Effect of Plasma in Molecular Beam Deposited Germanium-Sulfur Thin Films

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Due to their high carrier mobilities and extreme anisotropies, few-layer 2D materials are expected to lead the charge into utilization for advanced electronic and optical devices. Germanium Sulfide (GeS) stands out as an excellent candidate for such uses due its low toxicity[1]. In line with this, our group has been working on the thin-film growth of GeS. While we have obtained high quality crystals via the vapor transport method, significant challenges remained in controlling film thickness[2]. On the other hand, RF plasma-assisted molecular beam deposition (MBD) demonstrated excellent film thickness control, but crystal growth remains to be achieved[3,4]. In this work, we present our findings into the investigation of the relation between the germanium-sulfur bonding in MBD films and the RF plasma power of the sulfur cell attached Ar plasma unit.

In the experiment, separate-source germanium and sulfur were deposited within the MBD chamber onto silicon(111) substrates at room temperature for 30 min. A conventional solid-source K-cell was used for germanium evaporation while sulfur was evaporated from a cell equipped with an argon (Ar) plasma chamber at its outlet (Fig. 1) to enhance the reactivity of sulfur which typically exists as a stable eight-membered ring. The deposition of GeS_x was conducted by varying the output of the RF coil in the plasma cell as a parameter, and the thickness, composition and bonding of the resulting films was evaluated.

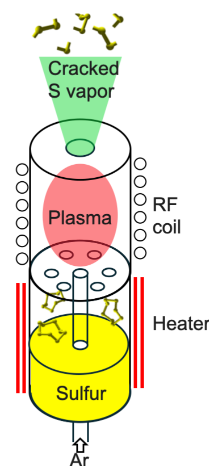

Fig 1. Illustration of the argon plasma (sulfur-cracking) unit

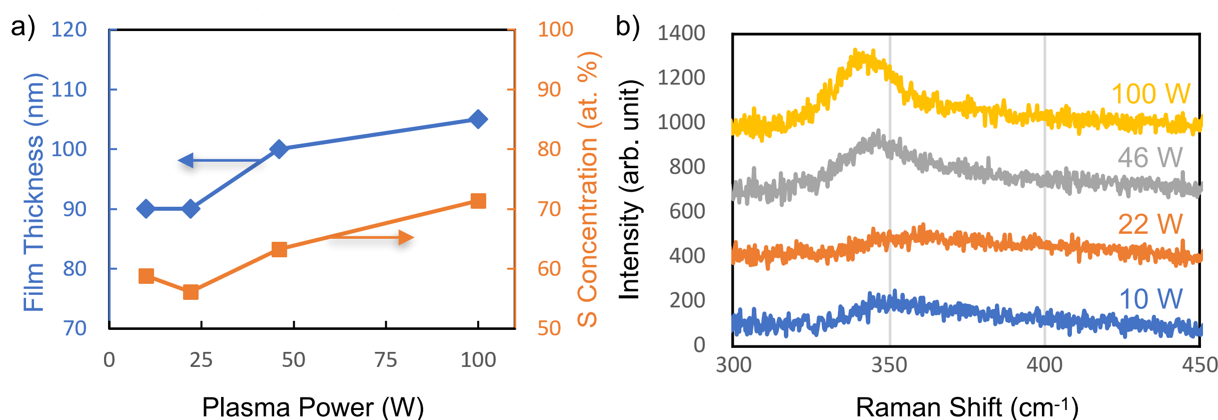

Fig. 2 (a) Film thickness and composition dependence against RF coil plasma power, (b) Background-subtracted Raman spectra for the deposited films

The deposited films' thickness and sulfur concentration are shown in Fig. 2(a). Interestingly -despite identical germanium and sulfur evaporation rates- the film thickness indeed increases when higher RF plasma power is applied. To explain this phenomenon, the sulfur concentration in the films was evaluated, revealing a trend of increased sulfur concentration with higher plasma power. This suggests that the rise in film sulfur integration is responsible for the observed increase in film thickness.

Consequently, Raman spectroscopy was used to examine the Ge-S bonding within the films. As the plasma power increased, a shift to lower wavenumbers in the peak position and increases in peak intensity were observed. These results indicate that the bonding of Ge-S in the films is affected by changes in plasma power. We believe this reflects the enhanced reactivity of sulfur under RF plasma conditions, which promotes its greater incorporation into the films.

References

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