**Supplementary Information**

Gas-phase calculations of vibrational spectra (both IR and Raman) were done for L-leucine cation, Maleic acid anion, and LLHM dimer to provide a more reasonable assignment of experimental modes. Ions of L-Leucine, maleic acid, and their dimer were extracted from LLHM crystal structure from [1] and further were freely optimized at B3LYP/6-311+G(d,p) level of theory, providing vibrational calculations subsequently. Gaussian09 package was used for all calculations [2].

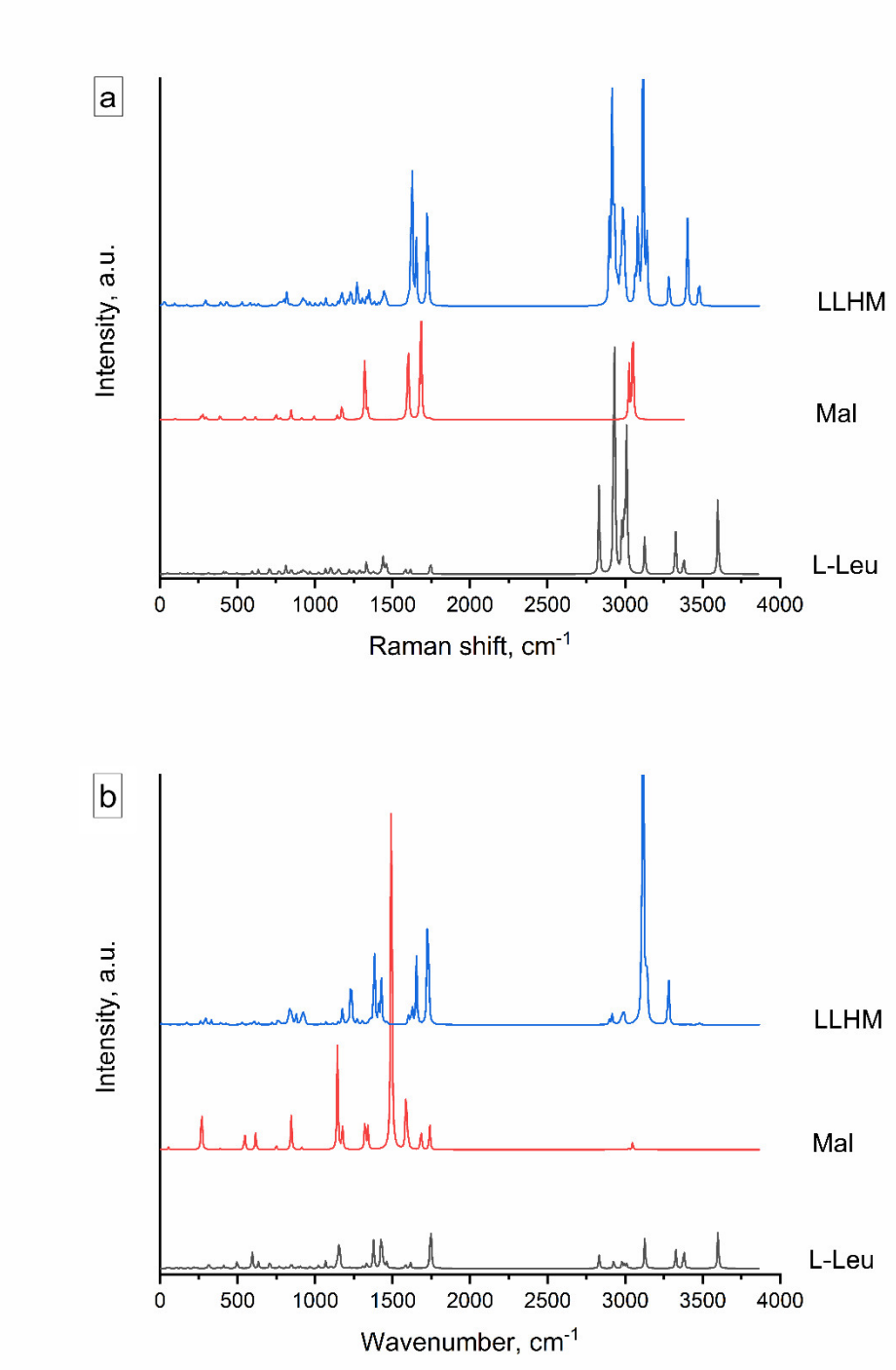


**Figure S1.** Gas phase calculated (**a**) Raman and (**b**) IR spectra of L-Leucine (Black), Maleic acid (Red), and L-Leucinium hydrogen maleate (Blue).

Low temperature Raman spectra were analyzed in three specific regions and no significant changes were found. Several lines split below 100K due to thermal motion decrease.

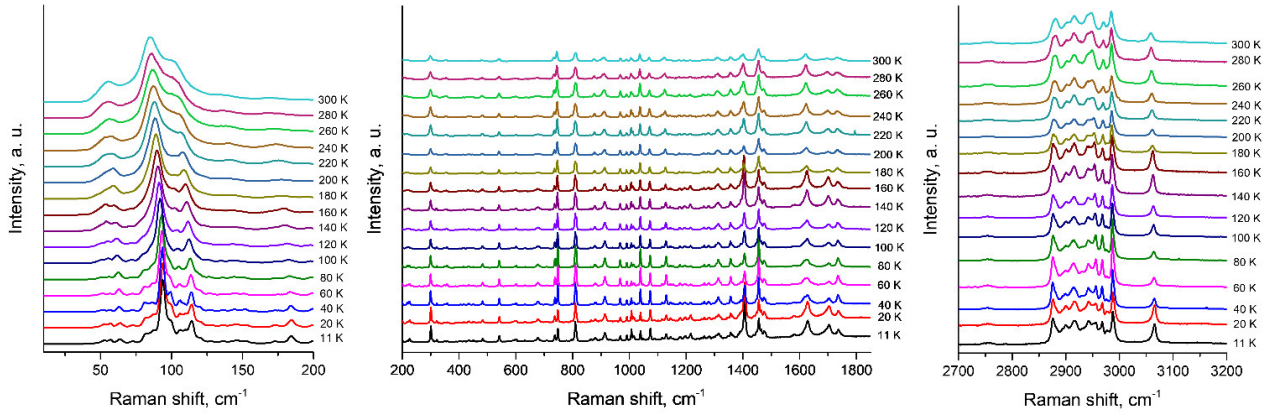


**Figure S2.** Experimental LLHM Raman spectra at low temperatures, showing no phase transition on cooling. Detailed three different regions of Raman spectra.

High pressure experiments were conducted at pressures where PTP preserves hydrostatic properties. Raman spectra of LLHM in PIP series at 0 GPa were recorded for isolated crystal (before being put in diamond anvil cell). No changes on pressure release were found.

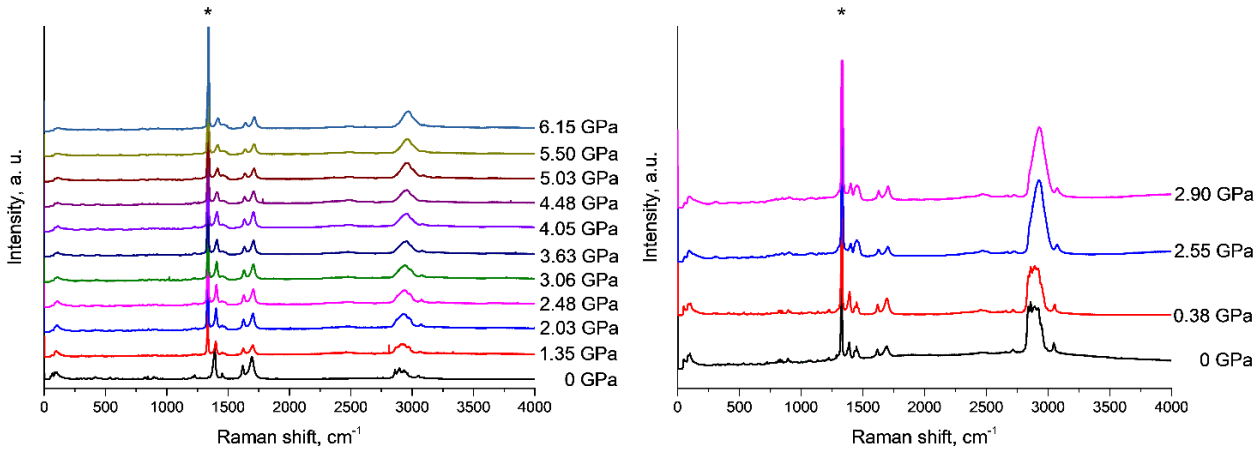


**Figure S3.** Raman spectra of LLHM at multiple pressures in PIP (**left**) and paraffin (**right**) PTM. Diamond peak from anvil cell is marked with *.

**References.**


1. Arkhipov, S.G.; Losev, E.A.; Nguyen, T.T.; Rychkov, D.A.; Boldyreva, E. V. A large anisotropic plasticity of L-leucinium hydrogen maleate preserved at cryogenic temperatures. *Acta Crystallogr. Sect. B Struct. Sci. Cryst. Eng. Mater.* **2019**, *75*, 143–151.
2. Frisch, M.J.; Trucks, G.W.; Schlegel, H.B.; Scuseria, G.E.; Robb, M.A.; Cheeseman, J.R.; Scalmani, G.; Barone, V.; Mennucci, B.; Petersson, G.A. et al. GAUSSIAN 09 2009.