Layer Growth of high-quality $BaSO_4:Mn^{6+}$ using Liquid Phase Epitaxy

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Single-crystalline host materials doped with transition-metal ions are of high interest for applications as tunable lasers. Mn^{6+} ions exhibit broadband luminescence, however, Mn^{6+}-doped crystals or waveguide structures could as yet not be grown in sufficient quality. The active material has to be free of inclusions or defects larger than $\lambda/10$, with λ, the wavelength of the porpagating beam. The interface between active layer and substrate must be optically flat to receive low-loss guiding properties. Finally, in the case of homo-epitaxy of $BaSO_4$, the doped layer has to be arranged on the substrate (001) direction.

The growth temperature of $BaSO_4:Mn^{6+}$ is limited by the decomposition of $BaSO_4$ at 1590°C, its phase transition above 1010°C, and especially the chemical reduction of the manganese dopant from Mn^{6+} to Mn^{5+} above 620°C. Therefore, the growth of $BaSO_4:Mn^{6+}$ from a solution at lower temperatures is the most suitable method. Liquid phase growth is close to the thermodynamic equilibrium and has enabled us to grow high-quality layers.

First, we prepared undoped $BaSO_4$ crystals of 10 x 5 x 1 mm^3 in a, b, and c-direction, respectively, using the flux method with LiCl as solvent. Subsequently, growth of high-quality undoped $BaSO_4$ was performed by liquid phase epitaxy (LPE), using the additive ternary CsCl-KCl-NaCl solution. We obtained crystalline layers free of inclusions, grown in the Frank-Van der Merwe mode (layer-by-layer growth). Finally, layers of $BaSO_4:Mn^{6+}$ were fabricated with thicknesses up to 150 μm, at growth rates of 3 μm/h and temperatures of 500–580°C. The thickness was controllable with a precision of ±0.1 μm. The Mn^{6+} concentration in the doped layer was up to 1 mol.% with respect to S^{6+}.

In collaboration with the University of Hamburg, absorption and emission spectra were measured, which confirmed that the manganese ion was incorporated in the layer solely in its sextavalent oxidation state. Room-temperature luminescence in the wavelength range 850-1600 nm was observed.