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Planar single-crystal thin-films of YAG obtained by ion implantation and thermal exfoliation: Mechanical properties ☆

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ABSTRACT

We report on the fabrication of single-crystal thin films of Yttrium Aluminum Garnet (YAG, $Y_3Al_5O_{12}$) obtained by thermal exfoliation from bulk crystals after deep He-ion implantation. The film qualities and exfoliation process were determined by AFM, optical microscopy, SEM, optical profilometer and nanoindentation. The resulting films were subjected to annealing at ~1200 °C to relax the residual strain and film curvature that arose from the ion implantation process.

1. Introduction

Garnet crystals, such yttrium iron (YIG) and yttrium aluminum (YAG) garnet and related crystals, e.g. yttrium lanthanum fluoride (YLF) are essential to a wide variety of laser-, photonic-, and acoustic-based devices [1]. For integrated versions of these devices, thin films of the oxides are required. Despite important advances in the growth of single-crystal thin-films of YAG and other materials, the formation of fully crystal freestanding films with bulk-like properties has yet to be fully realized. One approach to fabricating these films is via ion-implantation-induced exfoliation. In this approach, high-energy ion-implantation of light ions is used to form a heavily implanted region below the surface of a target bulk single-crystal sample. This heavily implanted region forms the basis for film lift-off either through selective etching or other crystal separation approaches. In the past we have developed a technique called Crystal Ion Slicing (CIS) that is based on light ion implantation for exfoliation of thin films of complex oxides and other materials [2–10]. Thin-film devices that were obtained using this exfoliation technique were for applications in telecom and sensing [11–22]. In addition, TEM studies have been used to understand the atomic-scale physics of heavily implanted oxide crystals and its role in the exfoliation mechanism of various material systems [23–26].

The present work demonstrates fabrication of thin films of YAG for microsystem applications. Our approach uses ion-implantation followed by rapid heating in air to obtain the exfoliated sample. The fact that garnets such as YAG have a complex bonding structure with nearly equal surface energies for several low-index crystal planes causes the YAG crystal planes, to separate via conchoidal fracture; this makes the ion-induced microcleavage more challenging, as no clear cleavage plane is present. Further, the realization of large films requires control over the vertical thrust that is caused by implantation-induced strain. Our study demonstrates that relatively large high-quality films can be obtained and delineates the procedure needed to obtain planarized films, as well as the factors that control the size of free-standing films.

1.1. Experimental

Fabrication of the thin films followed a modified approach to that in crystal-ion-slicing (CIS) where light ions are implanted in the crystal creating a sacrificial layer that is more chemically active or has a different thermal expansion coefficient. Samples were prepared for exfoliation by implanting He^+ ions several microns below the surface of the crystal. Samples of 1 cm^2 were diced from a Czochralski-grown YAG wafer with ⟨001⟩ axis perpendicular to

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the surface. These samples were ion implanted (Dynamitron ion implanter) at energies of 1–4 MeV, with a projected ion range of 3–10 μm due to the ion-energy-dependent stopping range and straggle region of 160–410 nm, calculated using Stopping and Range of Ions in Matter (SRIM) simulation package [27]. The ion dose was varied between 2.5 and 8×10^{16} ions/cm^2. The samples were mounted on a water-cooled (17 °C) holder and were implanted 7° off the perpendicular direction.

In order to exfoliate the films, the samples were subjected to high-temperature ramping in a rapid thermal anneal (RTA) furnace (HEATPULSE 410) or a tube furnace (Thermolyne 21100) that was pre-heated to the desired temperature. In all cases, the implanted samples were introduced to the furnace at the required temperature for each experiment in a time interval of 4 s. After exfoliation, samples were removed from the heating device and allowed to cool to ambient temperature.

The surface of the films was analyzed by several methods: an optical profilometer (Wyko NT9100) and a long-working-distance, side-mounted optical microscope (custom built) were used to determine the long-range curvature of the films. For high-resolution investigation of the film's surface morphology, a scanning-electron microscope (SEM; Hitachi 5000 & LEO 1450) and an atomic-force microscope (AFM; Veeco MutiMode V) were used.

Mechanical properties of the implanted samples were measured using a nanoindenter (Agilent G200). Such a nanoindentor allows probing the mechanical properties as a function of depth on a nanometer length scale.

1.2. Results and Discussion

Samples were irradiated over a range of energies from 1.2–3.8 MeV such that a variety of films thicknesses could be obtained; this variation allowed us to examine the influence of film thickness on the exfoliation process. Fig. 1a–c shows side-view images obtained, via a scanning electron microscope (SEM), of implanted YAG crystals at three different energies 1.2, 3 and 3.8 MeV. The crystals were lightly etched in phosphoric acid to indicate the narrow (a few hundred nm) stopping regions in each case. The yellow[1] curves in Fig. 1 shows the calculated projected ion range and straggle distance at each implantation energy for He in YAG crystal using SRIM. Earlier reports for other related material systems had shown that exfoliation occurred generally for doses of $\sim 5 \times 10^{16}$ ions/cm^2 for samples that were ~10 μm in thickness [8]. For the depth-dependent measurements, the dose was reduced as the implantation energy was lowered over the range given above from 5×10^{16} to 3.5×10^{16} ions/cm^2 because of the narrowing of the straggle region for lower energy. This procedure allowed us to maintain approximately the same He concentration in the implantation region at each implantation depth. A representative image in an SEM micrograph of the film can be seen in Fig. 1d. In this image, the implanted side of a 7 μm thin film is facing upward. Note that the films exhibit bowing due residual He near the back surface (top side in Fig. 1) as a result of the "tail" of the implant region.

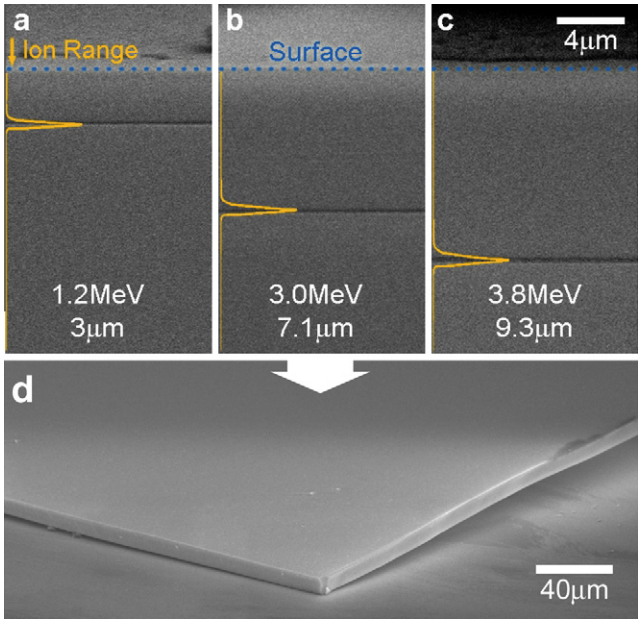

Fig. 1. (a–c) Scanning electron microscope (SEM) images of YAG crystal implanted at different energies (1.2 MeV, 3 Mev and 3.8 MeV). The doses for (a–c) are as follows: (a) (4×10^{16} ions/cm^2), (b) and (c) (5×10^{16} ions/cm^2). The overlaid light (yellow online) curve on the left of each inset is a TRIM calculation of He ions stopping range. (d) SEM image of exfoliated film of 3 MeV implanted YAG crystal on Si a wafer. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

A detailed atomic-scale mechanistic study of the slicing mechanism in other oxides, particularly $LiNbO_3$, have shown that He bubbles nucleate at implant-induced defects [24,28]. These defects then lead to the formation of a sheet of 'nanocleaves' in the highly-implanted crystal along the implant plane. The strong thermal gradients resulting from the presence of rapid oven heating provide localized stresses, which drive the exfoliation process. This mechanism can be characterized, as in many thermally activated processes [29], with an effective activation energy, E_a. This activation energy can be estimated using the usual Arrhenius expression,

$$E_A \propto K_B T \log(t_c) \quad (1)$$

where the time needed for exfoliation at temperature T is t_c and k_B is the Boltzmann constant. This thermally activated process was investigated by exposing samples to different exfoliation temperatures from 750 °C to 1200 °C and the thermal activation time was measured using a standard laboratory timepiece. The relation between slicing temperature and the time required to induce exfoliation, as indicated in Eq. (1), leads to an activation energy of 0.43 ± 0.04 eV. Experiments by Tong et al. [29] in Group IV semiconductors have shown that the exfoliation of a hydrogen-implanted sample has a characteristic activation energy, which for these elemental semiconductors is close to the bond energy of semiconductor. In our case, the material is more heavily implanted and thus a lower overall activation energy is reasonable.

Our nanoindentation measurements support the fact that implantation leads to a change in the mechanical properties in the implantation region. In this experiment, nanoindentation was used to probe the near-surface region of two different samples. Both samples were implanted with 1.2 MeV He ions at a dose of 3×10^{16} ions/cm^2. For the first experiment the sample was exfoliated and then nanoindentation was carried out on the *exfoliated* surface and compared to that on the surface of an unimplanted sample of YAG. The results showed that the straggle region has a Young's modulus that is repeatedly lower by at least 5% than that of the unimplanted sample, which is consistent with the crystal in the implantation region being damaged with point defects and dangling bonds [30]. In the second experiment, the Young's modulus of an implanted (unexfoliated) sample was characterized at different depths and compared with that of the unimplanted crystal. These measurements showed that the implanted sample had a decreased modulus beyond ~450 nm, which agrees with a decrease in the elastic modulus of the implanted region (~2.8 μm). For indentation depths significantly less than the straggle depth of the implanted samples, the Young's modulus was statistically

[1] For interpretation of color in Fig. 1, the reader is referred to the web version of this article.