Figure S1. Voids distribution in betaine at ambient pressure (different projections). Probe radius is 0.6 Å, grid is 0.1 Å. Void volume is 33.85 $Å^3$ (or 5.5% of the unit cell).

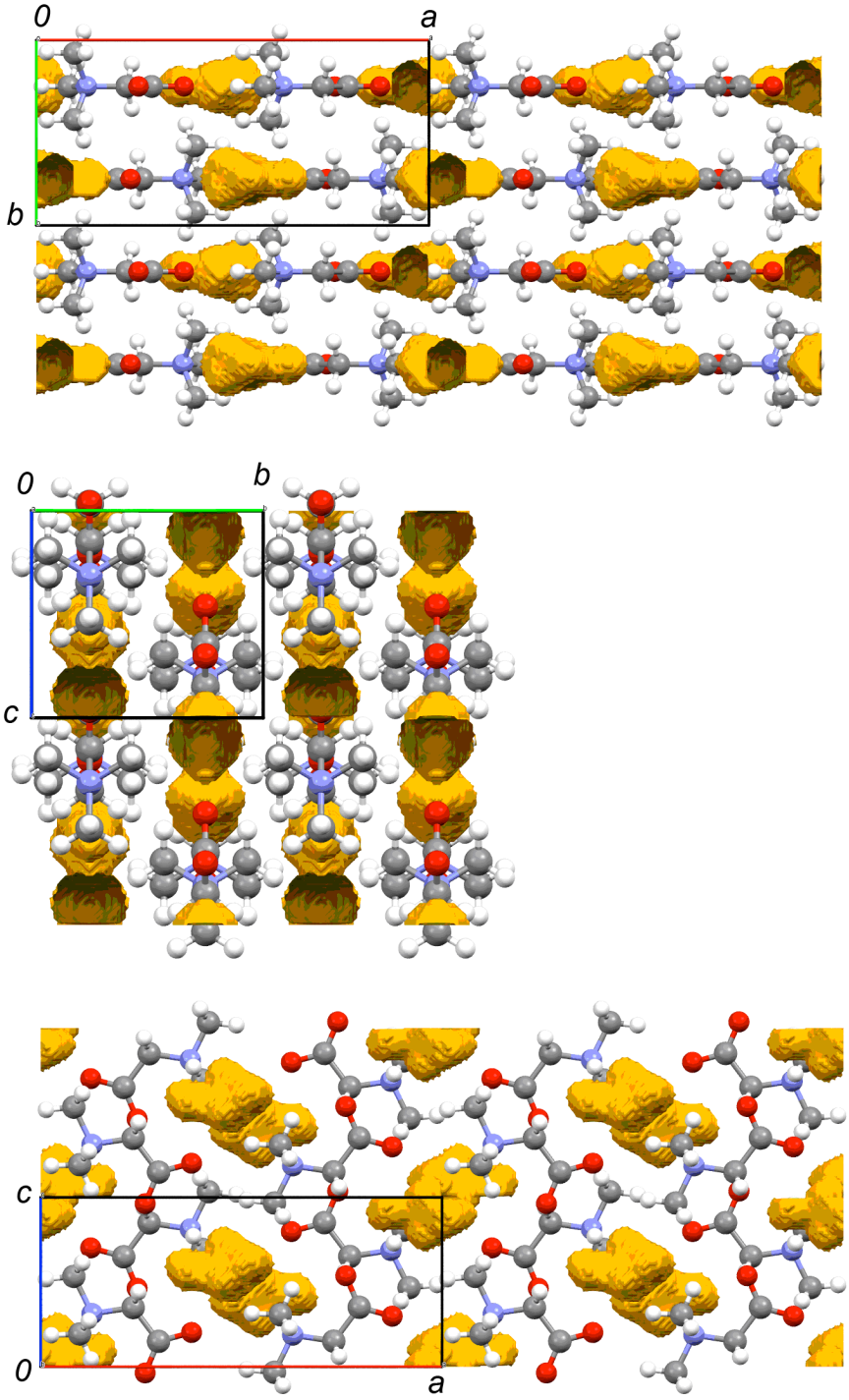

Figure S2. Photos of single crystal of DMG-II at 0.2 GPa (a) and 1.3 GPa (b) in DAC. Pressure medium: n-pentane/methylbutane.

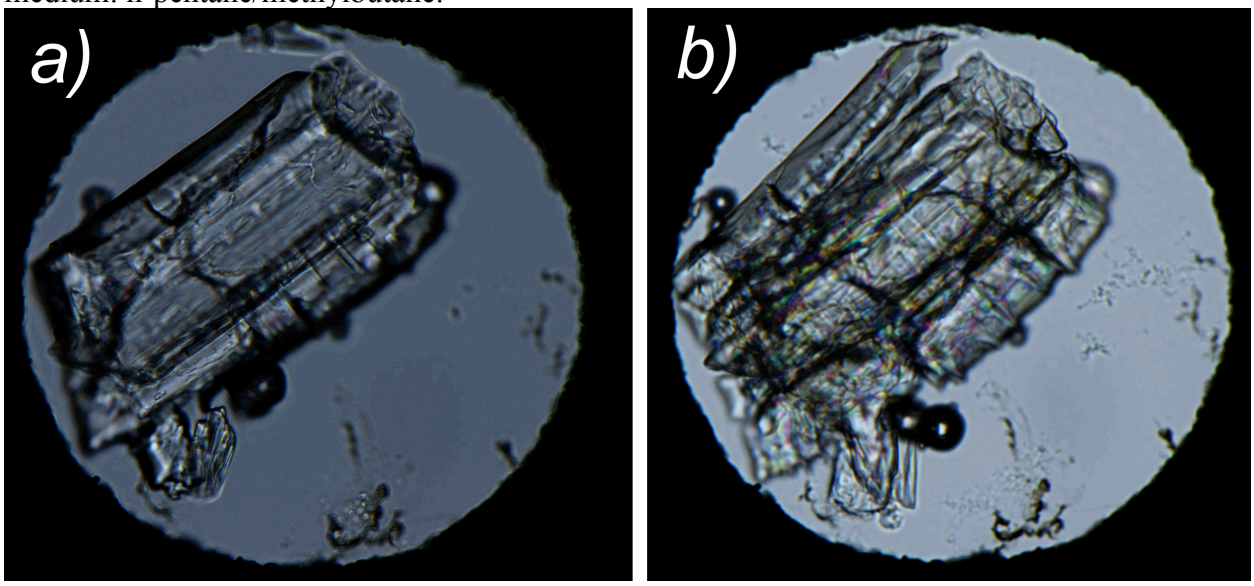

Figure S3. Reciprocal layer (***okl***) in betaine at 0.8 GPa (a) and 4.1 GPa (b)

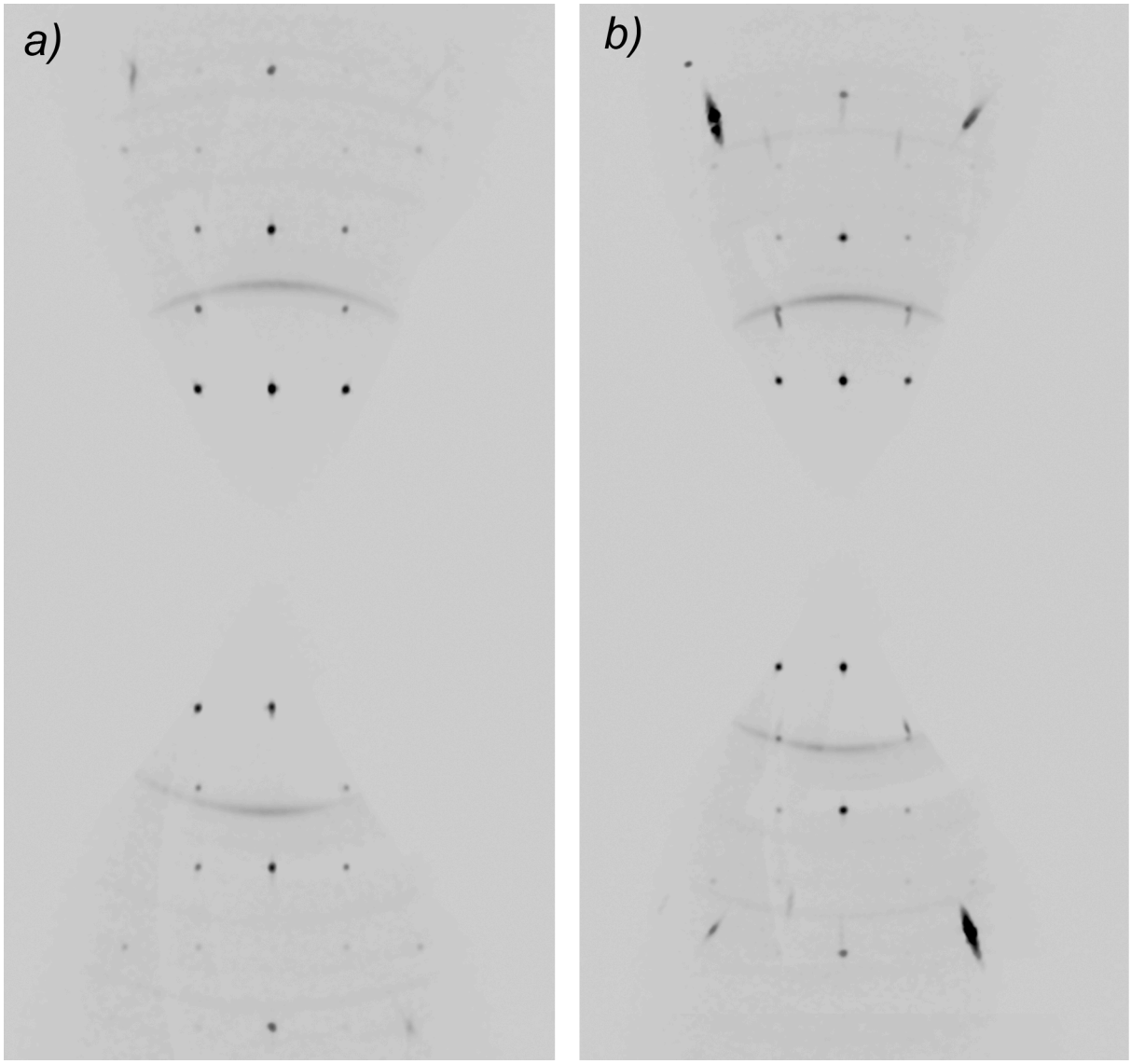

Figure S4. Voids distribution in betaine at 4.2 GPa (different projections).
Probe radius is 0.6 Å, grid is 0.1 Å. Void volume is 7.07 Å^3 (or 5.5% of the unit cell).

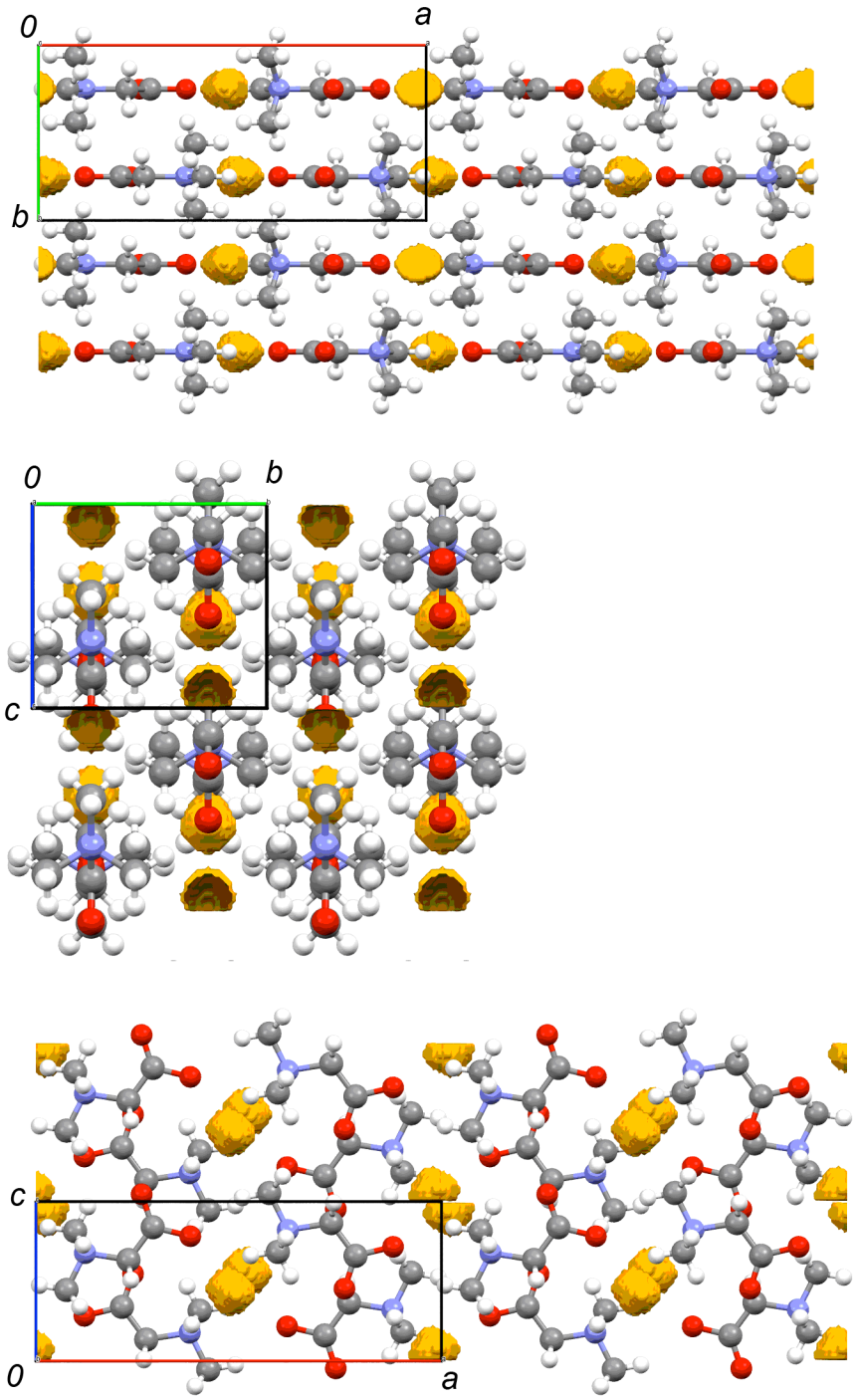

Figure S5. Relative volume change *vs*. pressure: α-polymorph of glycine (green rhombs), β polymorph of glycine (brown hexagons), γ-polymoprh of glycine (magenta triangles), L-alanine (blue stars), DL-alanine (orange triangles), betaine (red squares), sarcosine (black circles). Filled symbols correspond to increasing pressure, open ones - to decompression.