Crystal structures of racemic and homochiral forms of 2-phenylbutyramide (1) and 3-methyl-3phenylpyrrolidine-2,5-dione (2) were investigated in detail by a single crystal X-ray diffraction study. Absolute configurations of the homochiral forms of 1 and 2, obtained by chromatographic separation of racemates, were determined. It was revealed that racemate and homochiral forms of 1 are very similar in terms of supramolecular organization (H-bonded ribbons) in crystal, infrared (IR) spectral characteristics, and melting points. The presence of two different molecular conformations in homochiral forms of 1 allowed mimicking of crystal packing of the H-bonded ribbons in racemate 1. Two polymorph modifications (monoclinic and orthorhombic) comprising very similar H-bonded zigzag-like chains were found for the homochiral forms of compound 2 that were significantly different in terms of crystal structure, IR spectra, and melting points from the racemic form of 2. Unlike compound 1, homochiral forms of compound 2 have a higher density than the corresponding racemate which contradicts the Wallach rule and indicates that, in this case, homochiral forms are more stable than racemate forms.