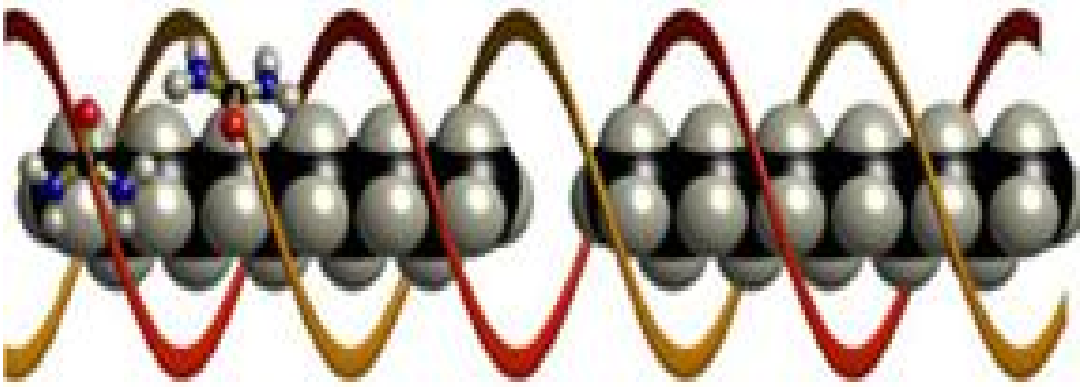
Phase Transitions and Pre-Transitional Effects in Aperodic Molecular Crystals



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Abstract

Supramolecular or self-assembled materials often result from a subtle balance of weak intermolecular interactions yielding to a large variety of ordered and disordered phases with many different physical, chemical or mechanical properties. Alkane urea inclusion compounds (UIC) are molecular composite structures where a "host" framework of urea molecules accommodates "guest" alkane molecules as long chains loosely held in a honey-comb like hexagonal network of adjacent channels. Most of the alkane-UICs are incommensurate or aperiodic, which means that the ratio of the host and guest periodicities is not a rational number M/N. As a consequence, very different types of short and long range ordering, as well as different phase transitions take place in these alkane-UICs just by changing the length of the alkane molecules. Even if these compounds are aperiodic by construction, periodicity can be recovered in superspaces of dimension greater than 3 and where full power of Fourier analysis can be used for crystallographic investigation.

Upon cooling, the aperiodic inclusion compound n-alkane/urea presents a hexagonal-to-orthorhombic group-subgroup phase transition at a critical temperature that increases the structure's superspace dimensionality from four to five. With changing the length we can have a family from this composite. With focusing the measurements on four composites n-nondecane/urea, n-nonane/urea, n-octane/urea, and n-dedacane/urea, new phases have been observed upon cooling down. The observation of an incommensurate diffraction pattern in urea inclusion compounds by neutron diffraction is also reported for the important composite n-nonadecane/urea. It reveals for the first time in these compounds the existence of all kinds of reflections expected for an intermodulated composite. The superspace analysis together with the temperature evolution of some characteristic reflections are presented.