Supplementary information

For the identification of D and L enantiomers we employed an adapted imagerecognition algorithm developed earlier [1]. For only partially ordered nuclei the search for specific molecular arrangement (such as the hydrogen bonded dimers and trimers described below) is limited to a tolerance allowed for the bond lengths. For the present work, the root-mean-square treshold of < 0.15 Å per atom to best distinguish between dimer and trimer motifs as illustrated in figure 2 of the main article. Note that in both the dimer and the trimer motifs all molecules are subject to further hydrogen bonding. The terms 'dimer' and 'trimer' shall hence refer to structural motifs (as part of a periodic arrangement) and not imply closed molecular entities.

The transition from dimer to trimer motifs requires molecular rotation and a slight shift of rows to reconnect the hydrogen bonded network without altering the total number of bonds. In all of our simulation runs this occurs at the boundaries of the (triangular) dimer domains – where one would expect a lowering of the transition barrier because of interfacial tension at the grain boundaries (fig. S1).

1 P. Duchstein, O. Hochrein, D.Zahn, Z. Anorg. Allg. Chem., 2009, 635, 649-652.



Fig. S1

Mechanism of the solid-solid transition at the boundary of a dimer domain (highlighted in red). Molecular rotation within a single row leads to hydrogenbonding in terms of trimer motifs (green arrows) and thus phase growth of the crystalline domain (highlighted molecules).