Synthesis of Type-II Ge Clathrate Films with Various Film Thickness

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The inorganic clathrates (e.g., Si, Ge and Sn based) are widely investigated material due to its unique properties such as higher band gap energy, superconductivity, phonon glass and an electron crystal (PGEC) and so on [1]. These unique properties mainly arise due to flexibility of sp³ bonded structure of group IV elements. The clathrate structure consists of cage like open framework of face sharing polyhedra which can encapsulate guest species inside (alkali or alkaline earth) Such guest species are known to be actively affecting the electronic properties of clathrates. However, the clathrates have been prepared mostly in powder forms, therefore, only a few studies were reported on film sample [2]. Fabrication of good quality clathrate film still possesses major challenge. In this study, we focused on the film fabrications of type-II Ge clathrate on transparent and insulating substrate with different thickness of starting Ge films (0.2-1 µm) using a new setup namely "Portable Vacuum Evaporation and Annealing System" (pVEAS) [3].

Ge films (RF sputtering of Ge on sapphire substrate) were transformed into clathrate structure by vacuum evaporation of metallic Na during IR lamp annealing under high vacuum. The synthesized films were characterized using X-ray diffraction (XRD), Raman scattering and optical transmission studies. Scanning electron microscope (SEM) was also used to analyze the morphology of the films.

XRD patterns and Raman spectra confirmed the synthesis of type-II Ge clathrate films on sapphire substrate (Fig. 1). SEM images showed an impression of continuous growth (no visible cracks or pinholes). Optical studies indicated the increase of absorbance with the film thickness.



Fig. 1: Raman spectra of type-II Ge clathrate film prepared from Ge film of initial thickness 290 nm. **Acknowledgment:** The work reported here is supported by the Grants-in-Aid for Scientific Research (No 16K21072, JP17H03234, JP20K03820, J21H01365).

References:

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