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Third order nonlinear optical, luminescence and electrical properties of L-phenylalanine L-phenylalaninium bromide bulk crystals

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ABSTRACT

Optical quality L-phenylalanine L-phenylalaninium bromide (LPLPB) bulk crystals were grown by slow evaporation technique. The third order nonlinear refractive index and nonlinear absorption coefficient of the grown crystal were measured by Z-scan studies. The third order nonlinear susceptibility was found to be 1.8792 x 10^{-4} esu which is fairly higher than the other compounds. The Photoluminescence spectra reveal the emission bands for LPLPB crystals. The Photoconductivity studies were employed to determine the dependence of photocurrent on the applied electric field. Negative photoconductivity was exhibited by the sample. The d. c. conductivity of the grown crystal was measured by the complex impedance analysis wherein the obtained plot in the form of semicircle finds application in Debye relaxation for materials having large dc conductivity.

1. Introduction

During the past three decades 'Optics' has secured an excellent position in application areas which was previously dominated by 'Electronics'. More than Discrete Electronics, the combination of electronics and optics leads to fantastic innovations in research applications. The field of 'Electro-Optics' emerged from the electromagnetic field of a laser beam incident on an atom, inducing electric polarization, manifesting interesting properties that are 'optically nonlinear'. Nonlinear optics is based on the interaction of laser light radiation with matter where the non-linear response of the atoms plays an important role. In a dielectric material, the influence of an electric field causes distortion in the spatial distribution between the electrons and the nucleus. These distortions cause electric dipoles, which in-turn manifest as polarization. At very low fields, the induced polarization is directly proportional to the electric field. The induced polarization is capable of multiplying the fundamental frequency to second, third order and even higher harmonics. The nonlinear susceptibilities have decreasing magnitudes as their order increases at $\chi^{(1)}$: $\chi^{(2)}$: $\chi^{(3)} = 1{:}10^{-8}{:}10^{-16}$. The first order susceptibility which is the linear term, $\chi^{(1)}$, gives rise to refractive index, absorption, dispersion and birefringence of the medium. The second order, $\chi^{(2)}$, gives rise to Second Harmonic Generation (SHG), frequency mixing and parametric generation, while the third order nonlinear susceptibility, $\chi^{(3)}$, gives rise to third harmonic generation, stimulated Raman scattering, optical bistability and conjugation [2]. Amidst all these properties, few properties are discussed in this paper.

$\chi^{(2)}$ materials are used for second harmonic generation that possesses an asymmetrical structure whose refractive index have been controlled with an external electric field, a property that is referred to as the electro-optic effect. This property is of prime importance for many applications and is currently used in electro-optic modulators. $\chi^{(3)}$ materials are expected to play a primary role in all optical switching devices since their optical properties can be constrained by light. However, due to the higher order of the nonlinearity, these materials are usually less efficient and have not reached the maturity of $\chi^{(2)}$ materials for device applications.

Photonic crystals are expected to play an important role in the development of new optical devices [3]. Nonlinear optical materials have acquired new significance with the advent of a large number of devices utilizing solid-state laser sources. They are essential for the fabrication of electro-optic modulators, which convert an electric signal into an optical one for transmission on a fiber optic cable [3]. Amino acids contain chiral carbon atom and crystallize in the noncentrosymmetric space groups and are potential candidates for optical second harmonic generation.

Among the amino acids, L-phenylalanine is an essential amino acid that is used by the body to build neurotransmitters. It is used in the

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manufacture of food and drinks and sold as a nutritional supplement for its reputed analgesic and antidepressant effects. Recently in one of the papers, the L-phenylalanine L-phenylalaninium bromide (LPLPB) growth process, crystal parameters, FTIR, UV–Vis spectral studies, dielectric, thermal and mechanical properties were thoroughly discussed [1]. However, to the best of author's knowledge, there is no report available on the third order optical nonlinear properties of the LPLPB crystal which is presented here for the first time. This paper brings out photoluminescence, photoconductive, complex impedance analysis of the crystal along with its z-scan studies that reveals the high-speed switching operations which are ideally faster than even the conventional electronics.

2. Materials processing

2.1. Synthesis

The LPLPB crystals were synthesized and recrystallized as per the method reported in the literature [1]. Initially the hydrobromic acid was diluted in the equimolar mixed solvents of water and acetone. To this aqueous solution, the calculated amount of L-phenylalanine was slowly added and homogeneously dissolved using a motorized magnetic stirrer at a temperature of 45 °C. The recrystallization procedure was repeated several times to reduce the impurity content. Finally the resultant supersaturated solution was filtered using a high-quality Whatman's filter paper. During filtration there was a quick formation of interesting needle-like crystals along the sides of the funnel that lacked stiffness. Hence, adopting a hot filtration method [4], such needle-shaped crystals were dissolved by warming the filtrate at 40 °C. After obtaining a clear filtrate, the contents were transferred to a beaker with a perforated lid and kept in a constant temperature bath set at 55 °C to achieve slow evaporation. After successive recrstyallization, over a period of 45 days, there was a bulk growth of L-phenylalanine L-phenylalaninium bromide having dimensions of $25 \times 3 \times 2\,mm^3$ as shown in Fig. 2. The harvested crystals were pale, non-hygroscopic, needle shaped and stable with appreciable transparency [1].

3. Results and discussions

3.1. Z-scan analysis

In nonlinear optics, z-scan technique is used to measure the non-linear index n_2 (Kerr nonlinearity) and the non-linear absorption coefficient $\Delta\alpha$ via the "closed" and "open" methods, respectively. As

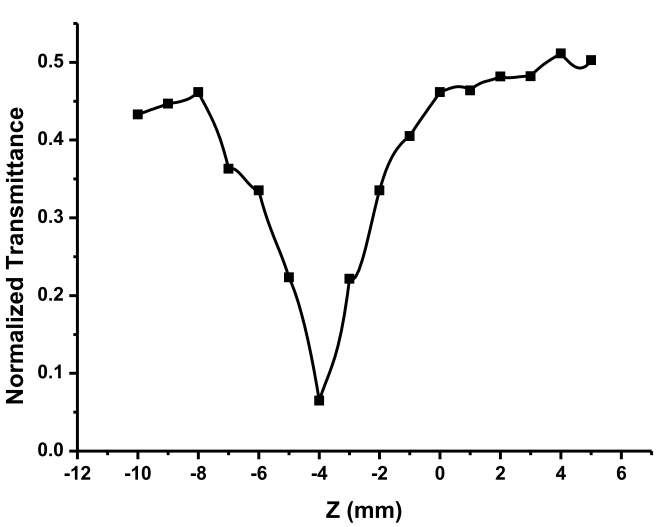

Fig. 1. Open aperture z-scan curve for lplpb crystal.

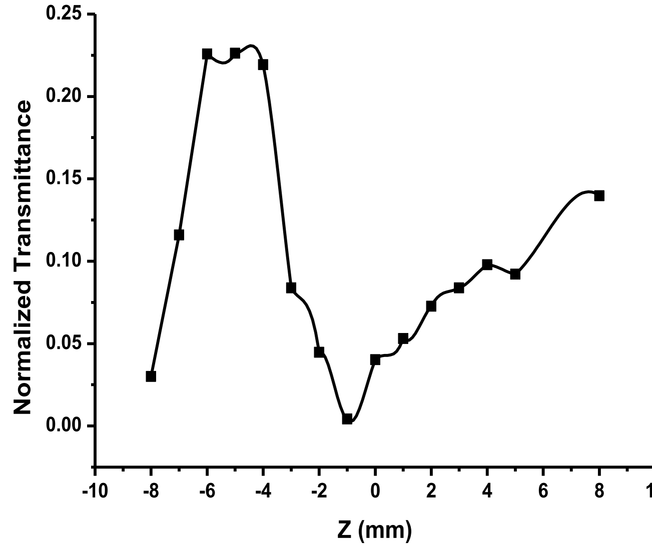

Fig. 2. Closed Aperture Z-scan Curve for lplpb crystal.

nonlinear absorption can affect the measurement of the non-linear index, the open method is typically used in conjunction with the closed method. For measuring the real part of the nonlinear refractive index, the z-scan setup is used in its closed-aperture form. It is a versatile standard tool in the study of nonlinear optics which evaluates the third order nonlinearity and determines the changes in nonlinear refractive index and variations in absorption. The third order optical nonlinearity was investigated by the Z-scan technique which enables simultaneous measurement of magnitude and sign of the non-linear refractive index (n_2) and the nonlinear absorption coefficient (β) of the samples leading to different properties like acoustically induced optical kerr effect, self-defocusing effect etc. The study of acoustically induced optical kerr effect in NLO materials leads to design of variety of acoustically-operated quantum electronic devices [5].

Materials such as insulating crystals and optical glasses typically possess nonlinear optical coefficients, $\chi^{(3)}$, of the order of 10^{-13}-10^{-14} esu. Electronic Polarization is believed to make the largest contribution to the nonlinear response of these materials.

The z-scan is a single beam technique developed by Sheik Bahae to measure the magnitude of nonlinear absorption as well as the sign and magnitude of nonlinear refraction [6]. Basically, the method consists of translating a sample through the focus of a Gaussian beam and monitoring the changes in the far field intensity pattern. When the intensity of the incident laser beam is sufficient to induce nonlinearity in the sample, it either converges (self-focusing) or diverges (self-defocusing) the beam, depending on the nature of that nonlinearity. By moving the sample through the focus, the intensity dependent absorption is measured as a change of transmittance through the sample (open aperture). The nonlinear refraction is determined by the intensity variation at the plane of a finite aperture placed in front of the detector (closed aperture), because the sample itself acts as a thin lens with varying focal length as it moves through the focal plane.

The Z-scan experiments were performed using a 532 nm diode pumped Nd:YAG laser beam (Coherent Compass TM 215M – 50), which was focused by 3.5 cm focal length lens. The distance between the lens and the laser was 13 cm. The LPLPB crystal is translated across the focal region along the axial direction that is the direction of the propagation laser beam. The transmission of the beam through an aperture placed in the far field was measured using photo detector fed to the digital power meter (Field master GS-coherent). For an open aperture Z-scan, a lens to collect the entire laser beam transmitted through the sample replaced the aperture.

The Z-scan theory is of vital significance in the field of nonlinear

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