition of an epitaxial thin film on a suitable substrate without the need for expensive, and often difficult to produce, bulk crystals. Furthermore, the large refractive index difference that can be obtained between the waveguide and substrate leads to confinement of the propagating light in a narrow mode in the waveguide layer. Deposition techniques, such as rf-sputtering, dc-sputtering, flash evaporation or thermal evaporation are not very suitable since they would not be able to reproduce the complicated stoichiometry of the target in the film. Furthermore the films grown by these techniques are rarely optically perfect, single-crystal waveguides.

Since 1987, when Dijkkamp et al. [4] prepared Y–Ba–Cu–O superconducting thin films by pulsed laser deposition (PLD), the technique has emerged as a viable means of growing stoichiometric and epitaxial thin-film optical waveguides for optoelectronic applications. These include $KNbO_3$ [5], $LiTaO_3$ [6], $BaTiO_3$ [7], and KTP [8], thin films with potential applications as diode-pumped frequency conversion devices, $LiNbO_3$ [9] and $LiTaO_3$ [10] thin films as surface acousto-optic wave devices and YIG [11,12] thin films as magneto-optic devices. Most importantly, PLD is able to reproduce the stoichiometry of the target onto the film and it was a natural progression to use the technique to deposit complicated multi-component, single-crystal, laser-host optical waveguides. To our knowledge, the successful growth of GGG thin films on single-crystal YAG substrates by PLD has not been previously reported.

In order to facilitate epitaxial growth of the GGG thin film, the substrate needs to have a close lattice match to the film. GGG is a cubic structure at room

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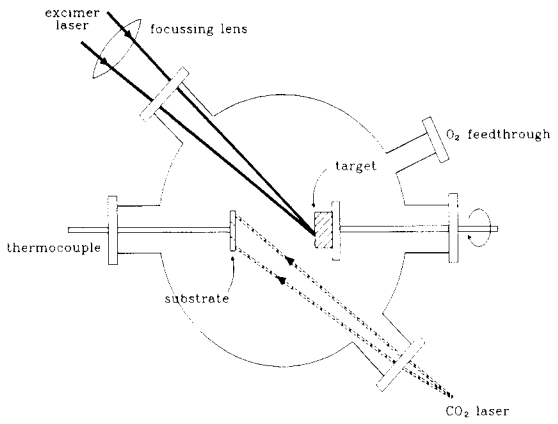

Fig. 1. Experimental arrangement used for PLD and heating the YAG substrate with a CO_2 laser.

temperature with a lattice parameter of 12.377 Å [13], and YAG is also cubic with a lattice parameter of 12.016 Å [13], at room temperature. This corresponds to a lattice mis-match between the film and substrate of only 2.9% which is far less than the maximum mis-fit of 7% which can be accommodated by uniform elastic strain and other crystal defects [14]. The YAG substrate also fulfils the refractive index requirement for optical confinement into the GGG thin layer. For the film to act as a waveguide, the substrate must have a lower refractive index ($n_{YAG} = 1.816$ at 633 nm) than that of the film ($n_{GGG} = 1.965$ at 633 nm) [15].

The experimental arrangement for the PLD of thin films is shown in Fig. 1. The pulsed laser is a KrF excimer laser (Lambda Physik, model LPX) operating at $\lambda = 248$ nm, pulse duration $\tau = 20$ ns, laser repetition rate = 20 Hz and an average laser energy per pulse of ≈ 200 mJ. The cylindrical GGG single-crystal target was ablated on its side using the excimer laser beam with an incident laser fluence of ≈ 5 J/cm [2]. The target had a spiral motion which reversed its direction every four revolutions allowing the ablation of as much fresh material as possible and minimised the growth of microstructures on the target due to the laser beam.

The YAG substrate was positioned 2.5 cm away from the target and heated on its front surface with a 10 W CO_2 laser beam as shown in Fig. 1. This arrangement allowed the substrate to be heated to a maximum temperature of 750°C, monitored with a chromel–alumel (K-type) thermocouple glued to the reverse side of the substrate with silver paint. The use of a CO_2 laser beam for substrate heating has several advantages to conventional substrate heating methods which use resistive wires or filament bulbs, since it is much cleaner without the problems of outgassing of the vacuum seals or oxidation of the heating element. The setup is far more efficient than the conventional heating methods since the maximum temperature is generated on the front surface of the substrate and not at its back or within the substrate heater.

Deposition was carried out in a vacuum chamber, at a background pressure of $\approx 10^{-5}$ mbar and then backfilled with an oxygen partial pressure ranging from 1.2×10^{-2} mbar to 1.9 mbar just prior to ablation. The films were grown for 20 min and then slowly cooled for 30 min in the ambient oxygen gas. The stoichiometry of the films was determined using both energy dispersive X-ray analysis (EDX) and Rutherford backscattering spectroscopy (RBS). The crystallinity of the films was investigated using both grazing incidence and conventional X-ray diffraction techniques.

Fig. 2 is a plot of the composition of the thin films (as measured by EDX) as a function of the substrate temperature, prepared in an ambient O_2 pressure of $\approx 10^{-1}$ mbar. From Fig. 2 it is seen that the [Gd]/[Ga] molar ratio of the films is close to the bulk GGG single-crystal [Gd]/[Ga] molar ratio of 0.6 (corresponding to the dashed line in Fig. 2) and not dependent on the substrate temperature. All the films appear to be very slightly Ga deficient with an average [Gd]/[Ga] molar ratio of 0.68.

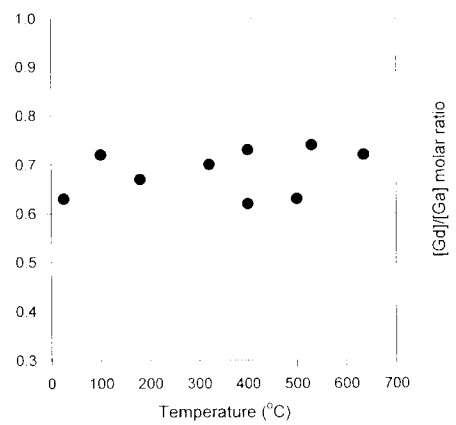

Fig. 2. Ratio of gadolinium, [Gd], and gallium, [Ga], concentration of the films as a function of the substrate temperature during deposition. The [Gd]/[Ga] ratio has been calculated by energy dispersive X-ray analysis. The dashed line corresponds to the [Gd]/[Ga] ratio for a GGG bulk crystal.

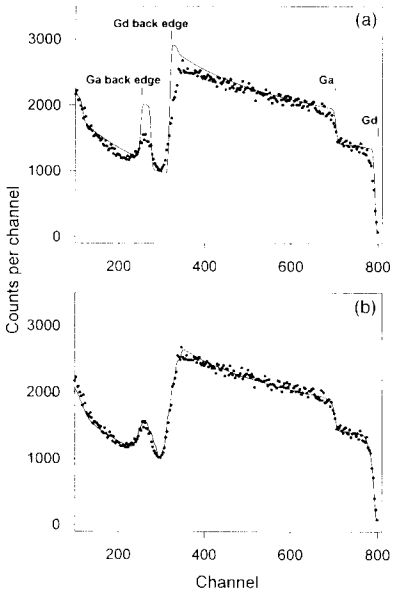

Fig. 3. Rutherford backscattering spectra of a film grown on a (444) oriented YAG single-crystal substrate in which the points are the experimental results and the solid lines represent the theoretical fit by assuming (a) an 1830 nm thick $Gd_3Ga_5O_{12}$ film and (b) a 1620 nm thick $Gd_3Ga_{4.5}O_{11}$ film over a 180 nm linear diffusion layer with the substrate and under a 100 nm linear diffusion layer of $GdGa_2O_{12}$.

The stoichiometry of the GGG thin films was further investigated using RBS analysis. Fig. 3 shows the RBS spectra of a film deposited at a substrate temperature of 675°C, in which the points represent the experimental data and the solid lines represent the computer simulation of this data by assuming a GGG film with a particular composition and thickness. In Fig. 3a, the experimental data is modelled by assuming a 1830 nm thick film with a composition of $Gd_3Ga_5O_{12}$ which fits well with the experimental data apart from near the Gd and Ga back edge where there may be some diffusion of the film into the substrate. The solid line in Fig. 3b represents the best fit to the experimental data by assuming a 1620 nm thick $Gd_3Ga_{4.5}O_{11}$ layer. Between this layer and the substrate we assumed a 180 nm thick layer where a linear diffusion of the film into the substrate has occurred. In addition, above the 1620 nm thick $Gd_3Ga_{4.5}O_{11}$ layer, we assumed an oxygen enriched surface layer ($GdGa_2O_{12}$) which has a 100 nm thick linear diffusion into the $Gd_3Ga_{4.5}O_{11}$ layer.

The crystalline quality of the GGG thin films was investigated with a D-500 Siemens, grazing incidence and conventional X-ray diffractometer, using the Kα emission from a Cu anode. Fig. 4 shows the X-ray spectra of GGG thin films deposited at various substrate temperatures. Fig. 4a represents a film grown at 530°C which is completely amorphous with only the (444) YAG substrate peak present. All the GGG films grown at this temperature or below were completely amorphous. Fig. 4b shows that at a substrate temperature of 650°C, a small amount of GGG crystallization was occurring and the (444) GGG peak was identified at a lattice d-spacing of 1.809 Å (cf. bulk crystal (444) peak at 1.787 Å [16]). At a substrate temperature of 685°C, the (444) GGG diffraction peak was more prominent and sharper with the d-spacing measured at 1.808 Å (Fig. 4c). Fig. 4d shows that at a temperature of 750°C, the (444) GGG peak was significantly sharper and located at a d-spacing of 1.798 Å so that the lattice spacing difference between the film and the bulk crystal of GGG was only 0.6%. In all the X-ray spectra, no other diffraction peaks were identified implying that the GGG had grown exclusively in the highly oriented (444) plane parallel to the (444) YAG surface, at a substrate temperature of 750°C and an oxygen partial pressure of 4×10^{-2} mbar. Subsequent annealing of the film shown in Fig. 4d, in a tube furnace for 5 h at a temperature of 1100°C, with 1 ℓ/min flowing oxygen, improved the crystalline quality of the film.

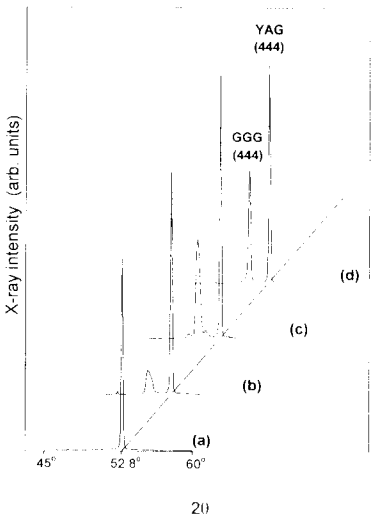

Fig. 4. X-ray diffraction spectra of the films formed on (444) YAG single-crystal substrates at an oxygen partial pressure of $\approx 10^{-2}$ mbar and (a) a substrate temperature of 530°C, (b) a substrate temperature of 650°C, (c) a substrate temperature of 685°C and (d) a substrate temperature of 750°C. The dashed line corresponds to the diffraction angle of the (444) YAG substrate peak at $2\theta = 52.8°$.

The (444) GGG diffraction peak increased in height and shifted slightly to a lattice d-spacing of 1.788 Å which is in excellent agreement with the (444) GGG peak for the bulk crystal. The lattice difference between the film and the bulk crystal of GGG corresponds to less than 0.06%.

The waveguiding properties of the GGG film shown in Fig. 4d were investigated by coupling light from a He–Ne laser at 633 nm into the layer using a rutile prism. The m-lines of the TE_0 and TE_1 modes were clearly visible and the effective refractive indices, n_{TE0} and n_{TE1} respectively, of these two TE modes were calculated by measuring the angles at which they were coupled into the film. It was thus calculated that n_{TE0} and n_{TE1} had values of 1.953 and 1.885 respectively. Substitution of these values into the guidance condition for an asymmetric waveguide enabled the refractive index and thickness of the films to be evaluated [17–19]. The refractive index was calculated to be 1.972 which is in close agreement with the bulk crystal value of 1.965 [15]. The thickness of the film was calculated to be 1.04 μm which implies that the deposition rate was ≈ 0.4 Å/pulse (≈ 50 nm/min).

In conclusion we have reported the fabrication of crystalline (444) GGG thin films on (444) YAG substrates by the pulsed laser deposition technique. The highly oriented GGG films were deposited at a substrate temperature of 750°C and an oxygen partial pressure of 4×10^{-2} mbar. Waveguide coupling measurements show a refractive index value of 1.972 in close agreement with the bulk crystal.

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