Laser-Induced Forward Transfer-printing of focused ion beam pre-machined crystalline magneto-optic yttrium iron garnet micro-discs

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Abstract: We present femtosecond laser-induced forward transfer of focussed ion beam pre-machined discs of crystalline magneto-optic yttrium iron garnet (YIG) films. Debris-free circular micro-discs with smooth edges and surface uniformity have been successfully printed. The crystalline nature of the printed micro-discs has not been altered by the LIFT printing process, as was confirmed via micro-Raman measurements.

OCIS codes: (350.3390) Laser materials processing; (220.4000) Microstructure fabrication; (140.7090) Ultrafast lasers; (160.3820) Magneto-optical materials.

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1. Introduction

Laser-Induced Forward Transfer (LIFT) is a laser-based additive printing technique [1–4] which has been routinely employed for the printing of many different materials, on various substrates for applications as diverse as embedded electronic circuits to integrated optical devices, biosensors and tissue engineering. LIFT is essentially a single-shot, serial printing approach that enables the creation of any arbitrary1D/2D user-defined pattern that is formed via a spatially controlled point-by-point pixelated deposition of the required material. This

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laser-based approach has been frequently exploited for printing because of several inherent advantages such as requiring a standard ambient environment for its implementation, providing the possibility of printing a wide range of materials, and allowing printing of multi-layered stacks composed of dissimilar materials on both structured and planar substrates. Furthermore, LIFT printing allows precise control of the size and shape of the printed pixels through the possibility of controlling the incident laser parameters such as wavelength, pulse-duration, and also precise beam shape.

Nevertheless, there are two important issues associated with LIFT printing which need to be addressed if precise and debris-free printing of high quality deposits is to be achieved. The first concerns the unavoidable partial ablation or melting of a surface region of the printed material (termed the donor) due to the absorption of the laser pulse responsible for the forward transfer process. Different research groups have proposed and demonstrated the use of some variants of the basic LIFT process to circumvent this issue. These LIFT-variants are based on the use of an absorbing layer [5–10] sandwiched between the donor and the carrier substrate which thereby eliminates the direct interaction between the laser radiation and the donor. This sacrificial layer, which is often referred to as a dynamic release layer (DRL), decomposes into gaseous fragments upon absorption of the laser pulse, and provides the explosive push required to propel the donor towards the receiver. However, the use of a DRL for LIFT-transfer of comparatively brittle materials such as single crystals is problematic since the growth of a thin crystalline donor film directly on top of a polymer DRL is impossible because the high substrate temperatures of ~600-800°C required for thin film epitaxial growth are far higher than the polymer decomposition temperature of ~250°C.

Growth of thin films that are fully crystalline is difficult, and only a few techniques such as pulsed laser deposition (PLD) are known to achieve this. However, PLD-grown crystals are very well bonded to the substrate due to the epitaxial growth that results from PLD on heated substrates. It is practically extremely difficult to separate such a crystal film from the substrate onto which they have been grown, and consequently LIFT-printing of such crystalline thin films would require an inventive step that would assist crystal transfer via LIFT.

The second problem is related to the unavoidable shearing and tearing of the donor around its edges during LIFT transfer. This presents therefore an important constraint and sets a limit both for debris-free transfer and also for transfer of donors that are fragile, brittle, are of substantial thickness (> a few μm), or composite stacks where edge quality or layer mixing is an issue. A plausible solution for this would be to implement, prior to the LIFT-printing, a pre-patterning step to precisely define the spatial extent of the donor to be transferred thus subsequently achieving a much more precise and controlled printing. A laser-based pre-indentation approach that uses preliminary laser pulses to define and weaken a boundary and a subsequent pulse to LIFT the donor has already been reported in the literature [11], however, the non-uniform edge quality remains an issue.

In order to alleviate all of the above problems, we have earlier proposed and demonstrated [12] an innovative approach that uses a Focused Ion Beam (FIB) machine to assist in partial pre-machining of the donor prior to LIFT. The FIB pre-machining defines and weakens a boundary along which the donor pixel can be easily ripped and propelled forward without undue shattering. We have observed that it is not essential to pre-machine throughout the entire depth of the donor as this prevents the build-up of laser-induced pressure behind the material intended for transfer, and hence can result in no transfer occurring. For an optimum pre-machining depth, a uniform and clean deposition of the donor onto the receiver substrate was observed. We have reported this successful LIFT-printing of intact amorphous ZnO pixels with smooth edge quality and well-defined shapes via this route. In this contribution we present our initial results from the implementation of this methodology to print debris-free, intact, crystalline yttrium iron garnet (YIG) pixels and we believe that is the first time that an intact single crystal material has been successfully LIFT-printed.

This successful result is an important step that will help spread the applicability of LIFT-printing for the fabrication of a range of different crystalline photonic devices. One immediate

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example is a whispering gallery mode micro-resonator where edge quality and feature shape are crucial parameters. Other examples would include gain-enhanced guided-wave devices fabricated via local deposition of a doped crystalline host onto waveguide structures or integrated modulator devices produced via deposition onto novel geometries such as an end-face of an optical fiber.

2. Experimental

A tunable ultrafast Ti:sapphire laser system operating at 800nm, with a repetition rate of 1kHz, a pulse duration of ~150fs, and delivering a maximum energy of ~2mJ per pulse was utilized for these LIFT-printing experiments. The laser beam was initially collimated and then homogenized using a refractive optical element to yield a spatially uniform top-hat intensity profile which was ~6mm in diameter. Transformation of the beam profile from a Gaussian into a top-hat profile ensured a uniform intensity profile which would result in uniform laser energy deposition, and hence a uniform thrust from the laser onto the donor that is being transferred. An aperture positioned in the path of the beam after the homogenizer allowed for the selection of the shape and size of the incident laser beam, which was then imaged with a large value of demagnification using a microscope objective lens onto the carrier-donor interface. The diameter of the imaged circular beam profile was set to ~10μm, and was intentionally adapted to match the diameter of the pre-machined donor.

The crystalline YIG donor films were previously grown onto $10 \times 10 \times 1mm^3$ yttrium aluminium garnet (YAG) carrier substrates by pulsed laser deposition. The YIG donor films were grown to have a thickness of ~1μm. YIG depositions were performed in oxygen at a pressure of $\sim 3 \times 10^{-2}$ mbar by ablating a single-crystal YIG target with a KrF excimer laser (λ = 248 nm) with a fluence of ~3 J/cm^2. Substrate temperature was set at a value high enough to ensure crystallization of the film, which was verified by XRD analysis: the film was found to be epitaxial with the same orientation as the YAG (100) substrate.

The choice of YIG as the donor was because its absorption is well matched with the two-photon absorption wavelength (400nm) when LIFT-printing with the fs-laser system that was operated at a wavelength of 800nm. For effective LIFT-printing the essential requirement is that the carrier is transparent at the fundamental laser wavelength, (and also at its harmonics if multi-photon absorption is the predominant mechanism), while the donor material exhibits a strong absorption that leads to effective detachment from the carrier. This absorption-contrast between the YAG carrier and the YIG donor at the two-photon absorption wavelength is shown in Fig. 1, and in this case the this contrast is almost ideal, with YIG transmission at 0% and YAG transmission at ~80%. Our earlier attempts to LIFT-print pre-machined crystalline gadolinium gallium garnet (GGG) were unsuccessful because absorption of the laser energy by GGG at the important two-photon absorption wavelength of 400nm was minimal and the absorption contrast (~5%) between the YAG carrier and the GGG donor was not sufficiently large. The absorption plots for YIG and GGG in Fig. 1 are indicative of this rationale underlying the selection of YIG over GGG for the successful demonstration of single crystal LIFT trials reported here. For a fixed operational laser wavelength, lack of absorption of the laser energy by the donor material at the corresponding multi-photon wavelengths can limit the range of donor materials that can be LIFT-printed.

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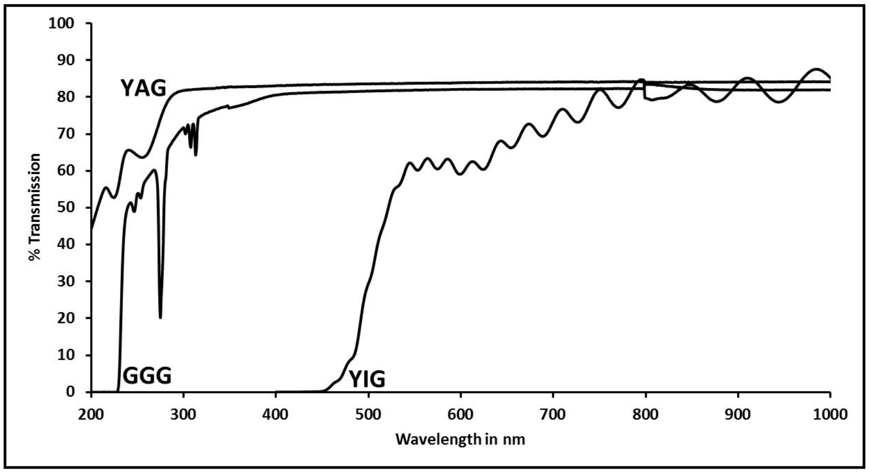

Fig. 1. Measured absorption spectra for YAG carrier and the YIG (reported in this paper), and GGG donors (used in previous unsuccessful LIFT trials) grown via PLD. The periodic fringes in the YIG spectra are thin-film etalon fringes resulting from multiple reflections.

Circular ring patterns that had outer and inner diameters of ~10µm and ~8.5µm respectively were pre-machined into the YIG donor films using the FIB. The accelerating voltage for the ion beam was ~30kV for an ion current of ~2.8nA. As FIB machining of non-conductive dielectric/crystalline material such as YIG with a beam of energetic positive (gallium) ions results in the build-up of the uncompensated excess positive charge over the surface of the material, a charge neutralizing gun incorporated in the FIB system was used and provided a stream of negatively charged electrons accelerated to an energy of ~50 to 80eV to compensate the positive charge. On average, under optimized FIB working parameters, milling a circular ring of the dimensions specified above requires between 23 and 37 seconds. If required, this time span can be further reduced using gas-assisted FIB machining whereby the (mechanical) machining is chemically assisted by use of the reactive species employed in the milling process.

Arrays of rings were pre-machined to different depths (ranging from 50 to 80% of the initial thickness of the donor) to investigate the effect of such a depth dependence on the laser fluence required for successful LIFT-printing. From our previous experiment with LIFT-printing of pre-machined ZnO amorphous donors, it was known that machining throughout the entire depth of the donor was not ideal as this would prevent the necessary laser-induced pressure build-up behind the material intended for transfer. The pre-machined YIG donor/YAG carrier composite was then placed in near proximity to a Parafilm-coated (supplied by Pechiney Plastic Packaging, USA) glass receiver for LIFT-printing; the Parafilm coating provided the dual requirement of greater adhesion and a softer ‘landing’ than on bare substrates such as glass or silicon. To maintain a uniform separation between the donor and the receiver a ~1µm thick Mylar spacer was introduced between them. The incident laser fluences that were trialed for the LIFT printing of the YIG-donor ranged from ~0.2-5.8J/cm^2.

3. Results and discussion

Our first results for LIFT-printing from pre-machined YIG donor films are shown in Fig. 2. Figure 2(a) and Fig. 2(b) show the top and side-view of two different discs LIFT-printed from a donor that had been FIB pre-machined down to depths of ~70%. The laser fluences necessary for the transfer of these pre-machined discs was ~5.8 J/cm^2. Below a fluence of ~1.7 J/cm^2 not even a partial transfer of the pre-machined discs was possible.

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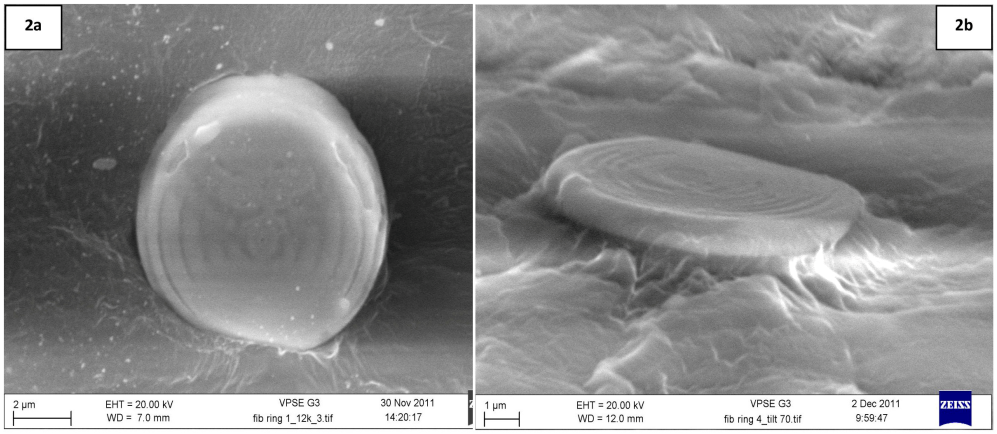

Fig. 2. SEM images showing (a) the top-view and (b) the side-view of FIB pre-machined LIFT-printed YIG pixels.

In the SEM image of Fig. 2(a) that shows the top-view of a LIFT-printed circular disc transferred from a pre-machined donor, the disc however appears to be slightly non-circular; this is because the front section of this disc is slightly embedded in the soft Parafilm that covered the glass receiver. This also restricted our ability to image the side of this LIFT-printed pixel, and hence we have not presented a corresponding image that depicts the edge of this LIFT-printed disc. To examine the edge quality of LIFT-printed pre-machined pixels we show a second SEM image of another FIB pre-machined LIFT-printed pixel (Fig. 2(b)). The SEM image of Fig. 2(b) was taken with the sample holder tilted to 70°. Note that the circular concentric rings that can be seen on the surface of the LIFT-printed discs in Fig. 2 are possibly due to incorrect imaging of the aperture or due to an optical standing wave effect resulting from multiple reflections from within the various carrier, donor and receiver surfaces [13].

To our knowledge this is the first report of LIFT-printing of intact crystalline materials from thin film donors. Considering the degree of difficulty associated with detaching a section of an epitaxially-grown (YIG) film from the lattice-matched crystalline (YAG) substrate, the quality of the LIFT-printed disc appears high with minimal evidence of edge damage or chipping.

To further underline the influence of the laser fluence on the LIFT of such pre-machined discs, Fig. 3 shows a sequence of SEM images for pre-machined donor spots (pre-machined down to depths of 80%) after their LIFT-transfer with increasing incident laser fluences. As seen in Fig. 3, the amount of donor material that is transferred onto the receiver increases with increasing fluence. Above a certain threshold fluence the entire pre-machined disc is LIFT-transferred onto the receiver.

The non-uniform LIFT-printing results (of Fig. 3) at non-optimal fluence could possibly be due to any of the following three possibilities; a small non-uniformity in the incident laser intensity profile, the strong non-linear two-photon absorption in the donor material, or a possible slight incident laser beam alignment error.

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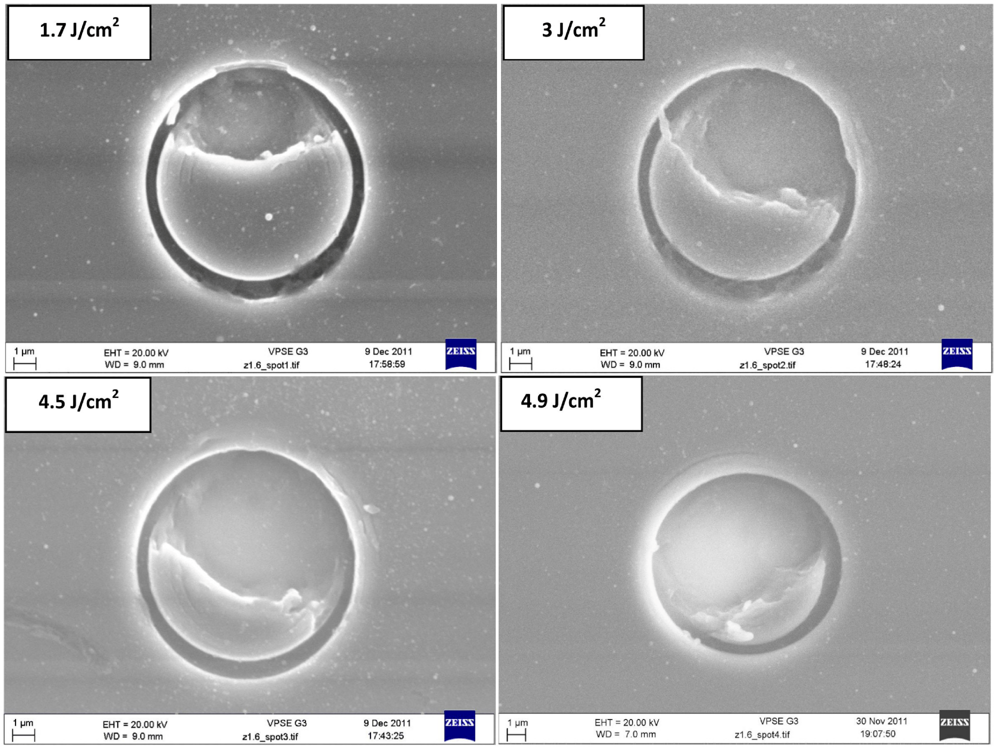

Fig. 3. SEM images of different pre-machined donor rings after LIFT-transfer with different laser fluences. The images are for donors that were pre-machined to a depth of 80% of the donor thickness. The fractional percentage of the donor material that has been LIFT-printed from the pre-machined donor rings at the incident laser fluences of 1.7J/cm^2, 3J/cm^2, 4.5J/cm^2, and 4.9J/cm^2, has been measured to be approximately 29%, 51%, 63% and 72% respectively.

To illustrate the importance of the FIB pre-machining step for effective crystal LIFT, we show results for LIFT from non-machined donors; however, there are major issues related to the shape and edge-quality of the printed pixels, and also importantly with the repeatability of printing. To illustrate this, Fig. 4 shows a non-machined donor after LIFT-printing an array of many pixels. It is clear from this SEM image that there is an inherent non-repeatability involved in LIFT-printing from a non-machined donor.

Fig. 4. SEM image showing a non-machined donor after LIFT-printing of an array of spots.

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In comparison to the results shown in Fig. 2 for LIFT-transfer of pre-machined donors, the SEM images of Fig. 5(a) and Fig. 5(b) show the top and side-view of an irregularly shaped, and partially fragmented pixel, LIFT-printed using a circular laser spot, but from a *non-machined* section of the donor. The undesirable and, in our experience unavoidable, shattering associated with pixels deposited with similar laser fluences, but without the essential pre-machining step is obvious. The SEM image of a non-machined LIFT-printed pixel in Fig. 5(b) further emphasizes the inevitable non-uniformity along its edge and in its thickness. Also seen in Fig. 5(b) is a large semi-circular fragmented section (indicated by an arrow) from another non-machined LIFT-printed pixel. We always observed such debris resulting from the fragmentation of LIFT-printing non-machined donors, and this characteristic can be attributed to the excessive force that the transferred donor pixel experiences while shearing-off from the remaining non-machined donor material. In contrast, for pre-machined donors, a smaller shearing force consequently results in transfer of donor pixels with better edge quality, and almost no fragmentation. The SEM images of Fig. 5(b) that shows side-on view was taken with the sample tilted to 70°.

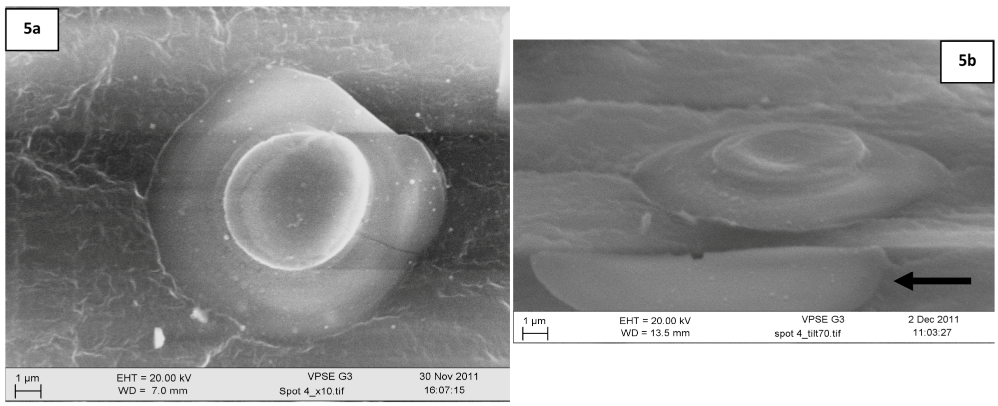

Fig. 5. SEM images showing (a) the top-view, and (b) the side-view of a non-machined LIFT-printed YIG pixel. Note that the arrow in Fig. 5(b) shows fragmented pixel debris associated with typical non-machined LIFT-printing.

The influence of the shearing and tearing of a non-machined donor pixel from the remaining donor, resulting in the transfer of irregularly shaped or fragmented donor pixels can be further visualized by imaging the corresponding donor material after transfer. Figure 6 shows a close-up SEM image of a non-machined donor after LIFT-printing. A direct comparison of this with images in Fig. 3 further emphasizes the usefulness of the pre-machining step that is crucial for the deposition of donor pixels with better edge quality and properly defined shapes. The facetted ring features of the donor (in Fig. 6) showing depth-dependent cracking can possibly be attributed to the preferential cleaving of the crystal along different planes during LIFT. The complementary ‘flying-saucer-like’ feature LIFT-printed as a result of such faceting is evidenced in Fig. 5(b). Moreover, even though the laser beam size was of the order of the inner hollow ring feature formed in the donor, the corresponding donor region influenced by this non-machined LIFT-printing extends over a larger area of the donor. In contrast, for LIFT-printing from pre-machined donors, the area on the donor influenced by the LIFT-printing is restricted to the size of the incident laser beam used to implement the LIFT-printing (Fig. 3).

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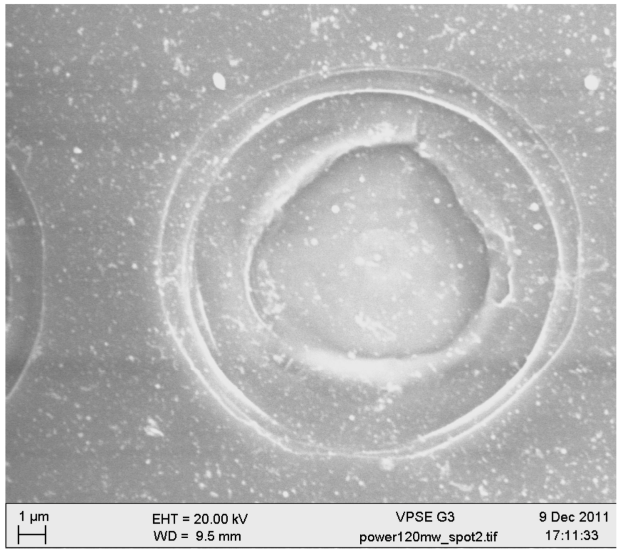

Fig. 6. SEM image of a non-machined donor after LIFT-transfer.

Finally, to confirm that we have indeed LIFT-printed crystalline YIG we have performed micro-Raman characterization of the circular pixels LIFT-printed from pre-machined donors. The Raman measurements were performed using a Renishaw inVia Raman Microscope. The Raman scans were acquired with an incident laser wavelength of λ = 633nm, and the grating used had a resolution of 1200 lines/mm. The trace labeled, "LIFT-printed pre-machined YIG pixel" in Fig. 7 is the micro-Raman scan for a circular disc LIFT-printed from a pre-machined donor. Since these discs were LIFT-printed onto a Parafilm coated glass receiver, we have also acquired a micro-Raman trace for an identical Parafilm on glass receiver that did not have any LIFT-printed material on it. This measurement is shown by the line labeled, "Parafilm" in Fig. 7. The trace labeled, "YIG crystal" in Fig. 7 is the measurement for the c-axis orientated YIG crystal which was used for the pulsed laser deposition of the YIG donor films which were used in the LIFT-printing experiments. Since the YIG donor films were grown on to crystalline YAG carriers, we have also acquired micro-Raman measurements for the YIG donor on the YAG carriers however, since the contribution to the spectrum from the underlying YAG donor is overwhelmingly dominant, it is hard to separate the contributions of the YAG and YIG crystal in this measurement, and we have not included any such spectrum in Fig. 7.

In order to separate the different traces in Fig. 7, and hence allow better visual comparison, the scan data for YIG crystal has been reduced to half of its original value. Similarly, the data for the Parafilm on glass has been shifted upwards by 1000 arbitrary units. Relative coincidence between at least the first five peaks observed in the Raman spectra for the LIFT-printed pre-machined donor pixel with that for the YIG crystal provide convincing evidence of the crystalline nature of the LIFT-printed pixels.

The peaks marked "P" in the spectra for the LIFT-printed pre-machined YIG disc are a contribution from the underlying Parafilm onto which the pre-machined YIG disc has been printed, and these have been indicated via the dashed lines in Fig. 7. The broadened peaks marked "P+Y" in the scan for the LIFT-printed pre-machined YIG disc are possibly a result of the convolution between the peaks observed for the YIG donor crystal and the Parafilm.

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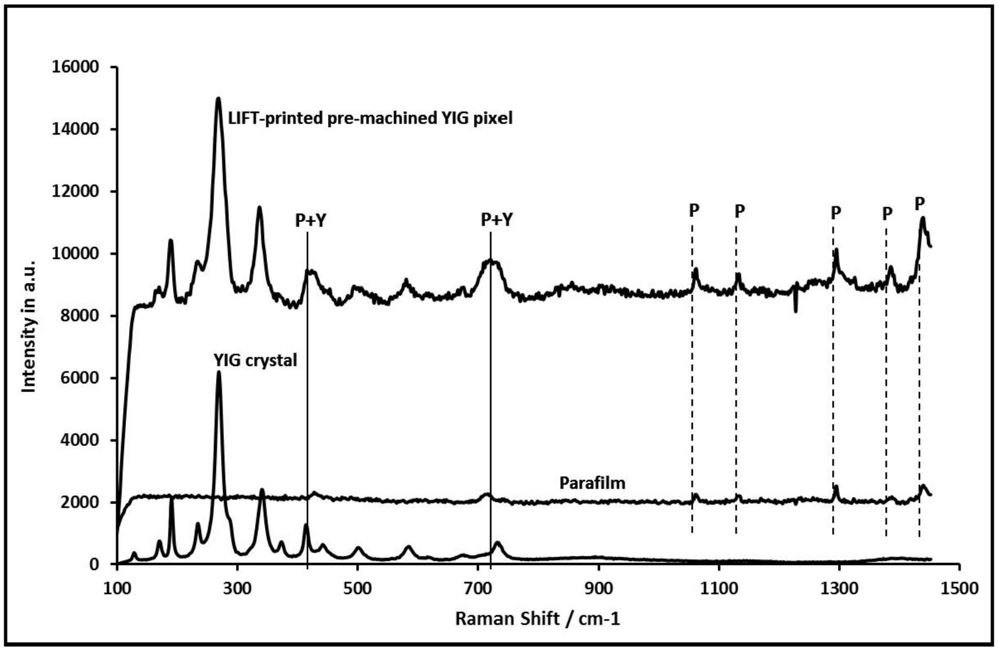

Fig. 7. Micro-Raman measurements of a pre-machined disc LIFT-printed on a Parafilm coated glass receiver, YIG crystal donor, and Parafilm on glass.

It is would have also been possible to characterize the crystalline nature of the LIFT-printed discs using micro-X ray diffraction. This approach would have been the preferred choice for verification of the crystalline nature of the LIFT-printed discs; however, since the discs are not necessarily flat on the Parafilm-coated receiver substrates, this would make the measurements practically difficult to interpret.

4. Conclusions

We have demonstrated successful debris-free, intact, LIFT-printing of FIB pre-machined crystalline YIG pixels with very good edge quality. To our knowledge this is the first report of LIFT-printing of intact crystalline materials from thin film donors. Taking into consideration the degree of difficulty that is associated with detaching a section of an epitaxially-grown (YIG) film from the lattice-matched crystalline (YAG) substrate, we consider these LIFT-printing results to be particularly encouraging. We believe that this is an important step that will help spread the applicability of LIFT-printing for photonic single-crystal devices where edge quality and feature shape are crucial parameters.

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