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Formation and Structural Characterization of $Cu_{15}Si_4$ Thin Films on Si(111) by RFMS and DCMS Magnetron Sputtering

Copper silicide nanofilms were synthesized via RF and DC magnetron sputtering (MS). RFMS deposition of Cu onto Si(111) at 467°C formed a heteroepitaxial $Cu/Cu_{15}Si_4/Si$ structure, with a 75 nm $Cu_{15}Si_4$ layer beneath a 130 nm Cu overlayer. DCMS-prepared Cu/Si(111) films were vacuum-annealed at 527°C for 90 minutes, also yielding $Cu_{15}Si_4$. Film thickness and morphology were characterized by scanning electron microscopy (SEM), and phase formation was confirmed by EDS. The results highlight the influence of Cu crystal size and substrate temperature on silicide formation, demonstrating the potential of copper silicides for enhancing performance in MOS transistors and high-speed integrated circuits.

Keywords: Copper silicides; Thin films; Plasma sputtering; Magnetron sputtering
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1. Introduction

There are many methods for the production of nano-sized Cu thin films and copper silicide films, which are widely used in electronic devices due to their high electromigration resistance and high electrical conductivity [1,2]. Cu and its silicide films are widely used in many fields, such as contacts in solar cells [3], metal-oxide-semiconductor transistors [4], and high-speed integrated circuits (ICs) [5,6]. Cu and its silicide films are commonly produced by various methods, including direct current magnetron sputtering (DCMS) [7-10], middle radio frequency magnetron sputtering (RFMS) [11-13], high-power pulsed magnetron sputtering (HiPIMS), molecular beam epitaxy (MBE) [14], and reactive pulsed laser deposition (RPLD) [15]. Cu and its silicide films have been reported in many articles. However, most of them used glass substrates to form contact films, and several papers have treated silicon substrates [16]. Copper and copper silicide thin films attract considerable attention due to their important role in modern microelectronic and nanoelectronic devices. Owing to their high electrical conductivity, relatively low resistivity, and good compatibility with silicon technology, Cu-based films are widely used as interconnects, contact layers, and diffusion barriers in integrated circuits and semiconductor devices. In particular, copper silicides are considered promising materials for ohmic contacts because of their favorable electrical properties and thermodynamic stability on silicon substrates.

The Cu–Si system has been extensively studied, and several copper silicide phases have been reported depending on the processing conditions, such as temperature, deposition method, and film thickness. Among these phases, $Cu_{15}Si_4$ is known as a thermodynamically stable silicide that can form through solid-state reactions between Cu and Si at elevated temperatures. Previous fundamental studies have demonstrated that Cu atoms actively diffuse into the silicon lattice when sufficient thermal energy is provided, leading to the formation of copper silicide phases at the Cu/Si interface. However, the exact formation mechanism and phase selectivity strongly depend on the deposition technique and substrate conditions.

Various physical vapor deposition (PVD) techniques have been employed to fabricate Cu and Cu–Si thin films, including direct current magnetron sputtering (DCMS), radio frequency magnetron sputtering (RFMS), molecular beam epitaxy, and pulsed laser deposition. Most reported studies focus on optimizing deposition parameters to control film thickness, grain size, and surface morphology. Nevertheless, a significant portion of these works has

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been carried out on amorphous or glass substrates, where the influence of crystallographic orientation and interfacial reactions with silicon is absent. As a result, the structural evolution and phase formation behavior of copper silicides on single-crystal silicon substrates remain insufficiently explored.

In the case of Cu films deposited on crystalline Si substrates, substrate temperature plays a crucial role in determining diffusion kinetics and phase evolution. Several studies have reported copper silicide formation after post-deposition annealing; however, fewer investigations address silicide formation directly during deposition, particularly under ion-assisted conditions. Moreover, comparative studies analyzing the differences between direct current magnetron sputtering (DCMS) and radio frequency magnetron sputtering (RFMS) modes under identical experimental conditions are scarce. This lack of systematic comparison limits a comprehensive understanding of how ion energy, plasma characteristics, and substrate temperature jointly influence copper silicide formation.

Another limitation of existing studies is related to phase identification. In many reports, conclusions regarding silicide formation are based mainly on compositional analysis, while a detailed correlation between elemental composition and crystallographic phase confirmation is often missing. Since techniques such as EDS provide information on elemental ratios but cannot uniquely identify crystal structures, reliable phase determination requires complementary X-ray diffraction analysis with proper indexing of diffraction peaks.

In this context, the present work aims to address these unresolved issues by investigating the formation and structural evolution of Cu–Si thin films deposited on single-crystal Si(111) substrates using a solid-state ion-plasma method. Copper films were deposited using both direct current magnetron sputtering (DCMS) and radio frequency magnetron sputtering (RFMS) modes under controlled conditions, including room temperature and elevated substrate temperature. Special emphasis is placed on the role of deposition mode and substrate temperature in promoting Cu diffusion into silicon and inducing the formation of the $Cu_{15}Si_4$ silicide phase. The phase composition is systematically analyzed using X-ray diffraction (XRD) with indexed reflections, supported by compositional analysis, allowing for a reliable confirmation of $Cu_{15}Si_4$ formation. The results provide new insights into the controlled synthesis of copper silicide layers on silicon substrates and contribute to a deeper understanding of ion-assisted silicide formation processes relevant to microelectronic contact applications.

Despite extensive studies on copper and copper silicide thin films, several important aspects remain insufficiently explored. Most reported works focus on films deposited on glass or amorphous substrates, while systematic investigations on single-crystal silicon substrates are still limited. In particular, the influence of deposition mode and substrate temperature on the formation pathways, phase composition, and microstructural evolution of copper silicides has not been fully clarified. Moreover, comparative studies addressing the differences between direct current magnetron sputtering (DCMS) and radio frequency magnetron sputtering (RFMS) techniques under identical experimental conditions are scarce. These unresolved issues highlight the need for a detailed investigation of Cu–Si thin film systems, with special emphasis on phase formation mechanisms and structural properties relevant to microelectronic applications.

2. Materials and Methods

Copper silicide nanofilms were fabricated using two different modes of the EPOS PVD DESK PRO magnetron sputtering system [9-11]. Argon gas with a purity of 99.99% was used. The working chamber pressure was set at 10^{-6} Torr. Using the RFMS method, copper silicide thin films were deposited onto a monocrystalline silicon substrate heated to 467°C. This was achieved by sputtering a copper target at a frequency of 100 kHz and with a duty cycle (D) of 70%, for the Cu target was 18 Å/s. The thickness of the resulting heteroepitaxial $Cu/Cu_{15}Si_4/Si$ film was measured using TESCAN MIRA3 scanning electron microscope (SEM). The elemental composition of the obtained samples was determined using energy-dispersive X-ray (EDS) spectroscopy at Peter the Great St. Petersburg Polytechnic University (Saint Petersburg, Russian Federation). Furthermore, the formation of a $Cu_{15}Si_4$ film was confirmed by thermally annealing Cu/Si(111) nanofilms, deposited via the DCMS method, in a vacuum at 800 K for 1.5 hours. The surface morphology of the obtained silicide films was examined using an Olympus LEXT™ OLS5100 laser confocal microscope. The phase structure of the Cu-Si system was studied using a SHIMADZU 6100 X-ray device.

3. Results and Discussion

During the experiments, it was found that the formation of copper silicide films depends on the substrate temperature during ion-plasma sputtering and the subsequent heating temperature. We describe two methods using a magnetron sputtering device. Initially, Cu was sputtered onto the Si surface by the ion-plasma method using the DCMS method at room temperature, and amorphous Cu films were formed, as confirmed by SEM (Fig. 1a), a $Cu_{15}Si_4$ film was formed by heating in a vacuum at a temperature of 800 K for 90 minutes. According to the results obtained using SEM (Fig. 1b), the islands on the surface of the film, as a result of subsequent heating, indicate the formation of a $Cu_{15}Si_4$ silicide film as a result of the crystallization of silicon.

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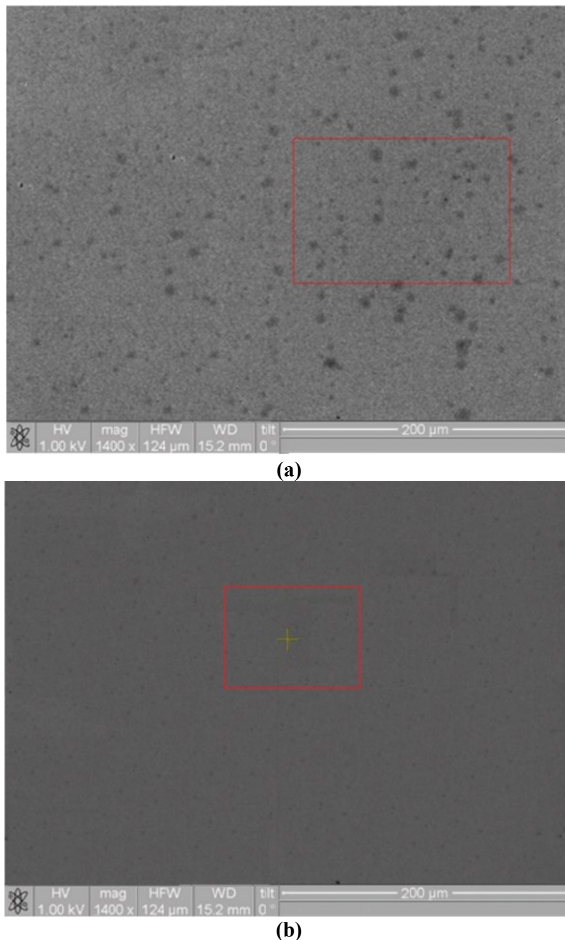
The thickness of the resulting heteroepitaxial $Cu/Cu_{15}Si_4/Si$ film was measured using SEM (Fig. 3). The formation of the copper silicide film depends on the copper crystal size and substrate temperature, and at 467 °C, a 75 nm thick $Cu_{15}Si_4$ film was formed under a 130 nm thick copper layer. To analyze these processes, the structure was studied using X-ray diffraction.



(a)

(b)

Fig. (1) SEM images of the copper film formed by DCMS on a Si(111) surface: (a) before annealing and (b) after annealing

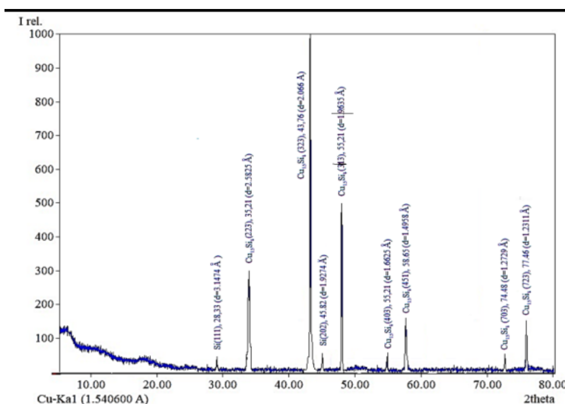


Fig. (2) XRD pattern of the Cu–Si thin film deposited on the Si substrate, with indexed diffraction peaks corresponding to the cubic $Cu_{15}Si_4$ phase

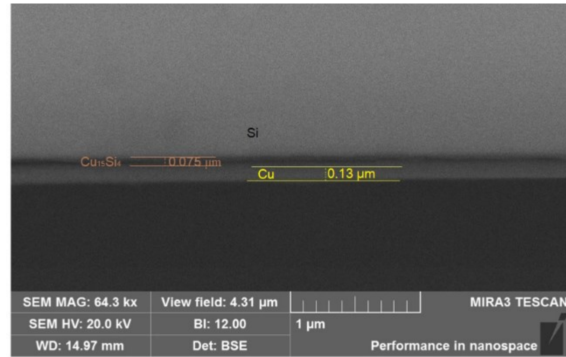


Fig. (3) SEM image of $Cu/Cu_{15}Si_4/Si$ layer by RFMS method

To further confirm the results, we performed energy-dispersive X-ray spectroscopy (EDS) analysis on the sample to determine the elemental composition and mass fraction of the thin film as shown in Fig. (4). The elemental composition of the Cu–Si thin films obtained under different deposition conditions was analyzed using EDS.

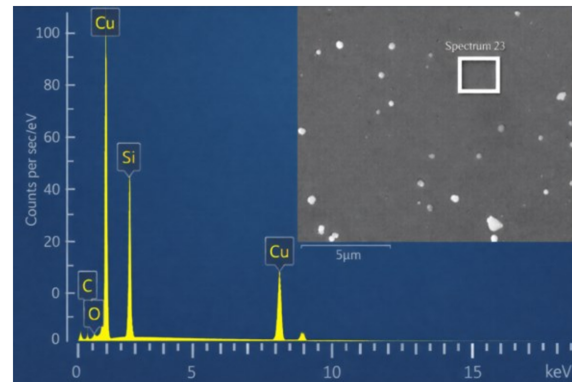


Fig. (4) EDS spectrum of the $Cu_{15}Si_4$ thin film formed on a Si(111) substrate, confirming the presence of Cu and Si elements. The inset SEM image illustrates the surface morphology and the selected region for elemental analysis

The compositional results for the RFMS sample are presented in table (1), while those for the DCMS-annealed sample are summarized in table (2). As shown in table (1), the RFMS-deposited sample contains 78.87 at.% Cu and 21.13 at.% Si. The calculated atomic ratio Cu/Si of about 3.73 is in excellent agreement with the theoretical stoichiometric ratio of $Cu_{15}Si_4$ (3.75), confirming the formation of this silicide phase. In contrast, Table (2) shows a different elemental distribution (49.69 at.% Cu and 45.60 at.% Si), along with small amounts of carbon and oxygen, which may originate from surface contamination or exposure to air. The deviation from the ideal $Cu_{15}Si_4$ stoichiometry suggests that phase composition depends strongly on the deposition mode and post-deposition treatment. The comparison between tables (1) and (2) indicates that substrate temperature and processing conditions play a

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crucial role in controlling the Cu–Si reaction and the resulting phase composition.

Table (1) Elemental composition of Cu–Si thin film deposited by RFMS

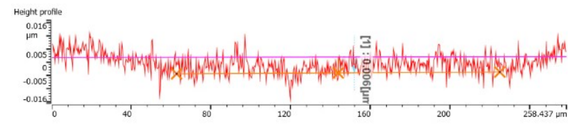
Element	mass. %	atom. %
Si	10.61	21.13
Cu	89.39	78.87
	100.00	100.00

Table (2) Elemental composition of Cu–Si thin film prepared by DCMS and annealed

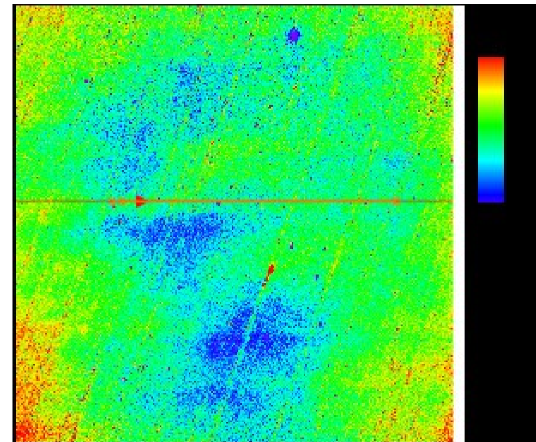
Element	Mass %	σ-mass %	Atom %
C	0.61	0.01	1.21
O	1.24	0.02	3.50
Si	29.04	0.09	45.60
Cu	69.11	0.10	49.69
Summation:	**100.00**		**100.00**

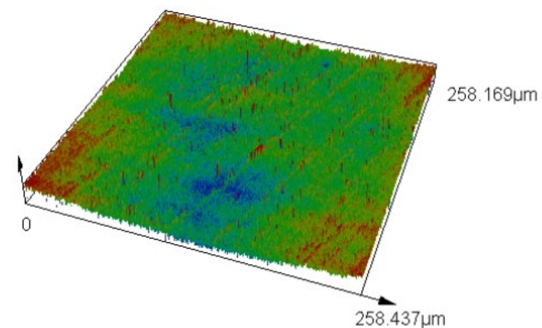
According to the results of the study, it was found that the formation of Cu and its silicide nanofilms from a copper target using the DCMS and RFMS methods of the magnetron device depends on the initial temperature of the substrate and the subsequent heating temperatures of the films. The optimal temperature for crystallization of the films was found to be 467 °C. The surface morphology of the $Cu_{15}Si_4$ thin film was investigated using an Olympus LEXT™ OLS5100 laser confocal microscope. The scanned area of approximately 258×258 µm² revealed a relatively uniform and continuous surface without pronounced cracks or defects. The corresponding 2D and 3D topographical images in Fig. (5) indicate that the height variations are confined within the nanometer scale. The height profile analysis shows that the average surface roughness (R_a) is about 6 nm. Such a low roughness value suggests good surface uniformity and indicates that the formed $Cu_{15}Si_4$ layer is suitable for microelectronic applications, where smooth interfaces are essential for minimizing contact resistance and ensuring reliable device performance.

X-ray analysis of the deposited Cu films confirms the formation of high-quality metallic copper layers, which serve as a precursor for copper–silicide formation during subsequent interaction with the silicon substrate. Under appropriate deposition conditions, particularly at elevated substrate temperatures, Cu atoms actively diffuse into the Si lattice, leading to the formation of thermodynamically stable copper silicide phases. In this work, special attention is paid to the influence of deposition mode (RFMS and DCMS), substrate temperature, and film thickness on the formation mechanism, phase composition, and surface morphology of Cu–Si thin films. The study focuses on the structural evolution and phase identification of the $Cu_{15}Si_4$ silicide formed by the solid-state ion-plasma method, providing insight into the controlled synthesis of copper silicide layers on Si substrates for microelectronic contact applications.



(a)


(b)



(c)

Fig. (5) Surface morphology of $Cu_{15}Si_4$ thin film formed by the DCMS method (a) Surface roughness image, (b) 2D image, and (c) 3D image

4. Conclusion

Copper silicide thin films were formed by sputtering copper onto a single-crystal silicon surface heated at 467°C. Additionally, $Cu_{15}Si_4$ film was formed by thermally heating Cu/Si(111) nanofilms in a vacuum at 527°C for 1.5 hours. The formation of the copper silicide film depends on the copper crystal size and substrate temperature. XRD analysis confirmed the formation of the cubic $Cu_{15}Si_4$ phase, supported by EDS compositional results consistent with its stoichiometry. The results demonstrate that substrate temperature and deposition mode play crucial roles in controlling silicide phase formation.

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