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論　文　要　旨
Thesis Abstract

(yyyy/mm/dd) 2022 年　9 月　1 日

※報告番号	甲第３２２号	氏　名 (Name)	Narin Sunthornpan
主論文題名	(Title) Metal-induced crystallization of germanium thin films		

内容の要旨　(Abstract)

Germanium (Ge) is one of the most intensively studied semiconductor materials because of its superior properties, such as higher carrier mobility and smaller energy bandgap compared to silicon (Si). These properties are advantageous in applications such as thin-film transistors, solar cells, next generation MOSFETs, etc. Nevertheless, because of its higher cost, fabricating high quality crystalline Ge thin films is a key to realizing these devices. Moreover, it would be advantageous if the low-temperature process is developed to fabricate these films on glass and plastic films. Metal-induced crystallization (MIC) technique is a rapidly emerging technique because it can crystallize Ge films at temperatures much lower than other methods like solid phase crystallization (SPC). Moreover, the method is simple and more suitable for large area processing compared to the laser annealing process. In MIC, to decrease the crystallization temperature, the introduction of metal layers such as aluminum (Al), silver (Ag) and gold (Au) is required as catalysts. Especially by using Au as a metal catalyst, the crystallization of amorphous Ge (a-Ge) films can be realized at a lower temperature (<300°C), which is low enough to fabricate high-quality crystalline Ge (c-Ge) thin films on plastic substrates such as polyimide films.

In the MIC method, an a-Ge/Au bilayer is annealed to crystallize the a-Ge layer. During the process, the Ge and Au layers exchange their positions. In most studies, a thin oxide layer is inserted between the a-Ge and Au layers to promote layer exchange. Nevertheless, this insert layer suppresses interlayer diffusion and a long annealing time, ~100 hours, is necessary. To solve this problem, the crystallization mechanism of Ge in the Au-induced crystallization method without an insert layer has been examined in this thesis. It is found that a layer exchange crystallization occurs even without an insert layer. The crystallization mechanism is clarified, and it is shown that by optimizing the process, it is possible to obtain a continuous c-Ge film with a smooth surface by annealing at 170°C for 1 hour. These findings will be useful for the fabrication of high-quality Ge thin films on various inexpensive flexible substrates which can be used as seeding layers for next generation solar cell application.

In chapter 1, the history of the MIC process is reviewed, and the aim of the thesis is explained.

In chapter 2, experimental methods adopted in the present study is introduced together with their operation mechanisms.

In chapter 3, we investigated the crystallization behavior of Ge thin films by Au catalysts without an insert layer. By annealing an a-Ge/Au bilayer up to 220°C, it is found that a layer-exchange type crystallization of Ge is possible even without an insert layer. As for the Au thickness dependence, it is found that the best Ge crystallinity is achieved with an initial Au

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layer as thin as 9 nm. This behavior seems to be brought about by the substrate which promotes heterogeneous nucleation of crystalline Ge. A higher (111) orientation is also realized for thinner Au samples. These findings are encouraging in terms of the smaller consumption of rare metals like Au.

The effect of initial a-Ge layer thickness on the crystallization behavior is also examined. It is found that the initial a-Ge layer thickness affects the morphology of the resulting c-Ge thin films. A double c-Ge layer structure has been confirmed. The bottom c-Ge layer has a thickness close to the original Au layer and has a better crystal quality compared to the top c-Ge layer. This morphology resembles that of crystalline semiconductor thin films obtained by annealing Al/Si and Al/Ge systems. It is possible to control the surface coverage of these layers by adjusting the initial Au and a-Ge thicknesses. Nearly 97% of the substrate surface is covered by the bottom c-Ge layer with a small amount of top c-Ge layer by annealing an a-Ge(46nm)/Au(29nm) bilayer at 220°C. The resulting ~30nm thick Ge film shows a hole mobility of as high as ~85 cm^2/Vs reflecting a high coverage.

In chapter 4, the crystallization mechanism without an insert layer has been investigated in detail. The results show that the Ge atom diffuse from the a-Ge layer into the Au layer and nucleation of c-Ge occurs inside the Au layer. Lateral growth of c-Ge proceeds by the Ge supply through the Au layer. This explains why the bottom c-Ge layer has a same thickness as the original Au layer. As the c-Ge layer grows on the substrate, Au is pushed up to the top layer and layer exchange completes. The top c-Ge layer starts to nucleate at a higher temperature compared to the bottom layer. A poor crystal quality implies a different growth mechanism for the top layer. A small amount of Au diffused into the a-Ge layer could have crystallized Ge without layer exchange.

In chapter 5, the findings in chapters 3 and 4 have been utilized to develop an efficient process to obtain a continuous c-Ge layer with a small surface roughness at low temperature. In chapter 4, it is found that the top layer starts to nucleate at a higher temperature compared to the bottom layer. This implies that by annealing at a low temperature for a certain period of time, it might be possible to increase the coverage of the bottom layer while suppressing the nucleation of the top layer. By annealing an optimized thickness bilayer, a-Ge(46nm)/Au(29nm), at 220°C with a heating rate of 0.5°C/min, the coverage of as high as ~99% for the bottom c-Ge layer with a small amount of top layer has been obtained. This result supports the assumption that it is possible to grow only the first layer by adjusting the bilayer thickness and annealing at a low temperature for a certain period of time, which is enough to complete the bottom layer. Moreover, to obtain a (111) oriented film, a thinner bilayer, a-Ge(18nm)/Au(10nm) has been adopted. By annealing this bilayer at 170°C for 1 hour, a highly (111) oriented c-Ge layer has been obtained with a small amount of top layer.

Chapter 6 includes the summary of the thesis and future prospects.

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