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Growth of Two-Dimensional Layers on Graphite Substrates

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Abstract. The work is devoted to studying the temperature dependence of epitaxial processes during the synthesis of Ge and Si on highly oriented pyrolytic graphite. In this work, the epitaxial growth of silicon and germanium is studied directly during the deposition of material onto the surface of highly oriented pyrolytic graphite using high-energy electron diffraction. In addition, the obtained samples were studied by Raman spectroscopy and scanning electron microscopy. A wide range of deposition temperatures from 100 to 800 °C is considered and temperature ranges are determined for various growth modes of silicon and germanium on highly oriented pyrolytic graphite.
Keywords: molecular beam epitaxy, reflection high-energy electron diffraction, silicon, germanium, silicene, germanene.

1. Introduction

Two-dimensional silicon (silicene) and germanium (germanene) have attracted special attention from researchers in recent years. The main method for creating graphene-like materials is their epitaxial growth on lattice-matched substrates. At the same time, highly oriented pyrolytic graphite (HOPG) and graphene are some of the promising substrates for growing silicene and germanene [1, 2]. However, to date, the processes occurring during the epitaxial growth of silicon and germanium on the surface of such substrates have been poorly studied [3, 4]. The purpose of the work is to study the dependence of epitaxial processes on temperature during the synthesis of Ge and Si on HOPG.

2. Results

The deposition of epitaxial thin layers took place in an ultra-high vacuum using molecular beam epitaxy. At the same time, the surface morphology was controlled by the reflection high-energy electron diffraction method. The diffraction pattern of a clean HOPG surface is shown in Fig.1.

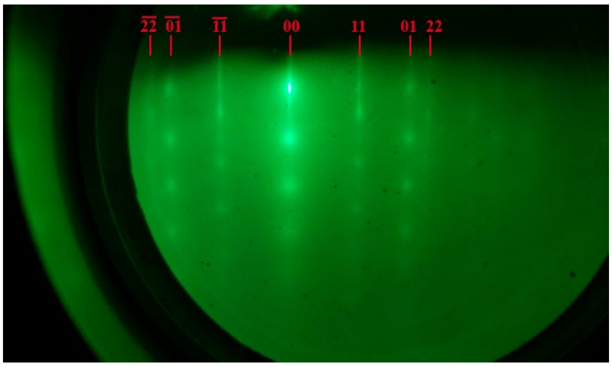

Fig. 1. Diffraction pattern corresponding to a clean HOPG substrate surface.

It has been shown that at temperatures close to room temperature, materials grow amorphously, and at high temperatures polycrystalline growth is observed. It has been established that in the temperature range of 250–400 °C the crystal structure of germanium during 1 monolayer repeats the structure of graphite. For silicon, this range is at higher temperatures and starts at 650 °C. Then reflections of the 1/N type appear in the diffraction pattern (N(Si) = 1.56 and N(Ge) = 1.62). Such values correspond to the lattice constants of silicene and germanene and may indicate the presence of areas of graphene-like 2D phases during epitaxial deposition of silicon and germanium onto the surface of highly oriented pyrolytic graphite. The corresponding diffraction patterns are shown as an example in Fig.2 and 3. The diffraction patterns in Fig.2 and 3 were selected at those temperatures at which the appearance of the 1/N reflection is observed.

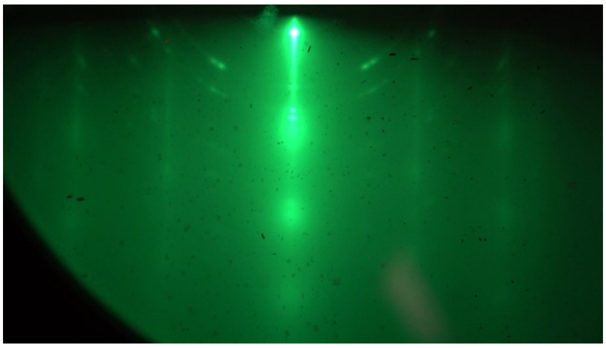

Fig. 2. Diffraction pattern corresponding to a HOPG surface with 10 ML Si on the surface. T = 650 °C.

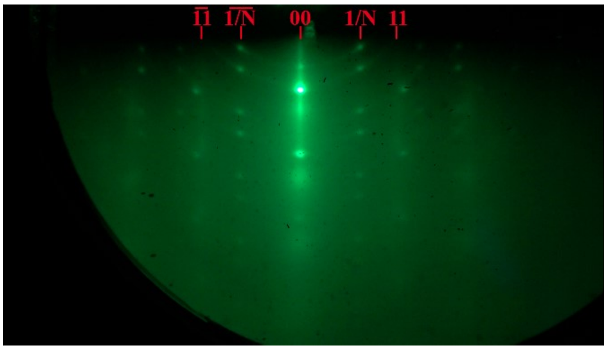

Fig. 3. Diffraction pattern corresponding to a HOPG surface with 10 ML Ge on the surface. T = 400 °C.

As Si or Ge materials are deposited, the 1/N reflections intensify, and the reflections from the HOPG substrate gradually fade. At temperatures below the specified temperature ranges, the intensity of the diffraction pattern decreases as deposition proceeds without the formation of additional reflections.

The growth pattern of one monolayer of germanium and silicon on HOPG is the same, judging by scanning electron microscopy (SEM) images (Fig. 4 and 5).

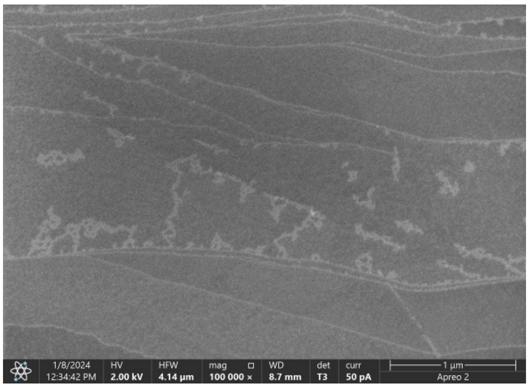

Fig. 4. SEM image of the surface after deposition of 0.9 ML silicon on HOPG.

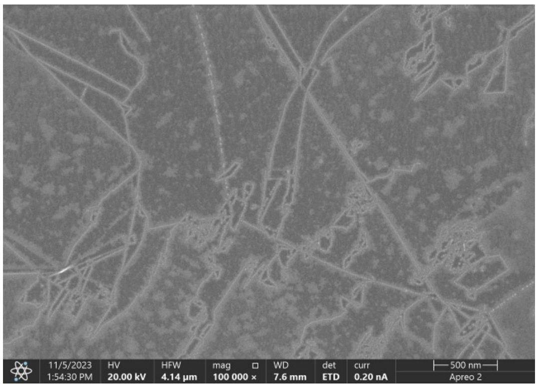

Fig. 5. SEM image of the surface after deposition of 0.9 ML germanium on HOPG.

As can be seen from Fig.4 and 5, the HOPG surface is covered with a flat layer of Si (Fig. 4) and Ge (Fig. 5) materials. At the edges of the steps, in some places there are accumulations of sprayed materials, but on the terraces there are only two-dimensional islands. It can be concluded that a two-dimensional islands growth regime takes place.

Additionally, the structures were studied by Raman spectroscopy. It was determined that the 1ML Ge/HOPG structure has peaks different from HOPG at 296, 300, and 304 cm^{-1}, and the 1ML Si/HOPG structure – at 1310 cm^{-1}.

3. Conclusion

The results of the work prove the importance of temperature choice for creating two-dimensional Ge and Si layers on HOPG. In addition, each material has its own temperature range at which the growth of a crystalline two-dimensional structure is possible. If, during the synthesis of Ge on HOPG, the 1/N reflection on diffraction patterns appears in the range from 250 °C to 470 °C, then for Si it begins only at 650 °C. It is important to note that 1/N appears only after the first monolayer. Therefore, SEM images and Raman spectra were obtained for 0.9 ML Ge and Si on HOPG. It has been established that there are two-dimensional islands on the surface that contribute to the Raman spectra (Ge – 296, 300, and 304 cm^{-1}; Si – 1310 cm^{-1}). The results obtained can be used to develop technology for growing silicene and germanene. In addition, the presented results will also be valid for the epitaxial growth of silicon and germanium layers on graphene.

Acknowledgement

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4. References

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