Heteroepitaxy of patterned ternary nonlinear optical materials for frequency conversion of laser sources in the mid and longwave IR

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Compact, high-power, tunable laser sources for the mid and longwave infrared (MLWIR) region are in great demand for a wide variety of applications such as IR countermeasures, enhanced laser radar, long-range IR communications, remote sensing of chemical and biological species, breath analysis, ultrafast spectroscopy of chemical reaction dynamics, etc. However, since the available direct sources in these spectral ranges do not satisfy requirements for power and tunability, up and down frequency conversion via phase or quasi-phase matching (QPM) is exploited. The QPM approach that consists of fabrication of orientation-patterned (OP) templates (thin periodic structures with alternating crystal polarity) and a subsequent thick growth on them, so far has resulted in frequency conversion demonstration in OP-GaAs [1] and OP-GaP [2] only. However, even these two leading materials suffer from shortcomings such as the large two-photon absorption (2PA) of GaAs that does not allow pumping with readily available sources between 1–1.7 µm, and the low crystal quality of the commercial GaP wafers, which results in low OP-GaP template quality and, from here, poor growth on them. In this work we propose resolving current material limitations by combining materials through heteroepitaxy and forming ternaries. After proving the heteroepitaxial concept in the GaP/GaAs case [3,4], nearly 800 µm thick high surface and crystalline quality (determined by XRD, PL, EDS, etc.) $GaAs_xP_{1-x}$ with composition in the range of x = 0.32–0.93 was grown by Low-Pressure Hydride Vapor Phase Epitaxy (LP-HVPE) on plain (001) GaAs substrates achieving composition variation of less than 1% along the layer thickness. Then, up to 600 µm thick OP-GaAsP QPM structures with excellent domain fidelity were also realized on OP-GaAs templates prepared by the MBE assisted polarity inversion technique consisting of patterns with domain widths between 20–62 µm. The growth on plain substrates demonstrated that at certain compositions $GaAs_xP_{1-x}$ exhibits lower 2PA than GaAs but higher nonlinear susceptibility than GaP, and intermediate IR-transparency and thermal properties. In addition, the dispersion of the refractive index of GaAsP allows pumping with shorter wavelengths throughout patterns with wider domains, which HVPE growth is more controllable. Consequently, the growth of OP-GaAsP/OP-GaAs provided large enough optical aperture for realizing femtosecond pulse SHG at 5450 nm with 19.1% internal conversion efficiency. Down conversion in OP-GaAsP in an Optical Parametric Oscillator (OPO) setup is also in progress. While forming ternaries improves desired material properties, their more favorable heteroepitaxial growth results in device quality material feasible for frequency conversion in the MLWIR region.

References

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