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Refractive index, birefringence, third-order non-linearity and piezoelectric resonance studies of L-lysine monohydrochloride dihydrate single crystals

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ABSTRACT

The principal refractive indices of L-lysine monohydrochloride dihydrate (L-LMHCl) single crystal for different wavelengths were measured by minimum deviation method at room temperature. The experimental values of refractive indices fit well with the theoretical Cauchy's equations. The birefringence and the crossing angle between the optical axes were calculated. The parameters of Sellmeier's single term dispersion equation were determined by least square method. A simple interferometric technique was used to observe the interference patterns along the optic plane and to qualitatively analyze the optical homogeneity of the grown crystal. The nonlinear refractive index (n_2) and nonlinear absorption coefficient (β) were also determined using Z-scan technique. The piezoelectric resonance in dielectric dispersions was observed at room temperature.

1. Introduction

In recent years much attention has been made in the development of nonlinear optical (NLO) crystals for second harmonic generation (SHG) and electro-optic modulation due to their widespread applications in high-speed information processing, optical communications and optical data storage. Amino acid based single crystals possess good NLO properties with better SHG efficiency than that of KDP [1,2]. Particularly, crystals with $P2_1$ space group have some advantages that they allow the maximal contribution of molecular non-linearity to the macroscopic crystal non-linearity compared to $P2_12_12_1$ structure [3]. L-lysine monohydrochloride dihydrate (L-LMHCl) is one of the amino acid based semiorganic materials, which crystallizes in monoclinic crystal system with non-centrosymmetric space group $P2_1$ [4] and is optically biaxial. Krishnakumar et al. [5] reported that the SHG efficiency of L-LMHCl is $\sim$2.5 times that of urea and Kalaiselvi et al. reported that its laser damage threshold is $\sim$52 MWcm^{-2} at 1064 nm [6]. Also L-LMHCl possesses dielectric and piezoelectric properties [7]. The conventional and unidirectional growth of L-LMHCL single crystals and their various physical properties were reported [8–11]. The effect of metal and amino acid dopants on L-LMHCl single crystals was reported by Vasudevan et al. [12]. In the present work, we report on the linear refractive indices, birefringence, Sellmeier dispersion coefficients, crossing angle between the optical axes, nonlinear refraction and absorption coefficients, and piezoelectric resonances of L-LMHCl single crystal for the first time.

2. Experimental

L-LMHCl single crystals were grown by slow solvent evaporation technique [12]. Grown single crystals of optical quality, without detectable solvent inclusion by optical microscope, were selected for characterization studies. The refractive index of a material can be accurately measured by minimum deviation [13,14] and ellipsometry [15] methods. The minimum deviation method is typically used for bulk materials, where some portion of the material is cut and polished into the shape of a wedge or a prism. In the present work the minimum deviation method was chosen to determine the principal refractive indices of L-LMHCl crystal. Two prisms (Fig. 1a) were cut from the as-grown crystal such that the axes of these prisms were oriented parallel to the crystallographic y-axis and z-axis, respectively, (Fig. 1b and c). The apex angles of the prisms ($A=58^\circ$ 2′ and $B=47^\circ$) were measured using sodium vapor lamp. The faces of prisms were polished using fine alumina powder to achieve good reflection. The mercury lamp was used as the light source for the measurement of angle of minimum deviation. The angle of minimum deviation at each wavelength was measured by a spectrometer. The optical homogeneity of L-LMHCl single crystal along the optical plane was observed through interference patterns using the simple interferometric technique [16]. For this measurement, L-LMHCl crystal was placed between the polarizer and analyzer,

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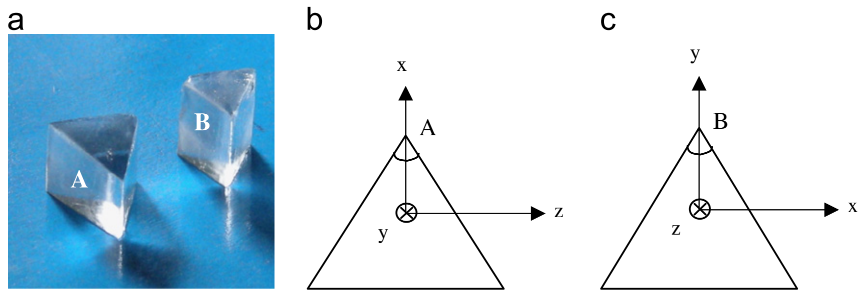

Fig. 1. (a) Cut and polished prisms and (b) and (c) schematic of prism orientations of L-LMHCl single crystal.

and Helium–Neon (He–Ne) laser source of wavelength 632.8 nm was allowed to transmit through it. The plane polarized light travels through the crystal at different phase velocities and splits into two orthogonal components (extraordinary and ordinary ray). These two components recombine when they pass through the analyzer and produce interference patterns on the screen.

The sign and magnitude of third-order nonlinearities of L-LMHCl single crystal was measured using Z-scan technique. He–Ne laser of wavelength 632.8 nm was used as the source and the experimental setup is reported elsewhere [17]. The transmittance of L-LMHCl was measured through a finite aperture in the far field from the focus as a function of the sample position Z. This gives information about the refractive nonlinearities of L-LMHCl crystal. The sample transmittance without aperture is also measured to extract complementary information about the absorptive nonlinearities of the sample. The nonlinear refractive index and nonlinear absorption coefficients were calculated using the sample transmittance collected from the closed and open aperture modes. A lens of focal length 12 cm was used to focus the laser beam on the crystal. The spot size of the beam at focus was calculated and found to be 45.12 μm. Dielectric measurements were carried out using Agilent E 4980 LCR meter in the frequency range 1 kHz–2 MHz. The cut and polished rectangular shaped single crystals were coated with high grade silver paste to provide electrical contact. The coated sample was placed between the two copper electrodes forming parallel plate capacitor. The dielectric behavior of the L-LMHCl single crystal was analyzed applying a suitable voltage across the sample.

3. Results and discussion

3.1. Refractive indices

Measurement of linear refractive indices as a function of wavelength is essential for designing the nonlinear optical devices such as second harmonic generator, optical parametric oscillator and electro-optic modulator. The linear refractive indices data are also useful to predict the phase matching angles for nonlinear optical crystals [18]. The refractive index of optical materials is influenced by the interaction between the atoms constituting the medium. It is therefore easy to recognize that in some materials the interaction between atoms is not the same in all directions and so anisotropy of the refractive index arises. In this regard, refractive indices (n_x, n_y, and n_z) along the principal directions of L-LMHCl crystal were measured using minimum deviation method. The value of principal refractive indices n_y and n_x were determined in the first prism (Fig. 1b) using vertically and horizontally polarized incident lights, respectively. In the second prism (Fig. 1c), the refractive index n_z was measured using vertically polarized light. Thus the refractive indices n_x, n_y, and n_z were measured for different wavelengths and they are represented by open symbols in Fig. 2. The solid lines represent theoretically calculated refractive indices using Cauchy's

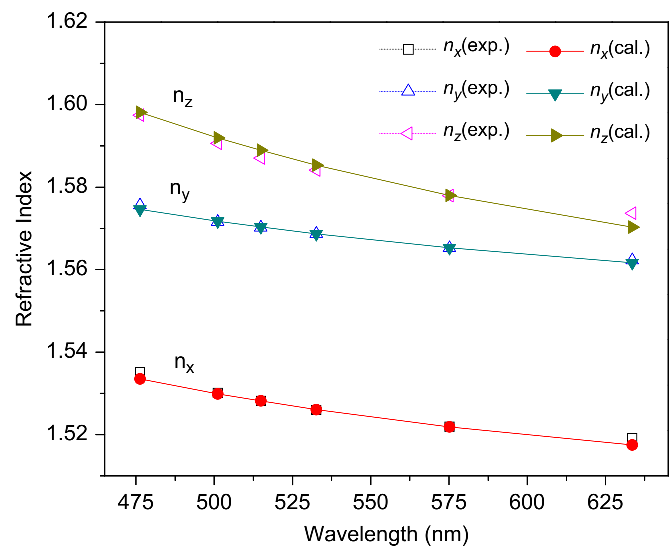

Fig. 2. Refractive indices (n_x, n_y, and n_z) of L-LMHCl single crystal measured at different wavelength. The solid lines represent the theoretically calculated refractive indices using Cauchy's equations.

Table 1
Birefringence (Δn) and crossing angle (2 V) of L-LMHCl crystal at different wavelengths.

λ (nm)	Δn (n_z-n_x)	Crossing angle (2 V)
476.3	0.0622	70
501.1	0.0604	66
514.9	0.0589	63
532.6	0.0581	60
575.2	0.0559	55
633.5	0.0544	53

equations. The Cauchy's constants of the principal refractive indices were obtained from the plot drawn between wavelength and the corresponding refractive indices (Fig. 2). The obtained Cauchy's equations are given below

$$n_x = 1.4968 + \frac{0.00832}{\lambda^2};\quad n_y = 1.5448 + \frac{0.006747}{\lambda^2};\quad n_z = 1.5340 + \frac{0.01448}{\lambda^2} \tag{1}$$

where λ is the wavelength. The calculated values agreed well with the observed values, which indicate that the theoretically calculated refractive indices using Cauchy's equations are reliable. The birefringence (Δn) of L-LMHCl crystals was calculated for different wavelengths and are given in Table 1. The maximum difference between the experimental refractive indices n_z and n_x (i.e., $\Delta n = n_z - n_x$), is 0.0622 at 476.3 nm. From the refractive index data it is observed that the difference between the values of n_z and n_y is smaller than that of n_y and n_x [19], which indicate the negative biaxial nature of