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Simple Binary Molecular Mixtures at High Density: the Methane-Nitrogen System

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Mixing different molecular species at high density may yield weakly bound compounds or van der Waals solids. These novel solids are stabilized by the application of high pressure and differ in physical properties from solids formed by pure molecular species at comparable thermodynamic conditions. In this contribution, we address the importance in studying the miscibility in dense molecular mixtures and present results of the formation of binary methane-nitrogen compounds at low temperature and high pressure. Methane and nitrogen, with similar potentials and molecular sizes, are expected to be partly miscible in the condensed state. Indeed, binary van der Waals solid phases of methane and nitrogen do occur with the application of pressure. Using single crystal and powder X-ray diffraction with synchrotron radiation, and vibrational spectroscopy, the pressure-concentration phase diagram for this system has been explored from 1 to 16 GPa at room temperature. The existence of novel van der Waals solid phases for samples with concentrations above 10% (methane per volume) is demonstrated. For example, at 7.6 GPa and at room temperature, whereas pure nitrogen and methane exist in a cubic (Pm3n) and in a rhombohedral structure (R-3/m), respectively, our study indicates that a methane-nitrogen sample with ~ 40% methane by volume exhibits, under the same conditions, a novel phase with a tetragonal symmetry with lattice parameters $a \sim 11.9 \text{ \AA}$ and $c \sim 6.2 \text{ \AA}$. Other novel structures in methane-nitrogen samples with different concentrations under varying pressure conditions have also been observed and will be discussed.

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