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Linear optical properties of L-arginine monohydrobromide monohydrate (LAHBr) single crystals

K. Sangeetha [a], R. Ramesh Babu [a,*], K. Ramamurthi [b]

[a] Crystal Growth and Thin Film Laboratory, Department of Physics, Bharathidasan University, Tiruchirappalli 620 024, Tamil Nadu, India
[b] Department of Physics and Nanotechnology, Faculty of Engineering and Technology, SRM University, Kattankulathur 603 203, Tamil Nadu, India

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ABSTRACT

L-Arginine monohydrobromide monohydrate single crystals have been grown by conventional and unidirectional solution growth methods. The quality of both crystals has been analyzed through high resolution X-ray diffraction, optical transparency and laser damage threshold studies. The orthoscopic pattern, three principal refractive indices and birefringence of the crystal at different wavelengths, optical dispersion parameters, crossing angle between the optic axes have also been determined for conventionally grown LAHBr single crystal.

1. Introduction

L-Arginine based crystals are effective nonlinear optical materials with favourable optical, thermal and mechanical properties. Their optical coefficients, laser damage threshold, transparency range and angular sensitivity are superior to potassium dihydrogen phosphate (KDP) crystal [1]. L-Arginine based compounds contain an optically active carbon atom and therefore most of them form acentric crystals. L-Arginine monohydrobromide monohydrate (LAHBr) is an L-arginine based nonlinear optical crystal first reported by Monaco et al. [2], since then some authors have studied their physical properties. Haussühl et al. [3] have studied the elastic, pyroelectric, electrostrictive and electro-optic properties including dielectric, optical and thermal expansion of LAHBr single crystal. Mukerji et al. [1,4–6] extended the studies on spectral, mechanical and micro-morphological properties of LAHBr single crystal. Thus, from the previous reports, LAHBr single crystal was known to be an electro-optical and nonlinear optical crystal. In this paper, LAHBr single crystal was grown by conventional and unidirectional solution growth methods. When compared to conventional solution growth methods (slow evaporation and slow cooling) unidirectional solution growth method is the best in which any one direction of the crystal can be grown i.e., growth along any significant direction such as the direction of optic plane, phase matching direction, direction of high mechanical stability, piezoelectric or dielectric constant is possible [7]. Hence in this work, the quality of crystals in both methods has been compared using high resolution X-ray diffraction, optical transparency and laser damage threshold studies. Also, the important optical studies such as orthoscopic pattern, three principal refractive indices, birefringence, optical dispersion parameters and crossing angle for different wavelengths were measured for conventionally grown crystal. These optical properties will be very useful to find the phase matching angle of the crystal and also when the crystal is used in laser pulse resonators and optical parametric oscillations. Therefore, the aim of the present work is two dimensional, viz., the discussion about the uniqueness of unidirectional growth and the determination of important optical parameters of LAHBr single crystal.

2. Experimental details

2.1. Growth of LAHBr single crystals

2.1.1. Slow evaporation method

L-Arginine monohydrobromide was synthesized by reacting L-arginine and Hydrobromic acid in 1:2 molar ratio at room temperature (28 °C). Single crystals of LAHBr were grown by slow solvent evaporation method (conventional solution growth method) at room temperature. Fig. 1(a) shows the conventionally grown LAHBr single crystal of dimension $47 \times 40 \times 8\ mm^3$.

* Corresponding author. Tel.: +91 431 2407057; fax: +91 431 2407045.
E-mail address: rampap2k@yahoo.co.in (R.R. Babu).

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Fig. 1. (a) Conventionally grown LAHBr single crystal (b) Unidirectionally (0 0 1) plane) grown LAHBr single crystal with ampoule and (c) cut and polished LAHBr single crystal.

2.1.2. Unidirectional solution growth method

⟨0 0 1⟩ Direction of conventionally grown LAHBr crystal was cut and polished and used as seed crystal for unidirectional growth. The experimental setup for unidirectional solution growth was followed as discussed in our previous report [8]. The seed crystal was loaded at the bottom of the ampoule and filled with saturated solution of LAHBr. The ampoule was placed inside the Constant Temperature Bath (CTB) in such a way that top portion of the ampoule experiences the room temperature (28 °C) and bottom portion experiences the temperature of CTB. The cryostat with temperature accuracy ±0.01 °C attached with CTB cools down (0.5 °C/day) the bottom portion to 22 °C which is the optimized temperature for obtaining optical quality LAHBr crystal. Fig. 1(b) shows LAHBr single crystal (12 mm in diameter and 60 mm in length) grown in glass ampoule by unidirectional solution growth method. Fig. 1(c) shows the cut and polished unidirectionally grown crystal.

2.2. Characterizations

The crystalline perfection, optical transparency and laser damage threshold were studied for conventionally grown LAHBr single crystal and compared with unidirectionally grown LAHBr. High resolution X-ray diffraction (HRXRD) determines the crystalline perfection by employing a multicrystal X-ray diffractometer developed at National Physical Laboratory, New Delhi [9]. The well-collimated and monochromated MoKα_1 beam obtained from the three monochromator Si crystals set in dispersive (+,−,−) configuration has been used as the exploring X-ray beam. The specimen crystal is aligned in the (+,−,−,+) configuration. Due to dispersive configuration, though the lattice constant of the monochromator crystal(s) and the specimen are different, the unwanted dispersion broadening in the diffraction curve of the specimen crystal is insignificant. The specimen can be rotated about the vertical axis, which is perpendicular to the plane of diffraction, with minimum angular interval of 0.4 arcs. The rocking or diffraction curves were recorded by changing the glancing angle (angle between the incident X-ray beam and the surface of the specimen) around the Bragg diffraction peak position θ_B (taken as zero for the sake of convenience) starting from a suitable arbitrary glancing angle and ending at a glancing angle after the peak so that all the meaningful scattered intensities on both sides of the peak include in the diffraction curve. Before recording the diffraction curve to remove the non-crystallized solute atoms remained on the surface of the crystal and also to ensure the surface planarity, the specimen was first lapped and chemically etched in a non preferential etchant of water and acetone mixture in 1:2 volume ratio. The optical transparency was analyzed for the grown crystals using UV–visible spectrometer. Laser damage threshold was calculated for the crystals using Nd:YAG laser source of wavelength 1064 nm.

The orthoscopic pattern and the three principal refractive indices were analyzed for conventionally grown LAHBr single crystal. Interference technique was used to observe the orthoscopic pattern and He-Ne laser source of wavelength 632.8 nm was used for the purpose. The three principal refractive indices (n_x, n_y and n_z) of biaxial LAHBr single crystal were measured for different wavelengths by prism method. In this method, a beam of white light is allowed to incident on the reflecting surface of the prism shaped crystal that disperses the white light into colours. The obtained spectrum is set in minimum deviation position and the refractive index is calculated using angle of prism and angle of minimum deviation. Birefringence ($n_z - n_x$) and angle between the optic axes of LAHBr single crystal are also measured from the refractive index data.

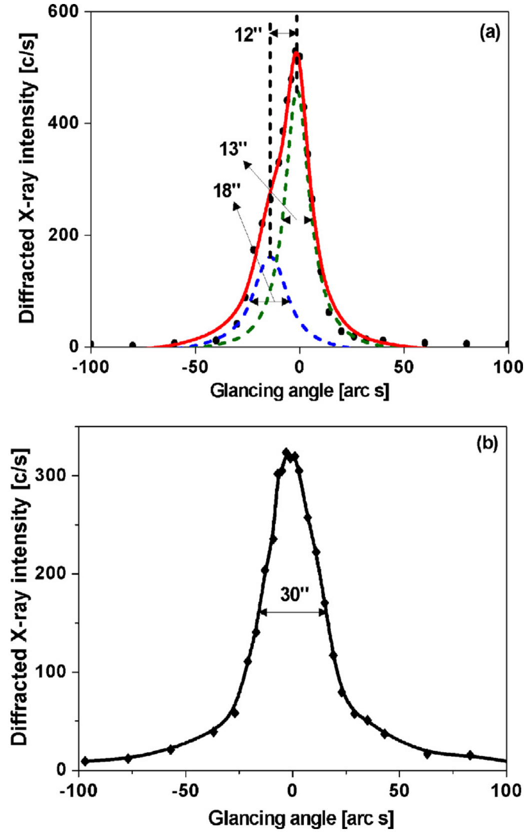

Fig. 2. HRXRD curve for (a) conventionally and (b) unidirectionally grown LAHBr crystal along (0 0 4) planes.

3. Results and discussion

3.1. Crystalline perfection

Fig. 2(a) and (b) shows the high-resolution diffraction curve (DC) recorded for conventionally and unidirectionally grown LAHBr single crystals, respectively, using (0 0 4) diffracting planes in symmetrical Bragg geometry by employing the multicrystal X-ray diffractometer with MoKα_1 radiation. The conventionally grown crystal contains one additional peak (Fig. 2(a)) which is 12 arcs from its adjoining region and it is a very low angle boundary. The FWHM (full width at half maximum) of the main peak and the very low angle boundary are, respectively, 13 and 18 arcs. The entrapment of impurities or solvent molecules and also the thermal fluctuations during the growth process could be responsible for the formation of the grain boundaries. The DC of unidirectionally grown LAHBr crystal shows a single peak indicating that the specimen is free from structural grain boundaries. The FWHM (full width at half maximum) of the curve is 30 arcs which is somewhat more than that expected from the plane wave theory of dynamical X-ray diffraction [10] for an ideally perfect crystal but close to that expected for perfect real crystals. The broadness of DC with scattered intensity along