CRYSTAL GROWTH OF OXIDES AND FLUORIDES AT THE IKZ

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Summary

The growth of volume crystals of oxides and fluorides has a long tradition at the IKZ but is also characterized by modern topics with outstanding international visibility. Our publications on the Czochralski method to grow big gallium oxide (β-Ga_2O_3) bulk crystals from the melt are “highly cited” in the field of bulk crystal growth [1,2]. The former head of the IKZ team, Reinhard Uecker, was awarded the IOCG Frank Prize in 2019 (together with Darrell Schlom) for pioneering "strain engineering" by providing substrates for “lattice mis-matched” films [3,4]. Recently, we succeeded in the growth of the first KTb_3F_{10} bulk crystals that can be used as optical isolators for high-power near infrared lasers.

In the presentation we will show how the preparation of β-Ga_2O_3 substrates of highest structural quality helped to provide a breakthrough in demonstration of novel power electronics devices [5]. Regarding crystal growth, the control of the local oxygen partial pressure is crucial to minimize the formation of suboxides and metallic gallium in the melt that would attack the crucible [2]. The growth of highly n-conductive β-Ga_2O_3 with large diameter remains a challenge, not only due to the self-absorption of heat radiation [1]. Current research is focused on the investigation of the segregation and doping of various elements during growth [6,7]. Also, the preparation and potential applications of gallates with spinel structure such as $ZnGa_2O_4$ [8] are discussed.

Perovskite-type substrates, originally used to prepare superconducting thin films, have been employed to push the limits of novel ferroelectric, superconducting, ferromagnetic, piezoelectric, multiferroic or high-mobility oxide electronic materials [9]. The rare earth scandates $REScO_3$ (RE = Dy...Pr) grown at IKZ are in worldwide use to cover any desired pseudo-cubic lattice parameter in the range from about 3.95 to 4.02 Å [3,4]. Recently, novel promising materials with lattice constants in the range of 4.08–4.15 Å have been developed at IKZ jointly with Cornell University [10,11] to accommodate promising thin film materials such as La:$BaSnO_3$, $BiScO_3$, $BiFeO_3$ or $PbZrO_3$ with high structural quality. This success was strongly based on thermochemical assessments of stability of compounds such as Ba_2ScNbO_6 and $BaSnO_3$ under melt growth conditions and phase diagram calculations in the La_2O_3–Lu_2O_3–Sc_2O_3–Nd_2O_3 system. We will present these results and also highlight perovskite-type crystals $SrHfO_3$ and $SrZrO_3$ grown by the IKZ in collaboration with the Institute of Physics CAS in Prague [12].

Regarding fluorides, we will briefly introduce our activities and demonstrate first crystals of KTb_3F_{10} that can be used to prepare superior optical isolators. The crystal growth is impeded by a peritectic phase transition slightly below the melting point, while accurate thermodynamic data is not available [13]. Formation of scattering centers as well as oxygen contamination must be mitigated by employing the right off-stoichiometry and purified starting materials.

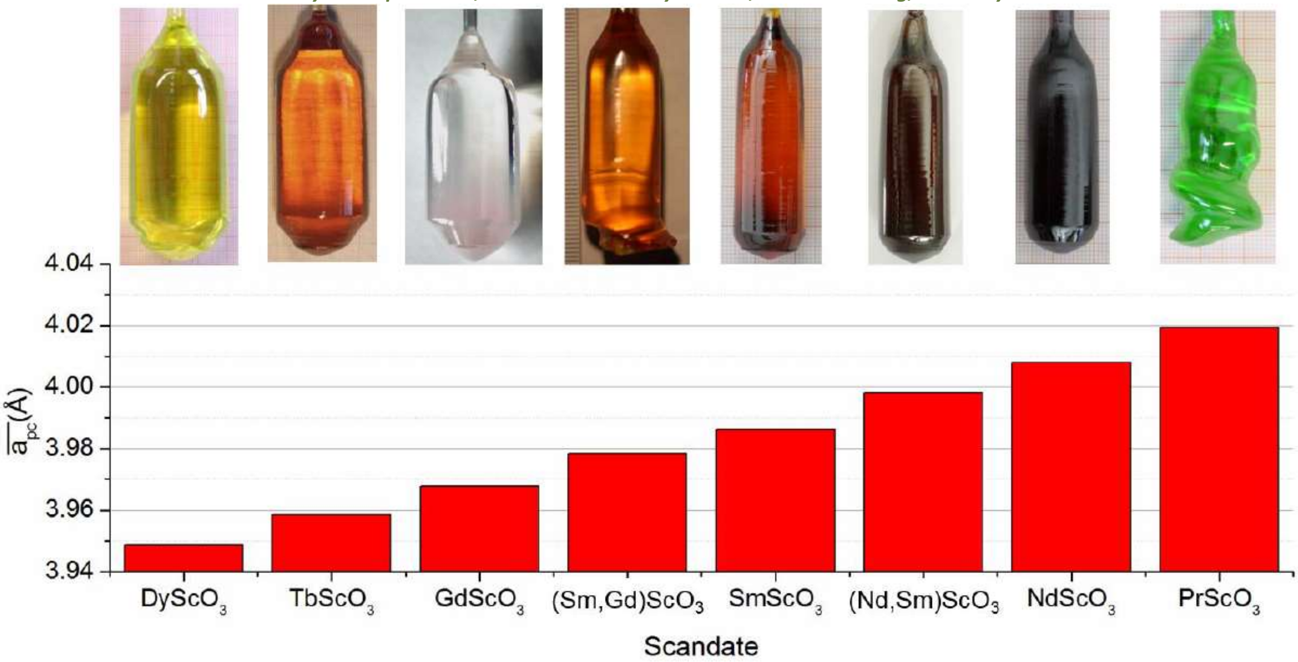

Fig. 1: Pseudo-cubic lattice parameters of different rare earth scandate single crystals (from [4])

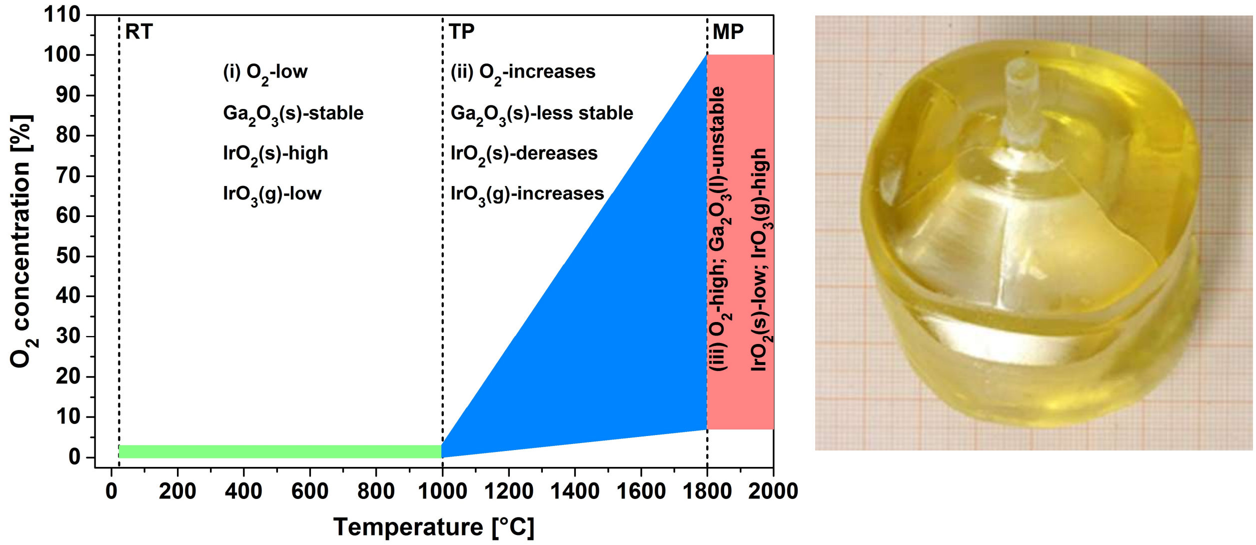

Fig. 2: Oxygen delivery vs. temperature for the growth of large diameter β-Ga2O3 single crystals from an Ir crucible (from [2]); 2-inch Al-doped β-Ga_2O_3 single crystal obtained by the Czochralski method (from [6])

[1] Z. Galazka, K. Irmscher, R. Uecker et al., Journal of Crystal Growth **404** (2014) 184
[2] Z. Galazka, R. Uecker, D. Klimm et al., ECS Journal of Solid State Science and Technology **6** (2017) Q3007
[3] R. Uecker, H. Wilke, D.G. Schlom et al., Journal of Crystal Growth **295** (2006) 84
[4] C. Guguschev, J. Hidde, T.M. Gesing et al., CrystEngComm **20** (2018) 2868-2876
[5] K. Tetzner, E.B. Treidel, O. Hilt et al., IEEE Electron Device Letters **40** (2019) 1503
[6] Z. Galazka, S. Ganschow, A. Fiedler et al., Journal of Crystal Growth 486 (2018) 82-90
[7] Z. Galazka, K. Irmscher, R. Schewski et al., Journal of Crystal Growth **529** (2020) 125297
[8] Z. Galazka, S. Ganschow, R. Schewski et al., Applied Physics Letters Materials **7** (2019) 022512
[9] H.J. Haeni, P. Irving, W. Chang et al., Nature **430** (2004) 758
[10] R. Uecker, R. Bertram, M. Brützam et al., Journal of Crystal Growth **457** (2017) 137–142
[11] C. Guguschev, D. Klimm, M. Brützam et al., Journal of Crystal Growth **528** (2019) 125263
[12] J. Pejchal, C. Guguschev, M. Schulze et al., Optical Materials **98** (2019) 109494
[13] K.T. Stevens, W. Schlichting, G. Foundos et al., Laser Technik Journal (Wiley) 03/2016, p. 18