

Homoepitaxial Growth of High-Quality BaSO₄:Mn⁶⁺ Using Low-Temperature Liquid Phase Epitaxy

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Single-crystalline host materials doped with 3d¹ transition-metal ions are of high interest for applications as tunable lasers. The Mn⁶⁺ ion exhibits broadband luminescence, however, Mn⁶⁺-doped crystals or waveguide structures could as yet not be grown with sufficient optical quality. The active material has to be free of defects and inclusions larger than 1/20 of the optical wavelength. In addition, the surface of the active layer and its interface to the substrate must be optically flat to receive low-loss guiding properties.

The structure of the barite-phase BaSO₄ contains SO₄²⁻ tetrahedra which are replaced partly by the MnO₄²⁻ dopant complexes. The BaSO₄:Mn⁶⁺ growth temperature is limited by the phase transition above 1010°C and especially the noticeable reduction of Mn⁶⁺ to Mn⁵⁺/Mn⁴⁺ above 600°C. Therefore, the growth of BaSO₄:Mn⁶⁺ 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 grew undoped BaSO₄ substrate crystals of 10 x 5 x 4 mm³ in a, b, and c-direction, respectively, using the flux method with LiCl as the solvent. Subsequently, the growth of high-quality undoped BaSO₄ was performed by liquid phase epitaxy (LPE), using the additive ternary CsCl-KCl-NaCl solution. We obtained flat layers free of inclusions with step heights of 1.4 nm, equal to 2 unit cells, and step distances of about 200 nm. Finally, layers of BaSO₄:Mn⁶⁺ were grown on c-oriented faces with thicknesses up to 150 μm, at growth rates of 3 μm/h and growth temperatures of 500–550°C. The Mn⁶⁺ concentration in the doped layer was up to 1 mol% with respect to S⁶⁺.

Absorption and emission spectra were measured, which confirmed that the manganese ion was incorporated in the layer solely in its hexavalent oxidation state. Room-temperature broadband luminescence in the wavelength range 850-1600 nm was observed.