Rapid growth of deuterated KDP crystals for frequency conversion in the "Laser MegaJoule" facility

The Néel Institute has a well established expertise in crystal growth. Among the many growth techniques used, crystal growth in solution (flux method) at high or low temperatures (close to room temperature) allows synthesis of crystals that undergo a phase transition or decomposition before melting. The major drawback of these techniques, when compared with growths from the melt, is the limited growth rates that can be achieved. As a result, growth runs can extend over several months for very large crystals, hence the interest in developing techniques able to increase growth rates by an order of magnitude. The expertise of the Néel Institute "Matériaux, Optique Nonlinéaire et Plasmonique" team in such techniques is the basis of a joint study involving the CNRS, the CEA and the company St Gobain Crystals in view of the growth of giant crystals for the "Laser Mégajoule" facility.

The "Laser Mégajoule" (LMJ) facility currently being set up near Bordeaux will be dedicated to the production and the study of high energy plasmas. Each one of the 240 laser lines constituting the LMJ facility includes three KH_2PO_4 (KDP) single crystal slabs of 40x40 cm² in aperture: one for the Pockels cell and two for the frequency conversion (IR-visible-UV) and focusing system. Such large aperture slabs require growing very large KDP boules (about 300kg).

So far, these giant crystals have been synthesized by lowering the temperature of a 1000 litre solution containing a KDP point seed. To reduce the growth time growth rates are increased by one order of magnitude through a technique based on the continuous ultra-filtration and thermal conditioning of the solution. This technique allows reaching very high supersaturations, to go far from thermodynamic equilibrium in the metastable region without spurious nucleation. The French company St Gobain Crystals uses this technique to produce all the giant KDP crystals needed for the LMJ. However, these crystals can not be used for the visible-UV convertors because of a residual absorption which lowers their laser damage threshold so much that the optical components would not resist. To avoid the Raman effect responsible of this absorption, partially deuterated KDP crystals ($K(D_{(1-x)}H_x)_2PO_4$) have to be used.

The LMJ requires single crystal slabs with a deuterium ratio above 60%. To grow this intermediate composition solid solution rapidly, the temperature lowering technique used for KDP is inapplicable. In fact, the continuous modification of the thermodynamic conditions of growth (temperature and supersaturation) leads to modifications in the growth mechanisms: diffusion to and on the growing surfaces, adsorption... There are consequences at the chemical, morphological and mechanical levels leading to a significant lowering of the quality of the grown crystals.

The Néel Institute team has developed an original technique that allows us to achieve rapid growths in stationary thermodynamic conditions. Done at constant temperature and supersaturation, the growth uses a transport technique where the solution is circulated between two different containers of the reactor. In the first one, at higher temperatures, a high solute concentration is maintained by the dissolution of a deuterated KDP powder whose synthesis had first been optimized.

This solution is then transported into another container (the growth zone) kept at a lower temperature containing a KDP point seed. The temperature is chosen so that the rich solution arriving becomes heavily supersaturated hence leading to high growth rates. To avoid spurious nucleation, the solution is treated in-line while it passes from the enrichment zone to the growth zone. Because the time for treatment is reduced, the treatment must be highly efficient. That is why ultra-filtration and thermal conditioning are supplemented by high power ultrasounds which efficiently disaggregate clusters of molecules that are the starting point of the spurious nucleation. The combined effect of solution treatment and high supersaturations allows us to reach growth rates over 10 mm/day instead of the fractions of a mm per day rates that are generally observed with standard solution growth techniques.

Crystals of Deuterated KDP of size several cm³ obtained by rapid growth in solution under stationary conditions.

This technique has been successfully applied for the growth of organic-inorganic hybrid crystals as well as KDP. The optimal rapid growth conditions for deuterated KDP (DKDP) are being investigated in partnership with the CEA which is in charge of building and operating the LMJ project.

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FURTHER READING

MATÉRIAUX, OPTIQUE NONLINÉAIRE ET PLASMONIQUE.
J. Zaccaro, A. Ibanez, brevet CNRS Ref. PCT/FR2007/00198