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Lead free KTN single crystals: from composition regulation, space charge field engineering to application

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Lead-free crystals, as a classic example of environmentally-friendly materials, have received the extensive concern and key development recently at the forefront of high-technology advanced materials. Among these lead-free candidates, $KTa_{1-x}Nb_xO_3$ (KTN) is the most potential systems due to their giant electro-optic, piezoelectric and electrocaloric properties. However, large-size and high quality KTN crystals are difficult to grow, which limits its development and application. In this work, we successfully grew a series of large-size and high quality pure and Fe doped KTN crystals by the top-seeded solution growth method. The as-grown KTN crystals exhibit good crystallinities and have large quadratic electro-optic coefficients ($s_{11} = 1.04 \times 10^{-14}$ m^2V^{-2}), indicating their quality and suitability for device application. We investigate the segregation of as-grown crystals which enables precise control of the individual components of crystals during growth, and propose effective energy models to explain the crystal morphology and guide the size uniformity crystal growth which ensures the quality of as-grown crystals. Component regulation and space charge field engineering are used to improve the optical and ferroelectric properties. We successfully utilize the composition gradient, light-induced space charge field and external electric field to achieve the micro-domains manipulations in KTN, which greatly enhance the crystal performances. Further, the variable gradient functional materials is proposed by the light induced space charge field engineering. Based on this variable gradient refractive index KTN, a MHz high-speed electro-deflector was produced. The controllable growth of high quality KTN crystals will be valuable for the lead-free crystal performance improvement and photonic and electronic device development, and provides a basis for exploring the origin of giant electro-optic, piezoelectric effects.