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Investigation on growth of 4-N, N-dimethylamino-N′-methylstilbazolium p-chlorobenzenesulfonate: An efficient organic NLO crystal with potential THz properties

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ABSTRACT

The growth of an efficient organic nonlinear optical material 4-N, N-dimethylamino-N′-methylstilbazolium p-chlorobenzenesulfonate (DASC), a derivative of the stilbazolium tosylate family has been reported by employing the slope nucleation method coupled with slow solvent evaporation. The crystal system and lattice parameters are estimated by single crystal X-ray diffraction. The second harmonic conversion efficiency of DASC was determined using the Kurtz and Perry powder technique. The thermal stability of the compound is determined by thermo gravimetric/differential thermo gravimetric (TG/DTG) and differential scanning calorimetric (DSC) techniques. The hardness profile of the sample is investigated by Vickers microhardness test. The dielectric constant and dielectric loss of the crystal were studied as function of frequency. The surface properties of DASC are reported by undertaking scanning electron microscopy (SEM) and atomic force microscopy (AFM) studies.

1. Introduction

Terahertz (THz) technology has entered into an unprecedented revolutionary era with ever growing applications in biology and medicine, covering pharmaceutical industry, medical imaging, material spectroscopy and sensing, security and high data rate communications. High power and reliable THz sources and high performance THz spectroscopy and imaging systems have been developed over the past few years making it possible to explore new areas in science and technology [1]. However, the large THz portion of the spectrum has not been well developed because there was neither convenient high power emitter to send out controlled THz signals nor efficient sensors to collect them and record information. The lack of high power along with low cost portable room temperature THz source is the most significant limitation of modern THz systems. Coverage of a wide frequency range requires several different source systems; however the use of an organic nonlinear optical (NLO) crystal has the feasibility of ultra wide tenability for breakthrough developing THz source [1,2].

Nonlinear optical effects such as difference frequency generation (DFG) or optical rectification (OR) are widely used for generating coherent THz radiation. Appropriate source materials have a high nonlinearity optical susceptibility $\chi^{(2)}$ and a low refractive index in the THz frequency range. Recent research indicates that 4-N, N-dimethylamino-N′-methylstilbazolium p-toluenesulfonate (DAST) and its derivatives are promising candidates for THz wave generation devices [3]. By far, the generation of broadband high power THz waves from DAST single crystal has been realized up to 30 THz by DFG and the generation of sub-10 THz waves are achieved by OR [4,5]. However, a number of absorption features in the THz spectra of DAST limits its application. Another major constraint for the development of DAST crystal is to overcome the formation of hydrated DAST co-crystals, which destroys the noncentrosymmetric crystal structure [6]. In this connection, researchers are aiming to develop alternate sources to generate THz waves of different frequency ranges and thereby explore the possibility of further expanding the areas of applications.

The studies carried out by Yang et al. and Okada et al. have provided important information on how a change of the counter anion will modify the crystal structure, and the second harmonic generation (SHG) activity of stilbazolium salts, thus offering almost unlimited design possibilities to obtain new materials by straightforwardly varying counter anions [7–9]. Various derivatives

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