Growth of Nd:potassium gadolinium tungstate thin-film waveguides by pulsed laser deposition

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Thin Nd-doped potassium gadolinium tungstate [KGW or $KGd(WO_4)_2$] films are grown by pulsed laser deposition by ablation of a stoichiometric monocrystal target. Rutherford backscattering, x-ray diffraction, atomic force microscopy, and waveguide propagation analyses are performed. The as-grown films are optically active, as evidenced by the photoluminescence spectra centered at 1.068 μm. In some of the films, fine photoluminescence spectra between Stark levels are observed. © *2000 American Institute of Physics.* [S0003-6951(00)00518-0]

The development of thin film lasers based on doped planar waveguides presents a very attractive route to the applications in integrated optics in particular, and compact optoelectronic devices in general. Nd-doped potassium gadolinium tungstate [KGW or $KGd(WO_4)_2$] is a relatively new and very promising material with interesting optical properties. α-KGW doped with Nd, Er, or Yb has spectral characteristics which provide an excellent match for diode pumping.[1–6] Nd:KGW additionally offers very efficient and ultralow-threshold stimulated Raman scattering. This in turn allows the simultaneous generation of the common Nd lasing lines together with several Stokes and anti-Stokes lines (at 1.18, 1.32, and 0.97 μm from $^4F_{3/2}\rightarrow{}^4I_{11/2}$ transition), the so-called stringer laser[7] or lasing at 1.35 μm plus the first Stokes line at 1.54 μm from $^4F_{3/2}\rightarrow{}^4I_{13/2}$ transition.[2,5] Moreover, use of various combinations of nonlinear crystals such as KTP, $LiIO_3$, KDP permit frequency doubled output at numerous additional wavelengths in the visible, between 500 and 700 nm. All these combined properties are very useful for communications, integrated optics, laser spectroscopy, ellipsometry, rangefinders, Raman spectroscopy, and so forth. Nd:KGW lasers have demonstrated lower threshold and higher slope efficiency (by more than a factor of 2) than Nd:YAG lasers.[2,4,8] It is possible to operate Nd:KGW lasers in cw and *Q*-switching mode.

Pulsed laser deposition (PLD) has recently produced very promising results in the field of optoelectronics material growth and device construction. Although the production of optical quality thin films and waveguides by PLD presents numerous difficulties and challenges, to date these problems have indeed been successfully overcome, and several high quality active waveguide layers prepared by PLD have been grown, and lasing demonstrated in the lowest-loss examples. Examples include: Er or Yb phosphate glass on Si or fused silica substrates,[9] Nd:YAG on Si, MgO, SGGG, or YAG,[10] $Er:Al_2O_3$ on Si.[11] Nd:GGG on YAG,[12] $Ti:Al_2O_3$ on Al_2O_3 and quartz.[13] An overview on such material growth can be found in Ref. 14.

In this letter, we report the growth of Nd-doped KGW thin films on *r*-cut (1102) sapphire substrates using the PLD technique. Rutherford backscattering (RBS) and x-ray diffraction (XRD) analyses have been made, and waveguide propagation and photoluminescence properties of the layers have been studied. This is the report on thin film growth of this material.

PLD films were grown from crystalline Nd:KGW targets doped with 3 at. % (equivalent to 0.62 wt % $= 1.9\times10^{20}$ Nd^{+3}ions cm^{-3}) using two different experimental setups, both of which used a KrF laser (248 nm, 20 ns, rep rate up to

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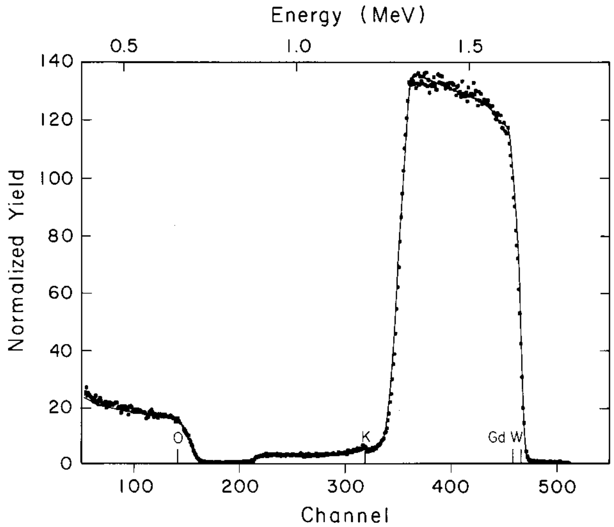

FIG. 1. RBS spectrum of Nd:KGW thin film. The surface channels for different elements are indicated. The deposition conditions were: $P(O_2) = 0.1$ mbar, $T_s = 700\,°C$, $d = 50$ mm, $E = 3$ J cm^{-2}.

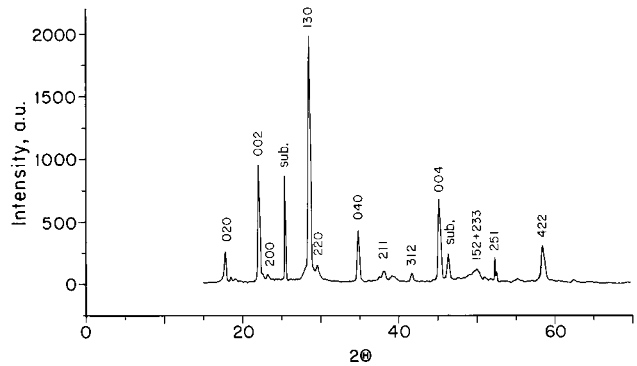

FIG. 2. XRD spectrum of Nd:KGW layer. The deposition conditions were: $P(O_2) = 0.1$ mbar, $T_s = 600\,°C$, $d = 40$ mm, $E = 3$ J cm^{-2}.

15 Hz) at fluences, E, between 2.2–3.2 J cm^{-2}. Substrate temperatures, T_s, were varied between 550 and 800 °C, and were situated at distances, d, of 30–50 mm in front of the target. Substrate heating was performed using a standard resistive heater or laser heated by a cw10.6 μm CO_2 laser.[15] The depositions were carried out in vacuum or in an oxygen environment (base pressure lower than 4×10^{-5} mbar) at pressures, $P(O_2)$, between 0.05 and 0.2 mbar. The average deposition rate, depending on the conditions, was between 0.15 and 0.60 nm per second for a laser repetition rates of 15 Hz.

The chemical composition and thickness of the films were evaluated by RBS. A typical spectrum of KGW thin film is illustrated in Fig. 1. The thickness of the film in this case was 0.7 μm. From the deviation of the points in the plateau in Fig. 1, it is evident that the surface of the film was not smooth. Atomic force microscopy (AFM) measurements have additionally confirmed the presence of particulates characteristic of the PLD process. The general conclusion from the RBS analyses was that for all the deposition conditions used, apart from depositions under vacuum, there is a faithful stoichiometric transfer of the oxygen content from target into the thin film. In some films, however, slight deficient of potassium content have been observed (0.90–0.95 stoichiometric transfer obtained), while at higher T_s, there is a proportionate increase of gadolinium and tungsten content (Gd: 1.1–1.2; W: 2.15–2.35). This is especially pronounced at substrate temperatures higher than 700 °C, where some phases of $Gd_2(WO_4)_3$ may also be presented, whereby enriching both the overall Gd and W content.[16] The Nd content cannot be accurately determined via RBS, due to both low percentage concentration, and also the proximity of the Nd signal to that of Gd in the spectrum. Finally, visual observation shows that the optimum films may well be those that match the target color, and which were grown under the conditions $P(O_2) = 0.1$ mbar, $T_s = 700\,°C$, $d = 50$ mm, and $E = 3$ J cm^{-2}.

The adhesion of the films to the sapphire substrates was very good. The crystalline structure of Nd:KGW films was investigated by XRD measurements with a Bragg–Brentano powder diffractometer and Cu $K\alpha$ radiation in the diffraction angle range from 15° to 65° in 2θ. Without post annealing, the films showed a strong textured polycrystalline nature as shown in Fig. 2. Although, we believe that the deposition conditions (E, $P(O_2)$, d, and T_s) are close to the optimum, such single-crystal growth might not be expected in any case, considering the relatively large lattice mismatch between KGW and (1102)-oriented sapphire. For laser applications, the usual orientation used for laser rods in α-KGW is (010) which has a lattice parameter in this direction $b = 10.38$ A,[17,18] which is appreciably bigger than the $a = 4.76$ A lattice parameter for sapphire. For our films grown by PLD, we have a mixture of low temperature phase of KGW (β-KGW), with high temperature modified form of α-KGW. Annealing in an Ar atmosphere at a temperature of between 1005 and 1075 °C should ensure the presence of α-KGW only.

Waveguide characterization was evaluated using a commercial instrument, with the Metricon model 2010 Prism coupler. Between 1 and 3 TM modes were obtained in the growth films, depending on the actual film thickness. However, the dips in the observed mode spectrum are shallow and wide, which would suggest polycrystalline material, rather than aligned epitaxial monocrystalline growth. A further complication with KGW concerns its biaxial material properties, as mode spectra will be particularly sensitive to nonuniform film growth and consequent refractive index nonuniformity. Thus, we are limited in our further discussions of waveguiding and must restrict our conclusions to ''local'' value of the refractive index. The refractive indices for KGW are $n_x = 2.00$, $n_y = 1.95$, $n_z = 2.05$ at a wavelength of 633 nm.[7] Waveguide propagation analysis will therefore return a mean value for local refractive index, and hence our value of about 1.90 fits quite well with such expected measured values. As shown in the XRD spectrum of Fig. 2, the film has little preferential orientation, and hence, the result of waveguide analysis is entirely consistent with this fact. On the other hand, however, the absence of any observed TE modes does perhaps suggest some inherent anisotropic growth behavior. Further growth runs with systematic post annealing would be necessary to look at this aspect further. Additionally, as seen in AFM scans, there are about μm sized particles present on the film surface, at measured up to 8×10^3 particles per mm^2, which further complicates effi-

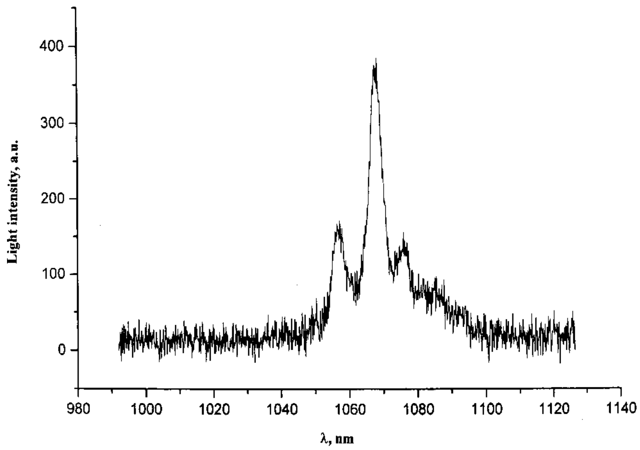

FIG. 3. Nd:KGW waveguide fluorescence spectrum over the spectral region of 1000–1120 nm. The deposition conditions were: $P(O_2)=0.2$ mbar, $T_s=800\,°C$, $d=50$ mm, $E=3.2\,J\,cm^{-2}$.

cient and reliable prism coupling. If the measurements are taken to reduce both particulate size and number density by using synchronized shutters, or rotating diaphragms, then the particulate size can be reduced to 0.2–0.7 μm in our experience, and this would greatly improve further systematic prism coupling measurements.

Finally, photoluminescence measurements have been performed at room temperature using an OMA 2000 Digital Triple Grating Spectrograph (EG&G 1235). A Ti:sapphire laser was used as a pumping source and efficient pumping wavelengths were found to be $\lambda_1=808$ nm, $\lambda_2=804$ nm, and $\lambda_3=751$ nm, with λ_1 twice as efficient as λ_3. The fluorescence spectrum of one of the Nd:KGW thin films is shown in Fig. 3. As can be seen, there is evidence of broadening of the emission peaks, which are assigned as the main laser transition ($^4F_{3/2}\rightarrow{}^4I_{11/2}$) of Nd:KGW, centered at $\lambda=1068$ nm, with two Stark-level satellites at $\lambda=1058$ nm ($2\rightarrow2$ transition) and $\lambda=1075$ nm ($1\rightarrow3$ transition), respectively. The broadening of peaks at full width at half maximum (FWHM) is evaluated to be $\pm2{,}1$ nm. This broadening of the main Nd line will reduce the peak emission cross section and is a result of the imperfect quality of the films. However, no special post-annealing step has yet been applied to the films, and our experience with other PLD grown lasing media, this invariably improves the spectral emission properties.[12]

In conclusion, thin films of Nd:KGW have successfully been grown via PLD and their waveguiding and fluorescence properties investigated. The films were grown on (1102) sapphire at substrate temperatures of between 550 and 800 °C from crystalline targets of KGW doped with 3 at. % Nd. The stoichiometry of the films shows slight potassium deficiency and a concomitant slight excess of Gd and W for higher growth temperatures. The films are polycrystalline with different orientation of the monocrystalline components. The photoluminescence spectra shows peaks centered at $\lambda=1068$ nm with two Stark shifted satellites. Without any post annealing, this emission have noticeably spectral broadening. Future experiments involve improvement of the deposition conditions and possible alternative to sapphire as substrate material. Once loss figures are obtained for these guides, lasing will be attempted.

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