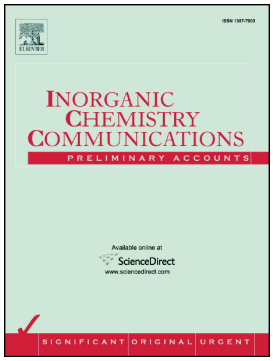

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Structurally Stable Borate as a UV Nonlinear Optical Material

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Abstract

A new phase-matching borate $K_3Sr_3Li_2Al_4B_6O_{20}F$ features double-layer structure similar to that of the outstanding ultraviolet nonlinear optical material $Sr_2Be_2B_2O_7$, and thus inherits its excellent linear and nonlinear optical properties in some degree. Furthermore, it overcomes the structurally dynamical instability problem existing in $Sr_2Be_2B_2O_7$. The first-principles calculations and anionic group theory are employed to explain these properties.

Keywords: Nonlinear optics, crystal structure, structure-property relationship, borate

Ultraviolet (UV, wavelengths below 400 nm) nonlinear optical (NLO) materials, which can double the frequency of incident light to UV region, are essential for a number of advanced scientific instruments [1-5]. Such materials should be of high second-harmonic generation (SHG) efficiency, short cutoff edge (absorption edge), moderate birefringence to achieve phase-matching, etc [6, 7]. Limited by these fundamental requirements, to date only one NLO material $KBe_2BO_3F_2$ (KBBF) [8] is able to directly generate UV coherent light even lower than 200 nm in practice. The required optical properties in KBBF are mainly ascribed to the layered structural units (namely, $[Be_2BO_3F_2]_\infty$ single-layers perpendicular to the crystallographic *c* axis), which afford the NLO-active $[BO_3]^{3-}$ units in perfectly coplanar and aligned arrangement [8, 9]. However, KBBF contains highly toxic beryllium and suffers from a severe layering growth habit owing to the weak bonding between the layered structural units. These problems hinder the commercial applications of KBBF, and thus it is an urgent need to develop new beryllium-free candidates for KBBF [10-15].

In order to inherit the brilliant optical advantages while overcoming the layering growth habit of KBBF, many attempts have been made to develop the layered NLO materials with reinforced interlayer bonding [10, 15-20]. Especially, $Sr_2Be_2B_2O_7$ (SBBO) was ever considered to be one of the most attractive substitutes for KBBF [7]. Its structure features $[Be_2B_2O_7]_\infty$ double-layers that are interconnected by Sr−O bonds, which enhance the interlayer bonding to some degree. Nevertheless, SBBO suffers from a structure instability problem and as a result its crystal structure has not been well solved yet [21].

On the basis of SBBO, Huang et al developed a new NLO material $NaCaBe_2B_2O_6F$ (with $[Be_3B_3O_6F_3]_\infty$ double-layers), in whose structure the bridged Be−O bonds inside $[Be_2B_2O_7]_\infty$ double-layers of SBBO are replaced by longer Be−F bonds [21]. $NaCaBe_2B_2O_6F$ resolves the

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