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Pressure-induced homothetic volume collapse in silicon clathrates

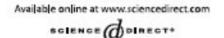
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PACS. 61.50.Ks — Crystallographic aspects of phase transformations; pressure effects. PACS. 61.48.+c — Fullerenes and fullerene-related materials.

Abstract. – The high-pressure properties of the Ba-doped silicon clathrate Ba₈Si₄₆ have been investigated combining X-ray diffraction and X-ray absorption spectroscopy. A pressure-induced isostructural phase transition associated with an important volume collapse takes place at 11.5–14 GPa. This transformation is characterized by the homothetic contraction of the silicon cages containing the Ba atoms. This transition is preceded by a change in the electronic structure at 5 GPa in good agreement with Raman spectroscopy observations (T. Kume et al., Phys. Rev. Lett., 90 (2003) 155503) that it is also of isostructural nature. The cage structure is preserved through the phase transitions allowing to obtain tetrahedral silicon with record interatomic distances as low as 2.13 Å. At the highest studied pressure of 49 GPa, the structure becomes irreversibly amorphous. The physical origin of the homothetic isostructural transitions is discussed.







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Synchrotron studies on silicon clathrates: Highly stable nanostructured materials

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Abstract

We have pushed the limits of X-ray absorption spectroscopy under high pressure in order to study the stability of the Ba intercalated silicon clathrate. EXAFS experiments at the Ba K-edge have been performed in the step by step mode up to a pressure of 14.5 GPa using the Paris-Edinburgh cell with sintered diamond anvils. XANES experiments at the Ba L-III edge were done in an energy dispersive set-up using hollowed diamond anvil cells up to a pressure of 20 GPa. Combining the obtained results with high pressure X-ray diffraction we demonstrate the high stability of the Ba intercalated silicon clathrates. Our conclusions could be extended to other clathrate systems.

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