## Supplementary Information

# Bulk Crystal Growth and Characterization of Imidazolium L-Tartrate (IMLT): A Novel Organic Nonlinear Optical Material with the High Laser-induced Damage Threshold

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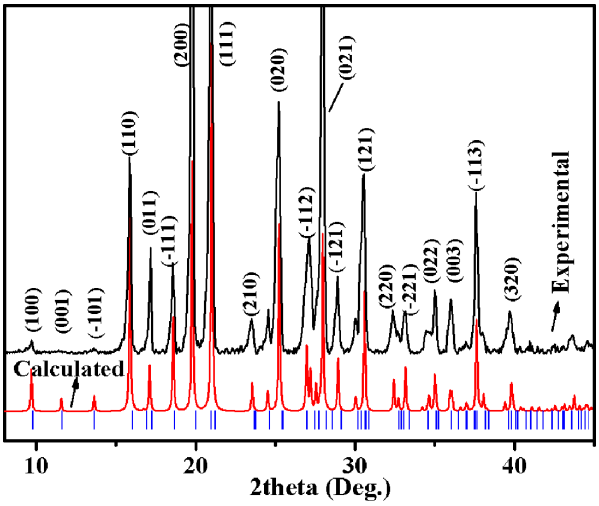


**Figure S1.** Simulated and experimental XRD powder patterns for IMLT

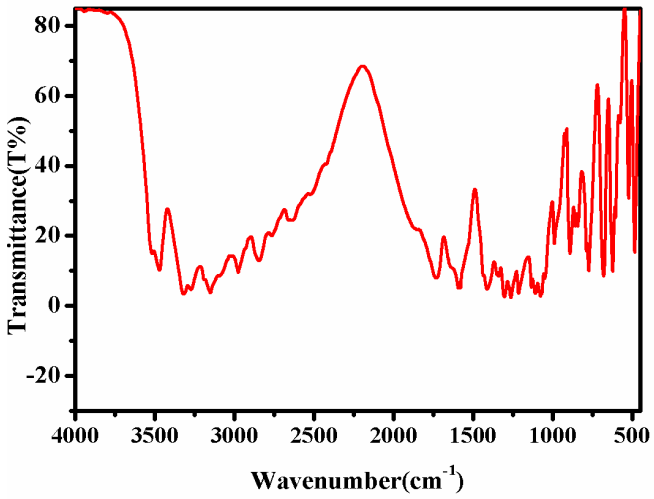


**Figure S2.** The FT-IR spectrum of IMLT

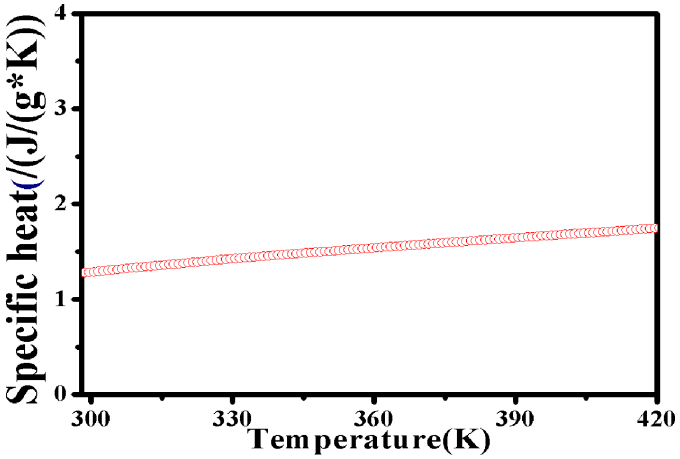


Figure **S3**. Dependence of specific heat of IMLT crystal with the temperature.

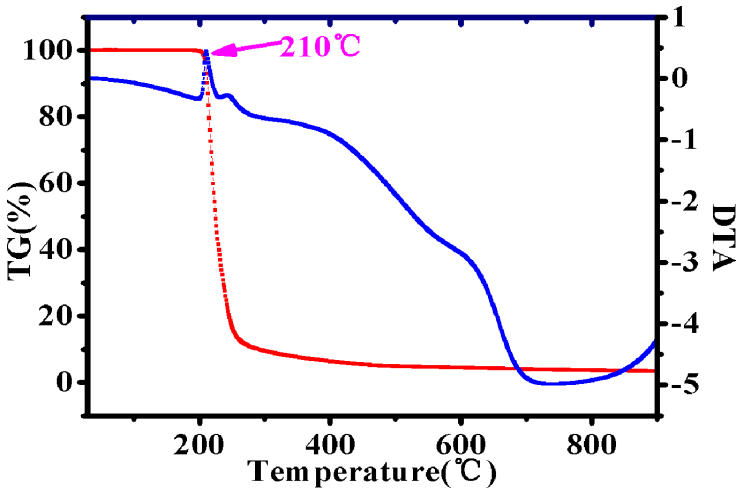


**Figure S4.** Thermogravimetric analysis for IMLT

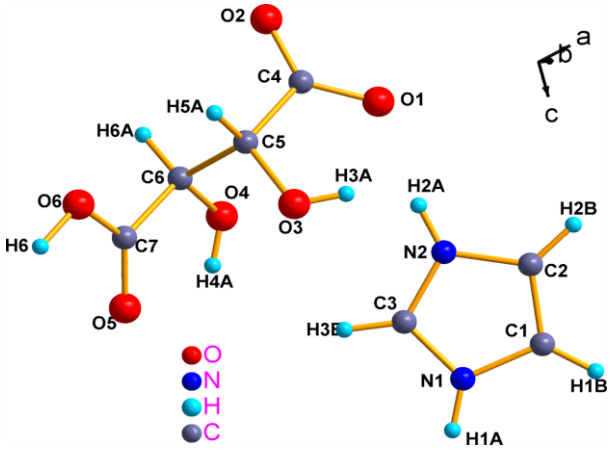


**Figure S5. Crystal structure of IMLT with the atomic numbering**

**Table S1.**. Crystal data and structure refinement for IMLT.

| | |
|---|---|
| Empirical formula | $C_7 H_{10} N_2 O_6$ |
| Formula weight | 218.17 |
| Temperature/K | 293(2) |
| Radiation | Mo-Kα (0.71073 Å) |
| Crystal system | Monoclinic |
| Space group | $P2_1$ |
| *a*/Å | 7.577(3) |
| *b*/Å | 6.894(2) |
| *c*/Å | 8.927(4) |
| *α*/deg | 90 |
| *β*/deg | 102.174(5) |
| *γ*/deg | 90 |
| Volume/ $Å^3$ | 455.9(3) |
| Z | 2 |
| Calculated density/g $cm^{-3}$ | 1.589 |

| | |
|---|---|
| Absorption coefficient/mm$^{-1}$ | 0.141 |
| $F$(000) | 228 |
| Crystal size/mm | 0.35×0.29×0.22 |
| Limiting indices | -9⩽h⩽9, -8⩽k⩽8, -11⩽l⩽11 |
| Reflections collected / unique | 5245 / 2019 [$R$(int) = 0.0177] |
| GOF | 1.041 |
| $R_1$, w$R_2$ [$I>2\sigma(I)$] | 0.0246/0.0635 |
| $R_1$, w$R_2$ (all data) | 0.0257/0.0641 |

**Table S2**. Bond lengths [Å] and angles [deg] for IMLT

| | | | |
|---|---|---|---|
| O(1)-C(4) | 1.2726(14) | C(3)-N(2)-C(2) | 109.32(10) |
| N(1)-C(3) | 1.3265(17) | C(3)-N(2)-H(2A) | 125.3 |
| N(1)-C(1) | 1.3782(17) | C(2)-N(2)-H(2A) | 125.3 |
| N(1)-H(1A) | 0.8800 | C(1)-C(2)-N(2) | 106.63(11) |
| C(1)-C(2) | 1.3523(17) | C(1)-C(2)-H(2B) | 126.7 |
| C(1)-H(1B) | 0.9500 | N(2)-C(2)-H(2B) | 126.7 |
| O(2)-C(4) | 1.2428(15) | C(5)-O(3)-H(3A) | 107.9(13) |
| N(2)-C(3) | 1.3266(16) | N(2)-C(3)-N(1) | 108.10(11) |
| N(2)-C(2) | 1.3756(17) | N(2)-C(3)-H(3B) | 125.9 |
| N(2)-H(2A) | 0.8800 | N(1)-C(3)-H(3B) | 125.9 |
| C(2)-H(2B) | 0.9500 | C(6)-O(4)-H(4A) | 109.5 |
| O(3)-C(5) | 1.4202(14) | O(2)-C(4)-O(1) | 126.63(10) |
| O(3)-H(3A) | 0.840(19) | O(2)-C(4)-C(5) | 117.24(10) |
| C(3)-H(3B) | 0.9500 | O(1)-C(4)-C(5) | 116.13(10) |
| O(4)-C(6) | 1.4145(14) | O(3)-C(5)-C(4) | 112.71(9) |
| O(4)-H(4A) | 0.8400 | O(3)-C(5)-C(6) | 109.17(9) |
| C(4)-C(5) | 1.5302(16) | C(4)-C(5)-C(6) | 109.62(10) |
| O(5)-C(7) | 1.2164(15) | O(3)-C(5)-H(5A) | 108.4 |
| C(5)-C(6) | 1.5339(17) | C(4)-C(5)-H(5A) | 108.4 |
| C(5)-H(5A) | 1.0000 | C(6)-C(5)-H(5A) | 108.4 |
| O(6)-C(7) | 1.3074(15) | C(7)-O(6)-H(6) | 111.2(13) |
| O(6)-H(6) | 1.04(2) | O(4)-C(6)-C(7) | 110.67(10) |
| C(6)-C(7) | 1.5265(16) | O(4)-C(6)-C(5) | 110.06(10) |
| C(6)-H(6A) | 1.0000 | C(7)-C(6)-C(5) | 109.42(10) |
| | | O(4)-C(6)-H(6A) | 108.9 |
| C(3)-N(1)-C(1) | 108.99(10) | C(7)-C(6)-H(6A) | 108.9 |
| C(3)-N(1)-H(1A) | 125.5 | C(5)-C(6)-H(6A) | 108.9 |
| C(1)-N(1)-H(1A) | 125.5 | O(5)-C(7)-O(6) | 125.27(11) |
| C(2)-C(1)-N(1) | 106.95(11) | O(5)-C(7)-C(6) | 121.37(10) |
| C(2)-C(1)-H(1B) | 126.5 | O(6)-C(7)-C(6) | 113.36(10) |
| N(1)-C(1)-H(1B) | 126.5 | | |

The positions of hydrogen atoms bonded to hydroxyl groups were fixed geometrically at calculated distances of 0.82 Å and refined using "riding" model with *Uiso* = 1.5*Ueq* (O).

**Table S3**. Torsion angles [deg] for IMLT.

| | |
|---|---|
| C(3)-N(1)-C(1)-C(2) | -0.16(16) |
| N(1)-C(1)-C(2)-N(2) | -0.20(15) |
| C(3)-N(2)-C(2)-C(1) | 0.49(16) |
| C(2)-N(2)-C(3)-N(1) | -0.60(16) |
| C(1)-N(1)-C(3)-N(2) | 0.47(16) |
| O(2)-C(4)-C(5)-O(3) | -168.92(10) |
| O(1)-C(4)-C(5)-O(3) | 11.09(15) |
| O(2)-C(4)-C(5)-C(6) | 69.26(13) |
| O(1)-C(4)-C(5)-C(6) | -110.72(12) |
| O(3)-C(5)-C(6)-O(4) | -65.12(12) |
| C(4)-C(5)-C(6)-O(4) | 58.78(12) |
| O(3)-C(5)-C(6)-C(7) | 56.69(12) |
| C(4)-C(5)-C(6)-C(7) | -179.40(9) |
| O(4)-C(6)-C(7)-O(5) | 5.81(16) |
| C(5)-C(6)-C(7)-O(5) | -115.64(12) |
| O(4)-C(6)-C(7)-O(6) | -174.51(10) |
| C(5)-C(6)-C(7)-O(6) | 64.04(13) |

Symmetry transformations used to generate equivalent atoms

**Table S4**. Hydrogen bonds for IMLT [Å and deg.].

| D-H···A | d(D-H) | d(H···A) | d(D···A) | ＜(DHA) |
|---|---|---|---|---|
| N(1)-H(1A) ···O(2) i | 0.88 | 1.86 | 2.7245(15) | 167.1 |
| O(6)-H(6) ···O(1) ii | 1.04(2) | 1.47(2) | 2.5107(14) | 179(2) |
| O(6)-H(6) ···O(2) ii | 1.04(2) | 2.65(2) | 3.3357(16) | 123.2(16) |
| O(4)-H(4A) ···O(3) iii | 0.84 | 2.17 | 2.9756(15) | 161.3 |
| O(3)-H(3A) ···O(5) iv | 0.840(19) | 2.291(19) | 2.8384(14) | 123.1(15) |
| N(2)-H(2A) ···O(1) | 0.88 | 2.14 | 2.9257(17) | 149.1 |
| O(4)-H(4A) ···O(5) | 0.84 | 2.19 | 2.6770(15) | 117.3 |
| O(3)-H(3A) ···O(1) | 0.840(19) | 2.151(18) | 2.6547(14) | 118.3(16) |

Symmetry transformations used to generate equivalent atoms:
( i ) x,y,z+1; ( ii ) x-1,y,z; (iii) -x+1,y-1/2,-z+2; (iv) -x+1,y+1/2,-z+2.

**Table S5**. Atomic coordinates and equivalent isotropic displacement parameters ($Å^2$) for IMLT.

| Atoms | *x* | *y* | *z* | *Ueq* |
|---|---|---|---|---|
| O1 | 0.84438(11) | 0.38253(14) | 0.76027(9) | 0.01443(19) |
| N1 | 0.89626(13) | 0.29480(17) | 1.31814(11) | 0.0153(2) |
| C1 | 1.06660(16) | 0.3669(2) | 1.32119(13) | 0.0159(2) |
| O2 | 0.69342(11) | 0.26321(13) | 0.53525(9) | 0.0165(2) |

| | | | | |
|---|---|---|---|---|
| N2 | 0.93557(15) | 0.28745(17) | 1.08689(11) | 0.0160(2) |
| C2 | 1.09119(16) | 0.3627(2) | 1.17554(13) | 0.0158(2) |
| O3 | 0.54525(12) | 0.49868(14) | 0.84377(10) | 0.0169(2) |
| C3 | 0.81931(17) | 0.24854(19) | 1.17505(14) | 0.0164(3) |
| O4 | 0.54471(12) | 0.08248(15) | 0.85399(10) | 0.0188(2) |
| C4 | 0.70166(15) | 0.33801(17) | 0.66320(12) | 0.0118(2) |
| O5 | 0.22583(11) | 0.19316(13) | 0.90922(9) | 0.01569(19) |
| C5 | 0.52286(15) | 0.38168(18) | 0.71017(12) | 0.0118(2) |
| O6 | 0.13453(11) | 0.32103(14) | 0.67478(9) | 0.0154(2) |
| C6 | 0.43062(15) | 0.19097(17) | 0.73805(13) | 0.0126(2) |
| C7 | 0.25141(15) | 0.23525(18) | 0.78319(13) | 0.0121(2) |

**Table S6.** Thermal expansion coefficients for IMLT and some other known NLO crystals

| Crystal | crystallographic system | Thermal expansion coefficients ($\times 10^{-5}$ $K^{-1}$) | | |
|---|---|---|---|---|
| | | $\alpha_1$ | $\alpha_2$ | $\alpha_3$ |
| NPNa dihydrate | orthorhombic | 7.48 | 0.43 | 1.99 |
| LABTF | monoclinic | 9.87 | -0.86 | 7.04 |
| LAP[1] | monoclinic | -1.75 | 0.96 | 1.64 |
| DLAP | monoclinic | 5.74 | 0.96 | 1.83 |
| BBO[2] | trigonal | 0.40 | 0.40 | 3.60 |
| LHPP[3] | monoclinic | 3.86 | 2.08 | 6.62 |
| IMLT | monoclinic | 7.63 | 7.18 | 2.75 |

**REFERENCES**

(1) G. Dhanaraj, M. Srinivasan, H. Bhat, H. Jayanna, S. J. Subramanyam, *Appl. Phys.*, 1992, **72**, 3464.

(2) D. Eimerl, L. Davis, S. Velsko, E. K. Graham, A. Zalkin, *J. Appl. Phys*., 1987, **62**, 1968.

(3) T. L. Chen, Z. H. Sun, C. Song, Yan. Ge, J. H. Luo, W. X. Lin, M. C. Hong, *Crystal Growth & Design*., 2012, **12**, 2673.