Single-Crystal Growth for Substrates. Experimental Details.

We used the Top Seeded Solution Growth (TSSG) method to grow single-crystals, which will be used for epitaxial substrates. The presence of WO$_3$ helps to lower the viscosity of the growth solution. Low viscosity is known to favor crystal growth by allowing the formation of high quality crystals in less time than is possible with self-flux solutions. However, although W$^{6+}$ is incorporated in the crystal structure with a very low distribution coefficient, the presence of W$^{6+}$ in the crystal can modify its properties. The choice of growth solution is therefore a trade-off between the viscosity of the solution and its amount of tungsten. The reagents used were Rb$_2$CO$_3$, (99%), NH$_4$H$_2$PO$_4$ (99%), TiO$_2$ (99.9%) and WO$_3$ (99.9%). The composition used was Rb$_2$O-P$_2$O$_5$-TiO$_2$-WO$_3$ = 44.24-18.96-16.8-20 (mol %), which has already been shown by our group$^9$ to be optimum. The bulk crystals were grown in 125 cm$^3$ cylindrical platinum crucibles, and the solution weight was about 200 g. All experiments were carried out in a vertical tubular furnace. The crucible was placed into the furnace in a region where the axial temperature gradient in the solution was 2.4 K/cm and where the bottom was hotter than the surface. The mixture was homogenized by maintaining the solution at about 75 K above the expected saturation temperature, $T_s$, for about 5-6 h. The $T_s$ was then accurately determined by placing the seed in contact with the center of the solution surface and controlling the growth or dissolution of the crystal seed with a micrometer comparer until neither growth nor dissolution was observed for a long time. Once the $T_s$ was determined, the seed was placed in contact with the surface of the solution and a cooling program of 0.1 K/h was applied to the solution until a final temperature 20-30 K below the $T_s$ (see Table 1) was reached. In several experiments, to obtain large crystals we pulled them at a rate of 0.5 mm every 12 h starting from a temperature of around 10 K below the $T_s$ or lower. In all experiments, the rotation was 40 rpm. All the crystals were grown on $c$-oriented seeds, where $c$ is the crystallographic direction perpendicular to the surface of the solution. After growth, these single crystals were cut and polished in slices perpendicular to the $c$-crystallographic direction before they were used as substrates.