Journal of Electronic Materials (2024) 53:2333–2339
https://doi.org/10.1007/s11664-024-10992-3

ORIGINAL RESEARCH ARTICLE

Investigation on the Structure, Spectral and Third-Order Nonlinear Optical Analysis of a New Organic Crystal: 4-Methylbenzylammonium Hydrogen Succinate

P. Deepa[1] · R. Aarthi[2] · S. Kalainathan[3] · C. Ramachandra Raja[1]

Received: 17 December 2023 / Accepted: 14 February 2024 / Published online: 11 March 2024

Abstract
4-Methylbenzylammonium hydrogen succinate (4MLBAHS) was synthesized and successfully grown via a slow solvent evaporation process. The structure was solved and the space group identified as $P\bar{1}$. In the 4MLBAHS molecule, 4-methylbenzylammonium and hydrogen succinate are linked by O–H···O and N–H···O bonds. Functional group vibrations were revealed through vibrational analysis. UV–Vis–NIR analysis was used to determine the transparency bandwidth range from 260 nm to 1100 nm. ^{1}H and ^{13}C nuclear magnetic resonance (NMR) spectroscopy established the carbon–hydrogen network in 4MLBAHS. In ^{1}H NMR, the chemical shift for the NH_2 group of 4-methylbenzylamine was revealed at 1.42 ppm. This was shifted to 8.094 ppm in 4MLBAHS, owing to the origination of the N–H···O bond between the 4-methylbenzylammonium cation and hydrogen succinate anion. The third-order nonlinear optical characteristics were analysed using Z-scan analysis. χ^3 was established as 3.0879×10^{-5} esu. The presence of hydrogen bond interactions in the 4MLBAHS crystal enhanced the χ^3 value.

Keywords Crystal growth · slow evaporation method · NLO materials · hydrogen bond · molecular structure · optical limiting

Introduction

Materials having optical nonlinearity with good physical and chemical flexibility are critical for a variety of applications. Novel materials have been utilized in the development of optical communication systems, where organic materials have significant potential for improving the systems.[1,2] Organic materials are widely used due to their high figure of merit in linear and nonlinear optical response during device fabrication.[3] Organic crystals offer synthetic flexibility for the design and production of higher-order nonlinear optical (NLO) materials used in optical switching applications, optical information processing and storage, and laser technology.[4,5]

Carboxylic acid has been found to be a potential component in the growth process owing to its strong hydrogen bond interactions with the reacting substance. Succinic acid exists in the neutral state as succinic acid and in the ionized state as succinate, forming a strong intermolecular hydrogen bond with other molecules. It is used extensively in the pharmaceutical and food industries, in the fabrication of high-electron-mobility transistors, and for the production of biodegradable polymers and surfactants.[6–8] Carboxylic acids, when associated with an amine group, give a higher-order nonlinear response.[9] The slow evaporation solution growth process is simple and economical, and can be carried out at low temperature.[10] Our researchers have worked on efficient organic NLO compounds under the amino acid category, such as L-alaninium succinate (LAS) and valinium succinate (LVS). Both crystals show wide optical transmittance, which enhances their NLO properties.[11,12] We have also reported several new 4-methylbenzylamine-based crystals.[13–20] Recently, new crystals showing enhanced third-order NLO

✉ C. Ramachandra Raja
crrajaphy@gmail.com

[1] Government Arts College (Autonomous) (Affiliated to Bharathidasan University, Tiruchirappalli), Kumbakonam 612002, India

[2] SRM Institute of Science and Technology, Tiruchirappalli 621105, India

[3] Centre for Nanotechnology Research, Vellore Institute of Technology, Vellore 632014, India

efficiency and with wide optical transparency have been developed and reported.[21–23]

All these investigations demonstrate that 4-methylbenzylamine plays a prominent role in the field of photonics. In this work, the structural, spectral and optical properties of 4-methylbenzylammonium hydrogen succinate (4MLBAHS) are reported. In the 4MLBAHS crystal, succinic acid is a carboxylic acid with the potential to form strong hydrogen bond interactions with 4-methylbenzylamine, forming a new structure. The crystal reveals a two-dimensional structure between the cation and anion connected by hydrogen bonds. 4MLBAHS can be utilized in night vision devices and in optical-limiting and photonic device applications.

Materials and Methods

Analytical reagent (AR)-grade solution of 4-methylbenzylamine (4MLBA) and succinic acid (SA) were used for synthesizing the title crystal 4MLBAHS [$(C_8H_{12}N^+)(C_4H_5O_4^-)$] by the slow evaporation solution growth method. 4MLBAHS was developed by mixing 4MLBA and SA in equimolar proportions. The mixture was stirred in 50 mL of deionized water for 3 h, and the resulting solution was kept undisturbed. Transparent crystals developed due to spontaneous nucleation and were harvested after 15 days. The crystal growth reaction of 4MLBAHS is given in Fig. 1.

Results and Discussion

Single-Crystal X-ray Diffraction (SCXRD) Analysis

Unit cell constants were estimated using a Bruker APEX3 diffractometer. The observed cell constants are $a = 9.3710(4)$ Å, $b = 12.0713(5)$ Å, $c = 22.5779(10)$ Å, $\alpha = 94.959(2)°$, $\beta = 90.964(2)°$, $\gamma = 96.082(19)°$ and $V = 2529.26(19)$ Å^3. The structure was solved using SHELX-97, and using the full-matrix least-squares method, refinement was performed to an R-value of 0.1680. A molecular diagram was generated using Mercury 3.8 software. The 4MLBAHS crystal data are given in Supplementary Table S1. The interatomic distances and angles of 4MLBAHS are presented in Supplementary Table S2 and Table S3. The 4MLBAHS structure contains $(C_8H_{12}N)^+$ cation and hydrogen succinate $(C_4H_5O_4)^-$ anion joined by N(1)–H(1C)···O(6) bonds. The anion molecules are connected with neighbouring anions through O(4)–H(4)···O(10) hydrogen bonds. Hydrogen bonds are shown in Supplementary Table S4. A two-dimensional structure arises because of the weak C–H···π interactions. In the structure of 4MLBAHS, the methylene unit (CH_2) from the positively charged ammonium group $(C_8H_{12}N)^+$ is a donor and the hydrogen succinate $(C_4H_5O_4)^-$ negatively charged group acts as an acceptor. An ORTEP (Oak Ridge Thermal Ellipsoid Plot) diagram of 4MLBAHS is given in Fig. 2a. Figure 2b shows the crystal packing diagram of 4MLBAHS viewed along the b*-axis. The morphology of the grown crystals was indexed using WinXMorph software. Figure 2c shows the morphology of 4MLBAHS. It contains 14 well-developed faces (1 0 0), (1 0 −1), (−1 0 0), (−1 0 1), (0 1 0), (0 1 −1), (0 −1 0), (0 −1 1), (−1 1 0), (−1 1 −1), (1 −1 1), (1 1 0), (0 0 1) and (0 0 −1). The growth rate of 4MLBAHS is extended along the crystallographic a-axis. The structure of 4MLBAHS is deposited as CCDC number 2233050.

Linear Optical Analysis

The optical transmission spectrum of the grown organic crystal of 4MLBAHS was recorded with a PerkinElmer ultraviolet–visible–near-infrared (UV–Vis–NIR) spectrophotometer as shown in Fig. 3. 4MLBAHS exhibits high transparency, with a cut-off wavelength of 260 nm. The wide optical transmission in the 260–1100 nm region reveals that 4MLBAHS possesses fewer structural defects and can be used in optical applications.[17] When compared with the previously reported crystals of 4-methylbenzylammonium oxalate hydrate (4MLBAOX) and 4-methylbenzylammonium hydrogen maleate (4MBAHM), their transmission range is from 262 nm to 1100 nm,[21,23] whereas the 4MLBAHS crystal possesses a more prominent wide transmission window and can be utilized in NLO device applications.

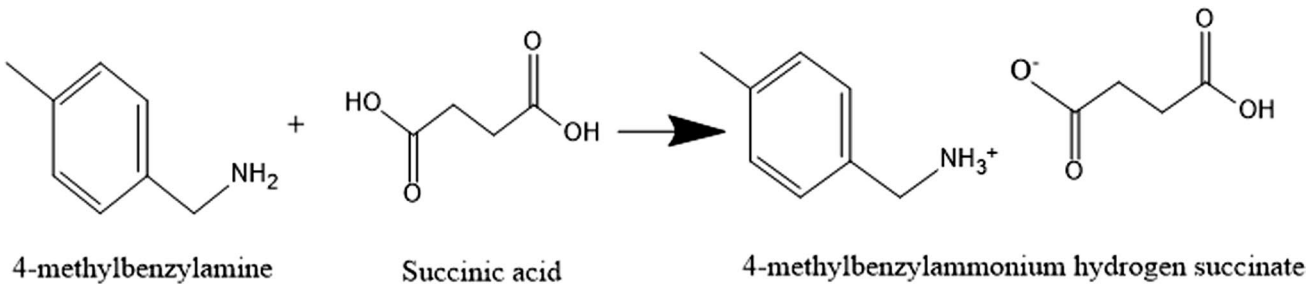

Fig. 1 Reaction scheme of 4MLBAHS crystal.

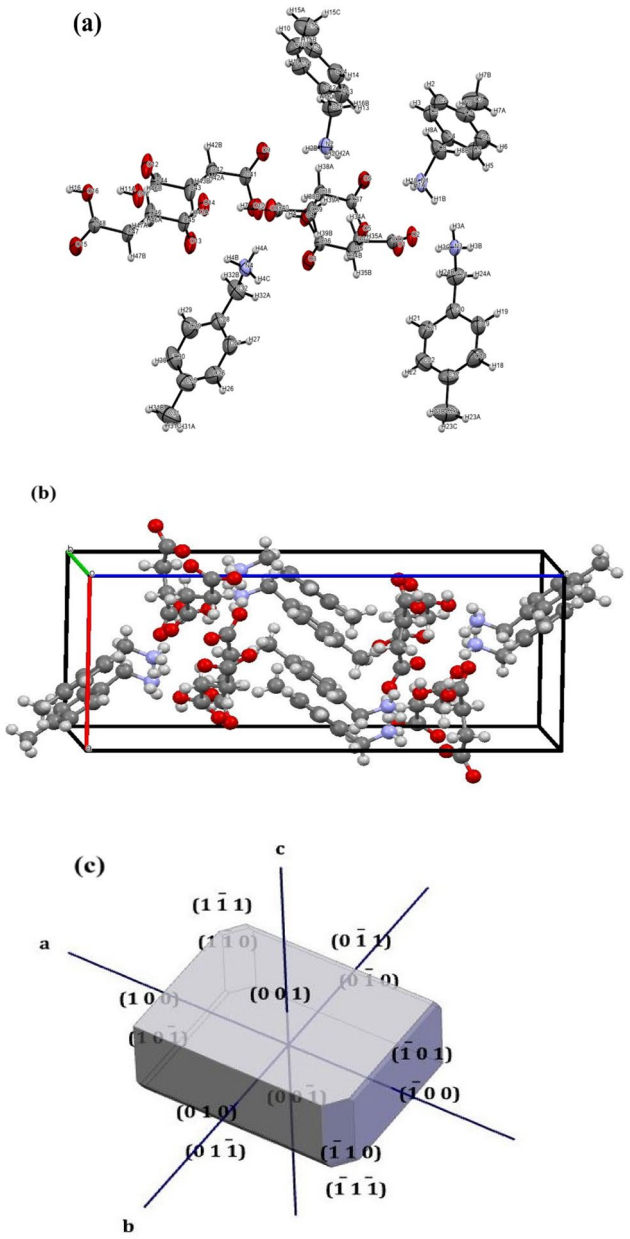

Fig. 2 (a) ORTEP view of the 4MLBAHS crystal. (b) Packing diagram of the 4MLBAHS crystal partly viewed along the *b**-axis. (c) Morphology of the grown crystal 4MLBAHS.

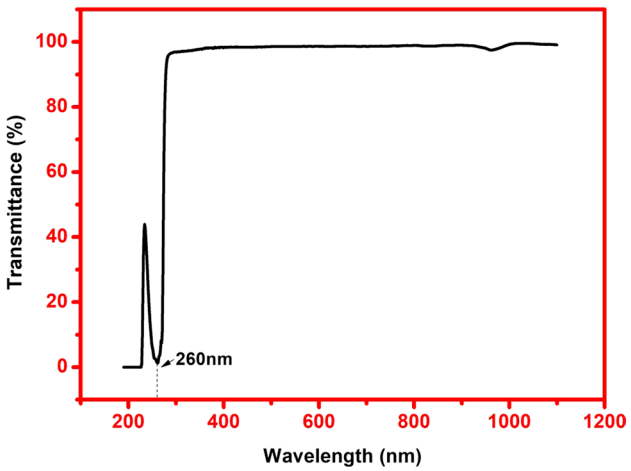

Fig. 3 Linear optical transmission spectrum of 4MLBAHS crystal.

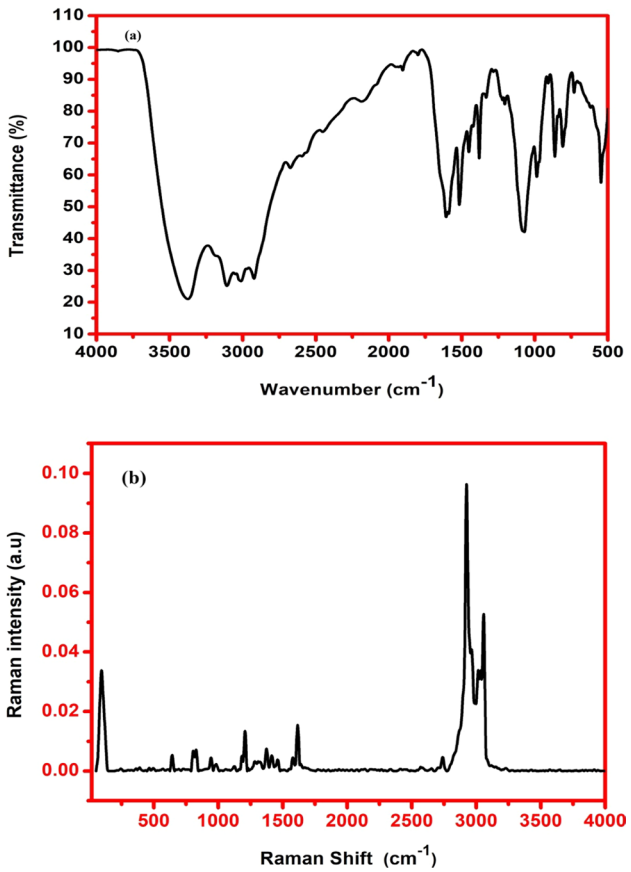

Fig. 4 (a) FTIR spectrum of 4MLBAHS crystal. (b) FT-Raman spectrum of 4MLBAHS crystal.

Fourier transform infrared (FTIR) and FT- Raman Vibrational Studies

The vibrational spectra of the 4MLBAHS crystal are given in Fig. 4a and b. The peak appearing at around 3360 cm^{-1} is due to NH_3^+ stretching. NH_3^+ symmetric bending modes were identified at 1610 cm^{-1} and 1644 cm^{-1} in IR and 1578 cm^{-1} and 1615 cm^{-1} in Raman spectroscopy. COO^- asymmetric and symmetric stretching of SA was observed at 1514 cm^{-1} and 1412 cm^{-1} in IR and 1414 cm^{-1} in Raman spectroscopy. COO^- bending was found at 639 cm^{-1} and 642 cm^{-1} in IR and Raman spectroscopy, respectively. The presence of NH_3^+ and COO^- vibrations ensures the formation of the 4MLBAHS

crystal. The vibrations of the other functional groups of 4MLBAHS were analysed in accordance with the literature[17,20,24–28] and are presented in Table I.

^{1}H NMR Spectral Study

^{1}H and ^{13}C NMR spectra of 4MLBAHS were recorded with a Bruker 400 MHZ spectrometer, and the spectra are depicted in Fig. 5a and b, respectively. Dimethyl sulfoxide (DMSO) was used as water (peak at 3.432 ppm) and DMSO solvent was observed at 2.506 ppm.[18]

In pure 4MLBA, the signals for CH_3, CH_2, CH (H in aromatic ring) and NH_2 groups were recorded at 2.320 ppm, 3.795 ppm, 6–8 ppm and 1.42 ppm, respectively. In 4MLBAHS, the signals for CH_3, CH_2 and CH (H in aromatic ring) were observed at 2.087 ppm, 3.982 ppm and 7.220−7.350 ppm, respectively.[19,24] Part of the spectrum appears as a quartet, and the coupling constant is given as J (0.02 ppm × 400.231 MHz) = 8.00462 Hz. The observed intensities show that the inner peaks are taller than the outer peaks, such as (7.239 ppm and 7.330 ppm) representing inner peaks and (7.220 ppm and 7.350 ppm) representing outer peaks. The resonating protons are coupled to form a strong coupling effect, leading to variation in the splitting pattern of the peaks known as the 'roof effect'.[29] The chemical shift for NH_2 is largely shifted and observed at 8.094 ppm. This is attributed to the protonation of the NH_2 group and ensures the development of the title crystal.[30] In succinic acid, the signal for CH_2 is revealed at 2.425 ppm, and in 4MLBAHS it is observed at 2.312 ppm.[12]

^{13}C NMR Spectral Study

In ^{13}C NMR, for pure 4MLBA, the signals for CH_3, CH_2 and CH (H in aromatic ring) are revealed at 20.97 ppm, 46.21 ppm and 126.95–140.47 ppm, respectively. In 4MLBAHS, the signals for CH_3, CH_2 and CH (H in aromatic ring) are shifted and observed at 20.23 ppm, 42.81 ppm and 128.83–139.56 ppm.[30] In succinic acid, the CH_2 and COOH signals are revealed at 28.75 ppm and 173.48 ppm. In 4MLBAHS, the signals are shifted and observed at 31.23 ppm and 179.57 ppm, respectively.[31,32] The intermolecular interactions, molecular structure and protonation of the amine group were well established by the shifts observed in both ^{1}H and ^{13}C NMR spectra. The shifts are tabulated along with the assignments in Table II.

Z-Scan Analysis

Z-Scan was carried out at 632.8 nm using a He-Ne laser (intensity = 3.13 mW cm^{-2}). Figure 6a and b depicts the closed and open aperture graphs of 4MLBAHS. The observed n_2, β and χ^3 values are displayed in Table III. The pre-focal peak followed by post-focal valley reveals the self-defocusing nature of the 4MLBAHS crystals, making them appropriate for use in optical sensors and night vision devices.[33–35] 4MLBAHS shows minimum transmittance at $Z = 0$ due to the reverse saturable absorption in the open aperture curve. This can be effectively employed in optical limiting applications.[36,37] The χ^3 value of 4MLBAHS is found to be 3.0879×10^{-5} esu, which is higher than the χ^3 values of LMSA, KSSA, MA-KDP, OA-KDP and BCSSA crystals reported in previous works, which are compared and shown in Table IV. The hydrogen bonds in the 4MLBAHS

Table I FTIR and FT–Raman spectral assignment of 4MLBAHS crystal

FTIR, cm^{-1}	FT-Raman, cm^{-1}	Assignments
3360	–	Stretching vibrations of NH_3^+
3109, 3008	3015, 3057	C–H asymmetric and symmetric stretching vibrations
2925	2925	C–H stretching vibrations of carboxylic acid
2681	2739	NH_3^+ overtone bands
2598	–	O–H stretching vibrations of carboxylic acid
1707	–	C=O stretching vibration of carboxylic acid
1610, 1644	1578, 1615	NH_3^+ symmetric bending vibration
1453, 1380	1460, 1379	CH_3 bending vibrations
1514, 1412	1414	COO^- asymmetric and symmetric vibrations of succinic acid
1281	1283	CH_3 deformation vibration
1204	1207	C–C bending vibrations
1022, 968	983, 943	C–H in-plane deformation vibration
808, 838	828	CH_2 rocking vibrations
639	642	COO^- bending vibrations
461, 547	–	Benzene ring vibration
–	95	Lattice mode vibration

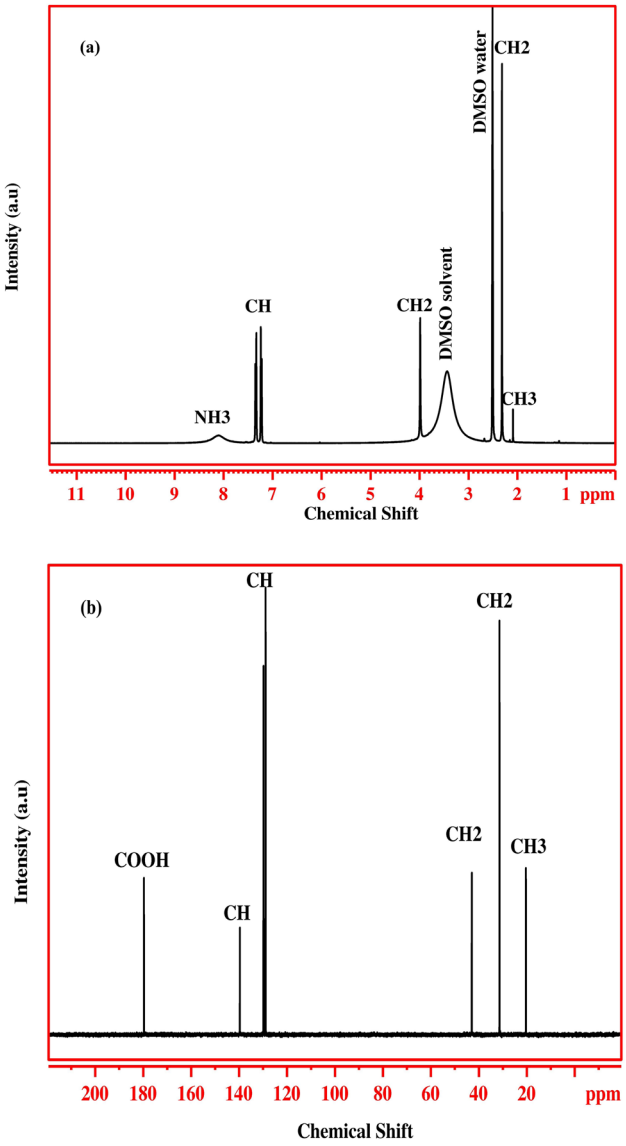

Fig. 5 (a) ^{1}H NMR spectrum of 4MLBAHS crystal. (b) ^{13}C NMR spectrum of 4MLBAHS crystal.

Table II Chemical shifts in ^{1}H and ^{13}C NMR spectrum of 4MLBAHS

NMR	4MLBAHS Chemical shift (ppm)	Group identification
^{1}H NMR	2.087	CH_3
	2.312	CH_2 in succinic acid
	3.982	CH_2
	7.220, 7.239, 7.330, 7.350	CH (H in aromatic ring)
	8.094	NH_3
^{13}C NMR	20.23	CH_3
	31.23	CH_2 in succinic acid
	42.81	CH_2
	128.83, 129.58, 129.72, 139.56	CH (C in aromatic ring)
	179.57	COOH in succinic acid

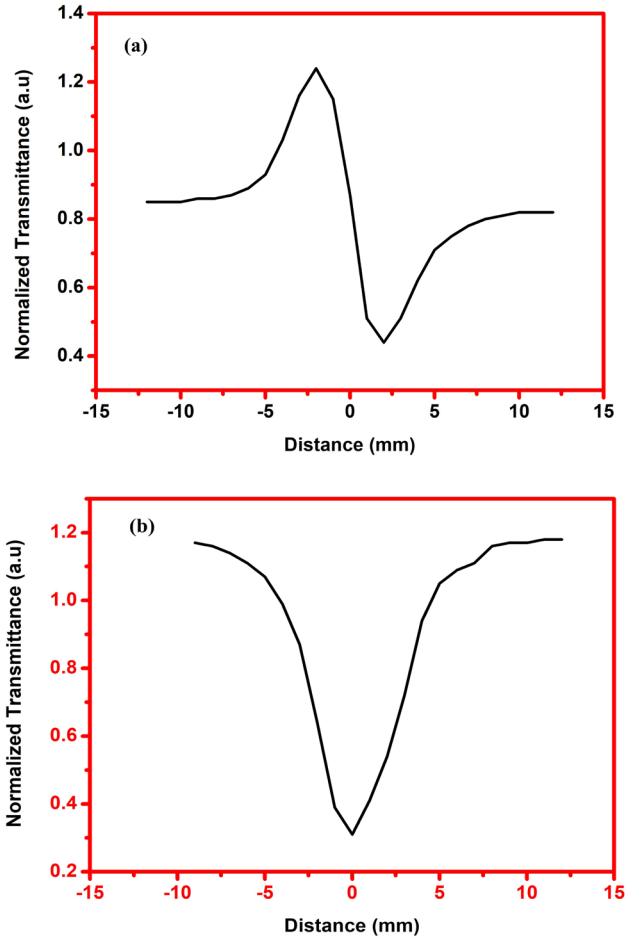

Fig. 6 (a) Closed aperture trace of 4MLBAHS crystal. (b) Open aperture trace of 4MLBAHS crystal.

molecule improve the charge transfer mechanism, which results in a higher χ^3 value.

Conclusion

Organic crystals of 4MLBAHS were effectively synthesized by a slow evaporation technique. The structure was solved and comprises 4-methylbenzylammonium $(C_8H_{12}N)^+$ cation and hydrogen succinate $(C_4H_5O_4)^-$ anion connected by hydrogen bonds. 4MLBAHS is transparent from 260 nm to 1100 nm. The functional group vibrations were analysed. The molecular structure and the significance of intermolecular hydrogen bonding were established by NMR spectral analysis. 4MLBAHS exhibits self-defocusing and reverse saturable absorption properties. The χ^3 value of the crystal is 3.0879×10^{-5} esu. The results reveal that 4MLBAHS can be utilized in night vision, optical-limiting and photonic devices.

Table III Z-scan parameters of 4MLBAHS crystal

Nonlinear refractive index (n_2)	7.3278×10^{-10}, m^2/W
Nonlinear absorption coefficient (β)	8.9732×10^{-4} m/W
Real part of the third-order susceptibility [Re(χ^3)]	4.9408×10^{-6} esu
Imaginary part of the third-order susceptibility [Im(χ^3)]	3.0482×10^{-5} esu
Third-order nonlinear susceptibility (χ^3)	3.0879×10^{-5} esu

Table IV Comparison of χ^3 value of some organic NLO crystals

Crystals	Third-order nonlinear optical susceptibility (χ^3) esu	References
LMSA	6.346×10^{-6} esu	38
KSSA	6.464×10^{-9}esu	28
MA-KDP	2.13×10^{-7} esu	39
OA-KDP	1.90×10^{-7} esu	39
BCSSA	2.274×10^{-6} esu	40
4MLBAOX	2.9457×10^{-6}esu	21
4MLBAHP	1.6949×10^{-6} esu	22
4MLBAHS	3.0879×10^{-5} esu	Present work

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11664-024-10992-3.

Acknowledgments The authors thank the Sophisticated Analytical Instruments Facility (SAIF), Indian Institute of Technology, Chennai, for providing SCXRD and Raman spectroscopy. The authors gratefully acknowledge the Instrumentation Centre of St. Joseph's College, Trichy for recording UV–Vis–NIR and FTIR spectra, and Gandhigram University, Dindigul, for providing the NMR spectrum. Also, the authors express special thanks to Dr. D. Sastikumar, NIT, Trichy, for providing us with a third-order nonlinear testing facility for recording Z-scan measurements.

Author Contributions PD: crystal growth, characterization and manuscript writing. RA: formal analysis. SK: structure determination. CRR: supervision.

Funding The authors declare that no funds, grants, or other support were received during the preparation of this manuscript.

Data Availability All data are presented in the manuscript. Crystallographic data have been deposited at the Cambridge Crystallographic Data Centre, CCDC number CCDC2233050.

Conflict of interest We wish to confirm that there are no conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

Ethical Approval Not applicable

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