Preparation of Perovskite, Pyrochlore, and Magnetoplumbite Crystals for Advanced Functional Oxides

Christo Guguschev, Michael Schulze, Julia Köpp, Steffen Ganschow and Matthias Bickermann

Leibniz-Institut für Kristallzüchtung (IKZ), Max-Born-Str. 2, 12489 Berlin, Germany
e-mail: matthias.bickermann@ikz-berlin.de

In this contribution, we will provide details on recent substrate crystals grown by IKZ that aid the development of heteroepitaxial advanced functional oxide layers. One renowned class of materials established by IKZ are the rare-earth scandates $REScO_3$ with RE = Dy to Pr. Their pseudocubic lattice parameters range between 3.95 Å and 4.02 Å and vary in steps of about 0.01 Å as the RE series is traversed. A fine-tuning of the pseudo-cubic lattice parameter between nearly all endmembers is also possible by the growth of solid solutions [1]. $REScO_3$ crystals enabled research on artificially strained oxide films that show interesting ferroelectric, superconducting, ferromagnetic, piezoelectric, multiferroic, or electron transport properties [2].

Recently, perovskite substrates with higher lattice constants up to 4.16 Å are demanded e.g. for thin films of $BiFeO_3$, $EuTiO_3$, $BaSnO_3$ or PZT. We have extended our accessible lattice parameter range with $(La,Nd)(Lu,Sc)O_3$ and $La(Lu,Sc)O_3$ mixed crystals that can be prepared by the Czochralski method in iridium crucibles, albeit at temperatures well above 2100°C [3]. A particular IKZ-Cornell development to provide lattice-matched substrates for $BaSnO_3$ (4.116 Å) was the double perovskite Ba_2ScNbO_6, grown from the melt by a novel crystal growth technique [4].

Other perovskite crystals such as $SrHfO_3$ and $SrZrO_3$ (around 4.10 Å) require temperatures above 2300°C for melting and thus are prepared by crucible-free methods which utilize Xenon lamp optical floating zone furnaces [5]. Doped with rare earth ions these compounds are attractive for scintillator applications, but the growth method leads to a limited achievable diameter and to increased internal stress in the crystals.

Thin films of pyrochlore structure are a matter of research because they show frustrated magnetic and spin effects. Again, substrates with tailored lattice parameters are required to prepare films of high structural quality. As an example, we are researching growth of $Gd_2Ti_2O_7$ substrate crystals by the optical floating zone method. Note that titanates with pyrochlore structure can also be interesting for other applications such as optical isolators. To satisfy the demand for reasonably long bulk crystals of appreciable structural quality, we developed growth of $Tb_2Ti_2O_7$ [6] single crystals with 16 mm x 16 mm cross sections and 54 mm in length using the edge-defined film-fed growth (EFG) technique. First results on high-quality $(Mg,Zr):SrGa_{12}O_{19}$ bulk crystals with magnetoplumbite structure will be also presented, which are grown by the top-seeded solution growth technique (TSSG) [7].

[1] C. Guguschev, J. Hidde, T.M. Gesing, M. Gogolin, D. Klimm, CrystEngComm, 20, **2018**, 2868-2876.

[2] J.H. Haeni, P. Irvin, W. Chang, R. Uecker, P. Reiche, Y.L. Li, S. Choudhury, W. Tian, M.E. Hawley, B. Craigo, A.K. Tagantsev, X.Q. Pan, S.K. Streiffer, L.Q. Chen, S.W. Kirchoefer, J. Levy, D.G. Schlom, Nature, 430, **2004**, 758-761

[3] R. Uecker, R. Bertram, M. Brützam, Z. Galazka, T.M. Gesing, C. Guguschev, D. Klimm, M. Klupsch, A, Kwasniewski, D.G. Schlom, J. Crystal Growth, 457, **2017**, 137-142

[4] C. Guguschev, M. Brützam, D. Schlom, H. Paik, United States Patent Application Publication, **2019**, US20200378030A1.

[5] J. Pejchal, C. Guguschev, M. Schulze, V. Jary, E. Mihokova, K. Rubesova, V. Jakes, J. Barta, M. Nikl, Optical Materials, 98, **2019**, 109494.

[6] D. Klimm, C. Guguschev, D. J. Kok, M. Naumann, L. Ackermann, D. Rytz, M. Peltz, K. Dupré, M.D. Neumann, A. Kwasniewski, D.G. Schlom, M. Bickermann, CrystEngComm, 19, **2017**, 3908-3914.

[7] C. Guguschev, C. Richter, M. Brützam, K. Dadzis, C. Hirschle, T.M. Gesing, M. Schulze, A. Kwasniewski, J. Schreuer, D.G. Schlom, Crystal Growth & Design, **2022**, in press, DOI: 10.1021/acs.cgd.2c00030.

Acknowledgments: The authors thank IKZ colleagues Markus Stypa and Mario Brützam for support and preparation of crystals. Parts of this work were performed in collaboration with, among others, the groups of Darrell G. Schlom and partners at Cornell University (USA), Jan Pejchal (FZU, Prague) and Kateřina Rubešová (UCT, Prague), Thorsten M. Gesing (Universität Bremen), Jürgen Schreuer (Ruhr-Universität Bochum). Recent work on $Tb_2Ti_2O_7$ is funded by the German Ministry of Economic Affairs BMWK as a ZIM project with EOT GmbH.