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Effects of source materials on fabrication of β-$FeSi_2$ thin films by RDE method

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The reactive deposition epitaxial (RDE) growth method has been employed extensively in the β-$FeSi_2$ thin film growth. It has been already clarified also that the Fe/Si ratio of source materials affects on the quality of deposited β-$FeSi_2$ thin films by ion beam sputter deposition under high-vacuum condition. On the other hand, to provide methods capable of depositing a high-quality β-$FeSi_2$ thin films inexpensively are important as well. In this study, we used a conventional vacuum deposition system and a few different kinds of iron silicides of Fe_2Si, α-$FeSi_2$ and ε-FeSi, which each contains Si, as the source materials in the RDE growth. We found that the depositing Fe_2Si onto heated Si substrate helps improving the film flatness and the electric properties of the obtained β-$FeSi_2$ thin films compared to that of Fe-deposited films, even at the relatively low vacuum depositing.

1. Introduction

Orthorhombic iron disilicide β-$FeSi_2$ is a semiconductor with the band gap energy at about 0.8 eV [1-4] and composed of Fe and Si that are abundant in earth's crust. Thus, β-$FeSi_2$ is expected to be a promising material for near-infrared light emitter, detector or solar cells. In terms of thin film growth, the reactive deposition epitaxial (RDE) growth, which deposit pure Fe onto heated Si substrate so that the reaction of deposited Fe and Si supplied from substrate proceed, was studied actively in the early researches on β-$FeSi_2$ thin films growth [5-7]. However, the RDE method is not appropriate to fabricate the thickness of films greater than 100 nm due to the epitaxial β-$FeSi_2$ film that easily aggregates into β-$FeSi_2$ islands and the interface between the grown β-$FeSi_2$ islands and Si substrate becomes relatively rough. To overcome the drawbacks of the RDE method, Yamaguchi *et al.* controlled the supply of Fe and Si atoms in the silicidation process by employing various chemical compositions of Fe-Si targets by means of ion beam sputter deposition (IBSD) under ultra-high vacuum, in which the sputtered atoms have relatively higher energy than that of thermally vaporized atoms [8], and found out that the highly oriented β-$FeSi_2$ thin films could be obtained by using the Fe_2Si targets. On the other hand, however, particularly in the field of the application, to provide methods capable of depositing a high-quality β-$FeSi_2$ thin films inexpensively are also important as well. In this study, we employed a conventional vacuum deposition system with the best base pressure at 1×10^{-6} Torr by using the oil diffusion pump and the resistance heating evaporation system. We used pure Fe and a few different kinds of iron silicides of Fe_2Si, α-$FeSi_2$ and ε-FeSi, which each contains Si, as the source materials to fabricate β-$FeSi_2$ thin films by the RDE method. The crystal structure and the electrical properties of the obtained thin films were investigated.

2. Experimental

We applied the conventional vacuum deposition system with the base pressure at 1×10^{-6} Torr using the oil diffusion pump, which is able to heat the substrate up to 900°C. The source materials of Fe,

Fe_2Si, ε-FeSi and α-$FeSi_2$ were placed in the alumina crucibles that were bound by W wire for the resistance heating and deposited onto the heated Si(100) surface, respectively. Typical gas pressure during deposition was around $3x10^{-5}$ Torr. The temperature of Si substrate during deposition, T_{sub}, was T_{sub}=650-750 °C. Some samples were then post-annealed under the vacuum at the temperature T_{ann}=750 °C. The crystal structures were evaluated by XRD. The amounts of Fe deposited were determined by SEM-EDS measurements from the integrated intensity of the Fe Kα line. Van der Pauw method and Hall measurements were employed for the electrical evaluations of the samples at room temperature. We used non-doped high resistive n-type Si wafer, so we neglected the conduction through Si substrate to evaluate electrical properties of the deposited thin films.

3. Results and Discussion

3.1 XRD measurements

Figure 1(a) shows XRD profile of the thin film which was fabricated by pure Fe deposition onto heated Si substrate at T_{sub}=700 °C. This XRD profile indicates that β-$FeSi_2$ polycrystalline thin film could be grown by this method. Based on our previous works, the T_{sub}=700 °C is the most suitable temperature for β-$FeSi_2$ growth using this technique. Figure 1(b) shows XRD profiles of the thin

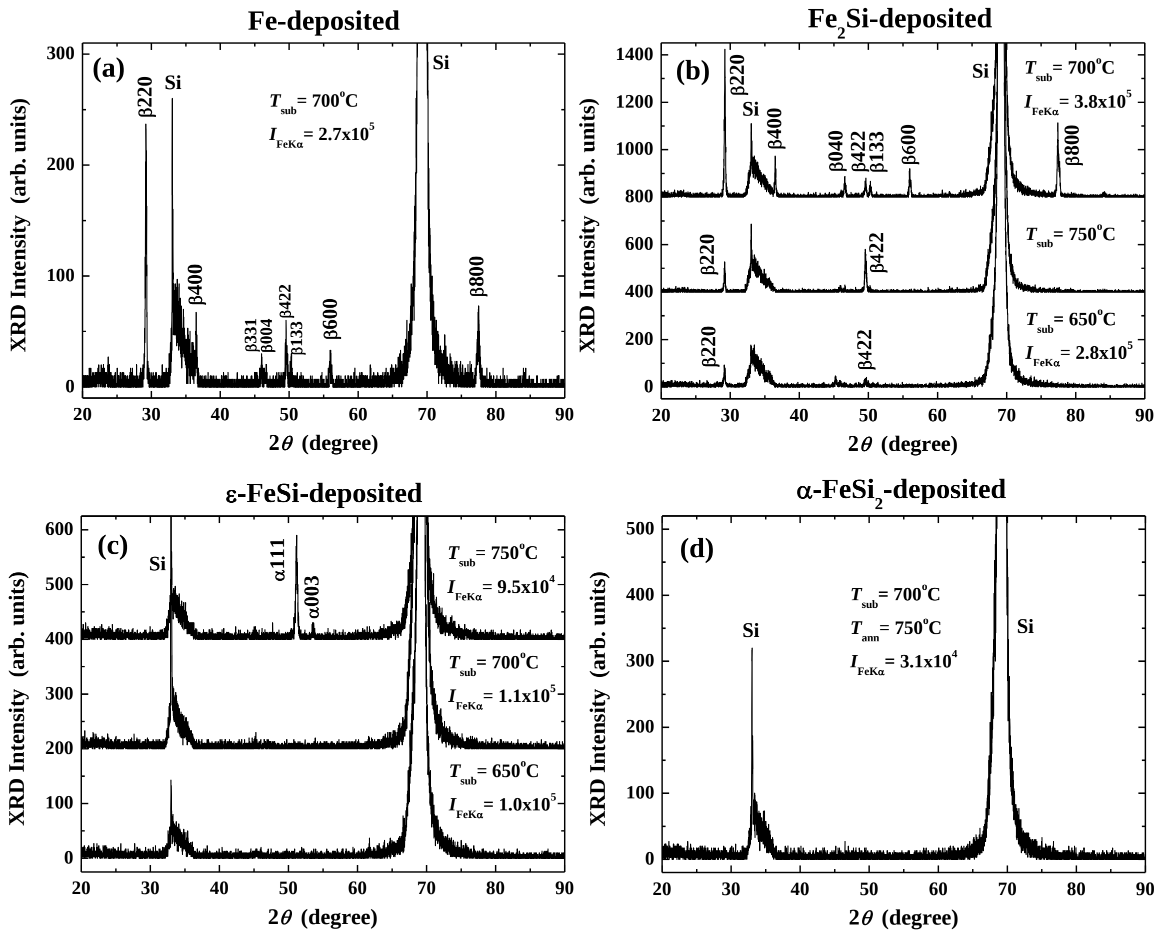

Fig. 1. XRD profiles of thin films fabricated in various conditions: (a) Fe-deposited, (b) Fe_2Si-deposited, (c) ε-FeSi-deposited, and (d) α-$FeSi_2$-deposited. $I_{FeK\alpha}$ represents the integral intensities of Fe Kα line from $1.28x0.96\ mm^2$ area of each film by SEM-EDS.

films that were grown from Fe_2Si deposited on Si substrate with temperature T_{sub} was 650, 700, 750 ^oC, respectively. It is readily seen from these XRD profiles that we could get the β-$FeSi_2$ polycrystalline thin films by using Fe_2Si as the source materials for the RDE growth. Particularly, in the case of T_{sub}=700 ^oC, the XRD peaks from *h*00 plane which corresponded to the epitaxial relationship to the Si(100) surface have become remarkable. Figure 1(c) shows XRD profiles for ε-FeSi deposition. There was no significant XRD peaks at T_{sub}=650 and 700 ^oC, on the other hands, α-$FeSi_2$ polycrystalline thin film was grown at T_{sub}=750 ^oC. In this case, Si was originated both from the deposited substance and the substrate, thus, the supply of Si was excessive to form the β-$FeSi_2$ thin films. Then Si-rich phase α-$FeSi_2$ (Fe_2Si_5) grow dominantly. This results coincide with the results of IBSD [8]. Figure 1(d) shows XRD profile for α-$FeSi_2$ deposition onto Si substrate at T_{sub}=700 ^oC and the post-annealed temperature in a vacuum T_{ann}=750 ^oC for 12 hours. Contrary to our expectations, there was no XRD peaks from the thin films, which means that no crystallization had occurred. Based on these experiment results, we found that only in the case of Fe and Fe_2Si as the source materials, the β-$FeSi_2$ single-phase polycrystalline thin films could be obtained.

3.2 SEM-EDS observations

Figure 2 shows the cross-sectional SEM images of β-$FeSi_2$ thin films, (a) Fe-deposited and (b) Fe_2Si-deposited, respectively. The EDS peak integral intensities of Fe Kα from 1.28x0.96 mm^2 area of these films, $I_{FeK\alpha}$, are roughly equal. As can be seen, the aggregation growth, which has many

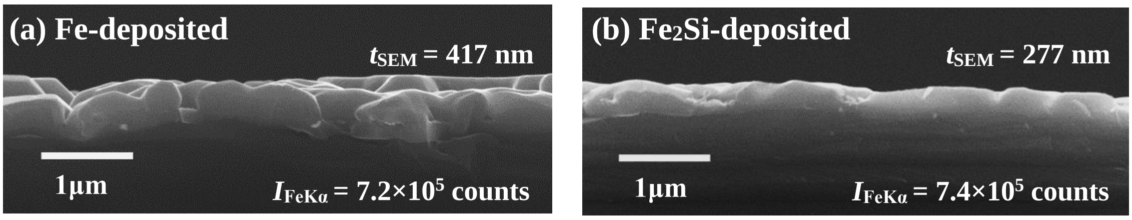

Fig. 2. Cross-sectional SEM images of β-$FeSi_2$ thin films, (a) Fe-deposited, (b) Fe_2Si-deposited. t_{SEM} are average thickness of β-$FeSi_2$ thin films evaluated from the cross-sectional SEM images, and $I_{FeK\alpha}$ represents the EDS peak integral intensities of Fe Kα line from 1.28x0.96 mm^2 area of each films.

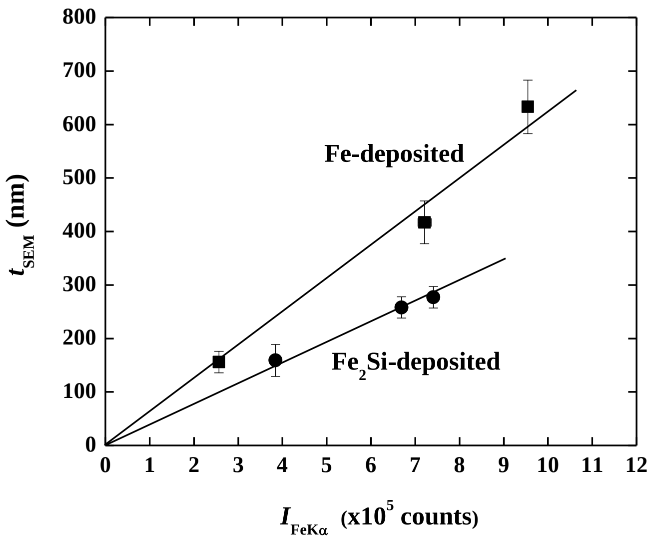

Fig. 3. Plots of the film thickness evaluated by cross sectional SEM (t_{SEM}) versus the integrated intensity of Fe Kα lines ($I_{FeK\alpha}$) evaluated by SEM-EDS. Solid squares are for Fe-deposited, and solid circles are for Fe_2Si-deposited.

interstices and irregularity of interface between β-$FeSi_2$ and Si substrate, has been remarkable more in (a) than (b). We fabricated two other thickness by same ways and those XRD patterns showed that β-$FeSi_2$ single phase thin films grown by using Fe and Fe_2Si as source materials. Figure 3 is the plots of the thickness evaluated by the cross sectional SEM, t_{SEM}, versus $I_{FeK\alpha}$ of all thin films we fabricated. The thickness t_{SEM} seems proportional to $I_{FeK\alpha}$ with different slopes indicating that the films prepared by Fe_2Si have the larger density than those by Fe as the source materials.

3.3 Electrical properties

Table I summarized the evaluated electrical properties of the β-$FeSi_2$ thin films. Both thin films by Fe and Fe_2Si deposited showed n-type conductions. The Hall mobility of β-$FeSi_2$ film by Fe_2Si deposited was 37% greater than that of the film by Fe deposited, and reached to almost the same value with which has been reported for β-$FeSi_2$ thin films fabricated by MBE method [9]. This improvement of electrical properties might be attributed by the continuing growth and smooth interface between β-$FeSi_2$ and Si substrate by using Fe_2Si for source materials.

Table I. Electrical properties evaluated by van der Pauw and Hall measurements at room temperature.

Source	T_{sub} [°C]	t_{SEM} [nm]	Conduction type	ρ [Ω·cm]	n [1/cm^3]	μ [cm^2/(V·s)]
Fe	700	148	n	0.25	$1.2\text{x}10^{17}$	202
Fe_2Si	700	132	n	0.28	$8.1\text{x}10^{16}$	276

4. Conclusion

We used a conventional vacuum deposition system whose base pressure was $1\text{x}10^{-6}$ Torr by the oil diffusion pump and the resistance heating evaporation system. We clarified that the depositing Fe_2Si onto heated Si substrate in RDE method helps improving the film flatness and electric properties of obtained β-$FeSi_2$ thin films compared to that of Fe-deposited films even in relatively low vacuum depositing. The electric properties of β-$FeSi_2$ reached to almost the same value as the Hall mobility reported for the β-$FeSi_2$ thin films fabricated by MBE method. However, we could not obtain β-$FeSi_2$ thin films from ε-FeSi and α-$FeSi_2$-deposition. This might be related to Yamaguchi's work [8] that pointed out excess Si makes the formation of β-$FeSi_2$ difficult. Further investigations of the relations between the conditions of deposition such as the deposition pressure, the deposition rate etc. and the crystalline and electrical properties of the fabricated β-$FeSi_2$ thin films are now in progress.

Acknowledgment

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