Upconverting Tm-doped BaYYbF₈ optical waveguides epitaxially grown on GaAs

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TM-doped BaYYbF₈ films were epitaxially grown on both (100) and (111) GaAs substrates using CaF₂ or LiF as intermediate layers. The BaYYbF₈ phase was found to be a previously unreported cubic phase with a lattice constant of 0.5711 nm, which is different from a monoclinic phase reported for bulk crystals. The films produced UV and visible radiation with wavelengths at 360 nm (UV), 450–480 nm (blue), and 500–550 nm (green) when pumped by laser radiation at 647 nm.© 1995 American Institute of Physics.

Upconversion lasers represent a class of optically pumped lasers that oscillate at frequencies higher than those used for pumping. The upconversion lasers produce output at wavelengths from violet to red spectral regions using infrared pump wavelengths accessible with III–V semiconductor lasers. These compact light sources are important in optical data storage, high-speed laser printers, large-screen displays, and undersea communications. Materials commonly used for upconversion lasers include rare-earth metal-doped BaY₂F₄, CaF₂, or LiF as intermediate layers. The BaYYbF₈ phase was found to be a previously unreported cubic phase with a lattice constant of 0.5711 nm, which is different from a monoclinic phase reported for bulk crystals. The films produced UV and visible radiation with wavelengths at 360 nm (UV), 450–480 nm (blue), and 500–550 nm (green) when pumped by laser radiation at 647 nm.© 1995 American Institute of Physics.

There are significant obstacles for the growth of epitaxial films of BaYYbF₈ on GaAs in waveguide form, because of their signals overlap on the spectrum. The successful deposition of Tm-doped BaYYbF₈ films was about 300–500 nm. The source used for BaYYbF₈ was prepared from powders using electronic grade BaF₂, YF₃, YbF₃, and TmF₃ having a mole ratio of 1:1:1:0.01. The mixed powders were pelletized under a pressure of 5000 psi. BaYYbF₈ films were deposited on CaF₂ or LiF-covered GaAs by e-beam evaporation. The deposition was carried out at a rate of 0.1–0.4 nm/s at temperatures of 400–550 °C, and the thickness of the BaYYbF₈ films was about 300–500 nm.

The composition of the resulting fluoride films was determined with Rutherford backscattering spectrometry (RBS) and the structural properties were characterized by x-ray diffraction (XRD) and ion channeling.

RBS measurements showed the evaporated fluoride film on GaAs having nominal composition BaY(Yb+Tm)₀.₉₅F₈. To determine the ratio of Tm to Yb it is not possible because their signals overlap on the spectrum. The successful deposition of the fluoride films using a single electron beam is attributed to the similar vapor pressures of the constituent fluorides.

The standard θ-2θ x-ray diffraction pattern in Fig. 1, obtained from a sample comprised of (100) GaAs/ CaF₂/BaYYbF₈, reveals two diffraction peaks centered at 2θ = 31.3° and 65.2° due to the BaYYbF₈ phase, in addition to the peaks corresponding to GaAs and (100)-oriented CaF₂. The two BaYYbF₈ peaks can be indexed as the (220) and (440) reflections of the well-known monoclinic phase, however, these peaks were not observed to be present for
powders. They can also be assigned as the (200) and (400) reflections for a cubic phase with a lattice constant of 0.574 nm. The full width of the rocking curves at half maximum (FWHM) was determined to be about 0.2° for GaAs, 0.5° for CaF$_2$, and 0.9° for BaYYbF$_8$. The highly oriented BaYYbF$_8$ film was further verified by ion channeling measurements. Figure 2 shows backscattering spectra with He ions at both a random and a $[100]$-oriented incidence. No interactions between GaAs and fluorides were revealed. The minimum channeling yield, which is defined as the ratio of the aligned yield to the random yield, was determined to be about 0.24. This value is substantially lower than one, indicating good crystal quality.

A highly oriented BaYYbF$_8$ film has also been grown on a (111) GaAs substrate with a (111)-oriented CaF$_2$ buffer layer. X-ray analysis showed two peaks centered at 2$\theta$ =27.0° and 55.5° other than the (111) reflections of GaAs and CaF$_2$. Neither of the two peaks can be assigned to the monoclinic phase, however, they can be readily identified as the (111) and (222) reflections of the cubic phase.

In-plane alignment of the BaYYbF$_8$ films grown on GaAs was determined by x-ray pole figure analysis. In the analysis, the fluoride phase was first assumed to be cubic with $a=0.574$ nm, and the assumption was found to be consistent with the following findings. The pole figure of Fig. 3(a), taken from the (220) peak of the BaYYbF$_8$ overlayer grown on (100) GaAs, exhibits four spotlike pole densities, suggesting that the BaYYbF$_8$ film is (100) planar oriented with a fourfold rotation symmetry. The (220) pole figures of BaYYbF$_8$, CaF$_2$, and GaAs were found to be aligned with the same in-plane orientation, indicating that the BaYYbF$_8$ film is epitaxial. The pole figure of Fig. 3(b), taken from the (100) peak of the BaYYbF$_8$ overlayer grown on (111) GaAs, exhibits six pole figure densities with almost equal intensity. By comparing the pole figure in Fig. 3(b) and that of the underlying CaF$_2$ and GaAs, one can find a (111) planar oriented BaYYbF$_8$ film grown epitaxially on (111) GaAs. The pole figure of Fig. 3(b) also reveals the presence of high twin density in the epitaxial film BaYYbF$_8$.

The presence of a cubic phase BaYYbF$_8$ was further verified by unit cell evaluation. Under the same deposition conditions mentioned previously, a BaYYbF$_8$ polycrystalline film was prepared on a Si wafer capped with a thermally grown SiO$_2$ layer. X-ray diffraction analysis showed more than twenty peaks, and all of them can be indexed based on a cubic unit cell with a lattice constant $a=0.5711$ nm. The intensities of the diffraction peaks suggest that this previously unreported phase is a defective face-centered cubic structure, since some very weak primitive cubic reflections are observed in the diffraction pattern. The difference in the lattice constant between BaYYbF$_8$ deposited on GaAs/CaF$_2$
and that on Si/SiO$_2$ suggests that one or both films may be strained. The presence of this cubic structure opens up the opportunity of growing BaY$_2$Yb$_2$F$_{8}$ epitaxial films on GaAs.

The investigation of phase stability and phase transformation of the two crystal structures is in progress.

Epitaxial growth of BaY$_2$Yb$_2$F$_{8}$ film was also achieved on (100) GaAs when using a LiF buffer layer, but it failed on (111) GaAs because the interposed LiF layer exhibited a mixed orientation. Since the refractive index of BaY$_2$Yb$_2$F$_{8}$ is substantially higher than that of CaF$_2$ and LiF, one can expect that a waveguide would be formed if the cladding layer is sufficiently thick.

The spectral response of the Tm-doped BaY$_2$Yb$_2$F$_{8}$ films was assessed by focusing radiation from a laser beam at 647 nm. Figure 4 shows a spectrum of a Tm-doped BaY$_2$Yb$_2$F$_{8}$ film grown on (100) GaAs with a LiF buffer layer. Similar results were obtained with a CaF$_2$ interposed layer, although the intensity was lower than that shown in Fig. 4. The films produced UV and visible radiation with wavelengths at 360 nm (UV), 450–480 nm (blue), and 500–550 nm (green). The presence of green upconverted luminescence can be clearly seen in the insert of Fig. 4. Chwalek and Paz-Pujalt$^5$ reported that the linewidths of upconversion excitation from Tm-doped BaY$_2$Yb$_2$F$_{8}$ were much broader in an amorphous film grown on a fused silica substrate than that in a bulk single crystal. The broader upconverting luminescence observed in Fig. 4 from epitaxial films is presumably due to high defect densities as suggested by x-ray analysis and/or nonstoichiometric composition revealed by RBS measurements.

In summary, Tm-doped BaY$_2$Yb$_2$F$_{8}$ films were grown epitaxially on both (100) and (111) GaAs with CaF$_2$ or LiF as buffers, providing a desirable structure for monolithic integration of upconverting waveguides and laser diodes. X-ray diffraction indicated that the BaY$_2$Yb$_2$F$_{8}$ films have a cubic structure with a lattice constant of 0.5711 nm, which is different from the monoclinic structure reported for bulk crystals. To our knowledge, this letter represents the first report on epitaxial growth of BaY$_2$Yb$_2$F$_{8}$ on GaAs and the presence of an alternative cubic structure of BaY$_2$Yb$_2$F$_{8}$.

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