THE Ti/c-Si SOLID STATE REACTION

III. The low-temperature reaction kinetics

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Thin Ti layers (~ 10 nm) are grown on top of a clean Si(111) substrate. Heating these layers initiates a solid state reaction, yielding a monosilicide phase at $\sim 350^\circ\text{C}$ and a C49 disilicide at $\sim 450^\circ\text{C}$. The present study concerns the growth kinetics of both phases by means of ellipsometry. A diffusion-limited growth kinetics is found for the monosilicide formation. However, two growth rates are observed, a fast initial one and a slow terminal growth rate. An enhanced Si diffusion in atomically disordered regions as compared to well ordered regions (grains or clusters) could be an explanation. From the measurements we have found a value of $2 \times 10^{-15} \text{ cm}^2/\text{s}$ for the diffusion coefficient at $\sim 370^\circ\text{C}$ and an activation energy of 0.62 ± 0.1 eV. Both values correspond to the fast process. Subsequently increasing the temperature to $\sim 450^\circ\text{C}$ permits the growth of the homogeneous C49 TiSi_2 phase. For this process, both planar layer growth and intermixing are observed, however, quantitative results could not be derived from the present study.

1. Introduction

In two preceding papers [1,2], we have depicted the phase formation of the Ti/c-Si solid state reaction. Upon heating a thin Ti layer (~ 10 nm) grown on top of a clean Si(111) substrate, we observed the growth of a metastable monosilicide ($\sim 350^\circ\text{C}$), a homogeneous C49 TiSi_2 at $\sim 450^\circ\text{C}$ and the final C54 disilicide ($\sim 700^\circ\text{C}$). In this paper we will present quantitative information on the growth kinetics of the monosilicide phase and the C49 disilicide.

The low temperature solid state reaction has already been studied by Tromp et al. [3], Chambers et al. [4], Holloway and Sinclair [5] and by Raaijmakers [6]. All these investigations agree on the formation of a monosilicide at low temperature ($\sim 400^\circ\text{C}$), however, there is a large scatter among the diffusion coefficients reported. Moreover, the applied experimental techniques and methods were not suited for an unambiguous de-

termination of the character of the actual diffusion mechanism; one just assumed that the growth is diffusion limited. Ellipsometry offers the advantages of depth sensitivity (~ 30 nm) [1], an accurate and quantitative measurement, and mathematical models that can be used for the analysis of the optical data. Therefore, ellipsometry is one of the most appropriate techniques for the study of the growth kinetics of a monosilicide layer buried by an unreacted Ti layer.

For the present study we have followed the intermixing process by three-wavelengths ellipsometry ($\lambda = 340, 450$ and 550 nm). The optical, time-resolved registrations of the monosilicide formation could be fitted to a simple planar layer model; a top layer of pure Ti above a monosilicide layer. In this way we could calculate the thickness of the monosilicide layer as a function of time. From fundamental considerations, one can distinguish two growth modes: interface limited and diffusion limited growth [7,8]. The analysis of our time-resolved data agree with a diffusion limited process for the monosilicide formation. However, two growth rates are observed, a fast initial one and a slow terminal kinetics. The diffusion coeffi-

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cient for the fast process can be easily calculated, a value of $2 \times 10^{-15} \text{ cm}^2/\text{s}$ at $\sim 370^\circ\text{C}$ is found. Because we have followed the solid state reaction at two temperatures (300 and 370°C), we could make an estimation of the activation energy: $E_a = 0.62 \pm 0.1 \text{ eV}$. The slow, terminal growth rate could be an indication for an enhanced Si diffusion through atomically disordered regions; the conversion of these disordered regions terminates upon total consumption of these regions whereas the Si indiffusion into the ordered regions (clusters or grains) still sustains. Such a diffusion mechanism has been suggested previously [3,4,9,10]. An alternative explanation is that proposed by Raaijmakers [6]. He, and Holloway and Sinclair [5], observed the growth of Kirkendall voids, which might block further Si transport. These Kirkendall voids can be expected if one of the diffusing species dominates the other, which is the case for the Ti/Si system. Si has been shown to be the diffusing component for high temperature [11] ($\sim 700^\circ\text{C}$) as well as for low temperature [12] ($\sim 300^\circ\text{C}$).

If the temperature is increased when the Ti layer has already been converted to the metastable monosilicide, then a second Si indiffusion is initiated and a C49 TiSi_2 phase is obtained. This process is studied too, unfortunately, it was impossible to make the same quantitative analysis as for the monosilicide formation. Nevertheless, the results do indicate that planar layer growth is involved, however, for the thin layers we have found proof that fast diffusion along short-circuits assists in the solid state reaction.

The optical experiments are not discussed here but one is referred to the first article of this series [1]. Therefore, we will proceed in section 2 with the analysis of the ellipsometrical registrations of the monosilicide growth. The results will be discussed in section 3. The paper closes with a presentation of the results of the C49 disilicide formation.

2. Optical Analysis of the monosilicide growth

The growth process is, as described in ref. [1], followed ellipsometrically at three wavelengths,

Fig. 1. Trajectories in the Δ - Ψ plane of the Ti-TiSi transformation. The solid lines present the planar growth of a TiSi layer consuming the above Ti layer. The dashed-dotted lines denote the trajectories of an initially linearly grown TiSi layer, but the final 5 nm of Ti are converted to TiSi_2 by a process of mixing.

340, 450 and 550 nm. One of the options we have, is to measure $\Delta(t)$ and $\Psi(t)$ at these three wavelengths upon a fixed temperature increment at a number of equal time intervals of 30 s. Fig. 1 displays the trajectories drawn in the Δ - Ψ plane by the monosilicide growth. Prior to commencing the registration, the sample has been annealed during 2 min at $T \approx 200^\circ\text{C}$, which allows the Ti layer to relax and which enables one to ascertain whether the player has a stable Ti micro-structure or constant ($\sim$ zero) voids fraction [1]. The measurement has commenced at a temperature of $\sim 300^\circ\text{C}$, which, after 40 min, is increased to $\sim 370^\circ\text{C}$. Only one measurement is shown, nevertheless, five measurements have been performed.

The optical data thus comprise six independent quantities, the Δ and Ψ values for each of the wavelengths. For the analysis of the first diffusion process, we have assumed that a planar monosilicide layer grows which consumes the above Ti layer. Such an intermixing process is easily modelled by a two-layer model, assuming that the dielectric constants are known. The dielectric function of Ti has been determined previously [13] whereas the dielectric constants of the monosilicide and c-Si were obtained from ref. [1]. Hence, we do have the dielectric constants at our disposal,

Fig. 2. Thickness of the non-consumed Ti layer (d_1) as a function of the TiSi layer thickness (d_2) as calculated from the ellipsometric trajectories for five experiments.

and thus we can fit this two-layer model to the optical data by optimizing both layer thicknesses.

The dielectric functions are measured at RT whereas the silicide growth occurs at higher temperatures. This discrepancy certainly is to affect the analysis and, as shall be shown further on, is observed in the measurements. Because of this perturbation of the dielectric constants, we have used the very coarse model of two pure layers which did not contain voids reflecting the microstructure of the layers [1,14]. One is forced to do so in order to limit the freedom of any solution, thus avoiding errors due to a too large ability to fit the model to the measured data. Hence, every time-point consisting of six independent quantities is fitted to a two-layer model, yielding two optimized layer thicknesses, d_1 and d_2 . For this purpose we have applied a LRA program based upon the Levenberg–Marquardt minimization routine [15]. A similar example of such an analysis can be found in ref. [13].

The analyses of the individual data points comprising Δ and Ψ at three wavelengths, are depicted in fig. 2. The result d_1 , the thickness of the Ti layer, is shown as a function of the TiSi thickness d_2 . For the five samples, we see that the Ti layer is consumed at the same rate for the initially thinner layers (~ 8 nm Ti) and for the thick ones (~ 15 nm Ti).

From the thickness relation found and the initial Ti layer thickness, we have calculated the Δ – Ψ

trajectories for the sample depicted in fig. 1. The calculated trajectories are denoted by the solid lines. Clearly shown is the large, systematic discrepancy between these optimized trajectories and the actual measured data points. The difference has to be ascribed mainly to the temperature effect; the dielectric constants used are those obtained at RT whereas the silicide growth occurs at higher temperatures. A similar shift of the trajectories is shown in fig. 6. The dots denoting the data points are measured at a moderate temperature of $T \approx 400^\circ\text{C}$, whereas the crosses mark the initial and final values for Δ and Ψ of the sample but at RT. For the wavelengths 450 and 550 nm, the data points are observed to shift to higher Ψ values, a shift which is also observed in the present analysis of the Ti–TiSi formation. Errors will be introduced by this shift, however, since the shifts are systematic, the errors in the optimized parameters d_1 and d_2 will have also a systematic character. It will be obvious that if the layers are allowed to contain a variable voids fraction, optimizing this fraction will yield erroneous results.

3. Discussion

From a simple consideration of the mass transport across an interface and through a layer, it shows that there are two growth modes, an interface limited mode and diffusion controlled mode [7,8]. The interface controlled mode corresponds

Fig. 3. The thickness of the TiSi layer as a function of time for two temperatures. Two experiments are shown.

with a layer growth proportional with time, i.e., $d_2 \propto t$, diffusion limited growth with $d_2 \propto \sqrt{t}$. In figs. 3 and 4, we have respectively plotted d_2 and d_2^2 as a function of time for two samples and two temperatures. Fig. 3 shows that the growth rate diminishes as time passes, and a fast increase of the growth rate is seen as a response to a temperature increment. Most revealing is fig. 4. At $T = 300^\circ\text{C}$, we see that d_2^2 is proportional to t for both samples. As far as we could study the growth process, there was already a thin 3–5 nm intermixed region [1], we do not observe any trace of an interface limited process. Hence, if there is any diffusion barrier for Si transport across the Si–silicide interface, it has to be small and we can safely conclude that the silicon diffusion satisfies the diffusion controlled mode.

The growth rate is strongly increased upon the temperature increment to 350 or 370°C . Initially at these temperatures, a diffusion limited growth is followed, $d \propto t^2$, however, in both cases it shows that the growth rate diminishes although the diffusion process has not yet terminated. Apparently, there is a second, slow diffusion process that sustains once the fast diffusion process has terminated. We will discuss this slow diffusion process further on and proceed with the fast process.

The diffusion coefficients at 300 and 370°C are 5×10^{-16} and 2×10^{-15} cm^2/s respectively. This is in good agreement with the result obtained by Chamber et al. [4], who found a value of

$\sim 7.5 \times 10^{-16}$ cm^2/s at $T = 340^\circ\text{C}$ for a Ti layer grown from an evaporation source. Raaijmakers [6] has studied the growth kinetics of the a-Si/sputtered-Ti system: he found a diffusion coefficient of about 1.0×10^{-16} cm^2/s at $T \approx 400^\circ\text{C}$. Hence, the intermixing process in the case of an evaporated Ti layer roughly evolves by one order faster than the interdiffusion of a-Si and sputtered Ti. Such a discrepancy is too large to be attributed to experimental errors and therefore it can be concluded that the reaction kinetics depend on the nature of the system. In particular, the deposition technique for the Ti layer is expected to affect the Ti film structure strongly, which, in turn, might affect the diffusion coefficient.

From the two diffusion coefficients we can easily calculate the activation energy for the monosilicide formation, a value of 0.62 ± 0.1 eV is found, which is somewhat smaller than the value reported by Chambers et al. [4], $E_a = 1.0 \pm 0.25$ eV. Other values are thus far not reported in the literature, but for the TiSi_2 (C49) formation an activation energy of ~ 1.8 eV is found [6,16]. This higher activation energy for the TiSi_2 growth explains why the growth of a monosilicide is not observed at high temperatures.

As mentioned above, there are at least two diffusion controlled growth mechanisms, a fast initial one and a second slow one. Further we have seen that the diffusion rate is an order of magnitude higher than that for sputtered Ti layers. A possible explanation for both phenomena might be given by the micro-structure of the Ti layer.

With regard to high-melting-point metals, molecular dynamics [17,18] show that the adatom mobility is low for low substrate temperatures, allowing adsorbed atoms to move no further than to a neighbouring lattice site. Due to this restricted lateral diffusion, a polycrystalline layer containing numerous lattice defects or even micro-pores or voids can be expected. A higher substrate temperature and a larger kinetic energy of the arriving atoms reduce the number of defects, or improve the layer structure. This effect is recently clearly shown by Yamada and Yoshida [19] who have studied Ti evaporated films grown at various substrate temperatures by means of Röntgen diffraction. Therefore, it can be expected

Fig. 4. Square thickness of the TiSi layer as a function of time for two temperatures. The data correspond with that of fig. 3.

that the as-deposited Ti layer is composed of extremely small grains or clusters of a few nm in size, separated by regions of disordered atoms. Heating such a layer should improve the crystallinity or ordering of the atoms, which in fact, is exactly what is observed in the first paper [1]. Moreover, sputtered Ti layers are expected to have a better structure (larger grains) as compared to evaporated films, since in this latter case the atoms arrive with but some thermal energy only, whereas sputtered atoms have a kinetic energy of the order of 1 eV.

It is frequently suggested that grain boundaries provide short-circuits for Si diffusion through a metal layer [3,4,9]. Solid proof, such as for some metal-metal systems [20,21], is not available. However, if we assume that Si diffusion is enhanced with diminishing film structure, it offers an explanation for the two growth rates and for the anomalous low diffusion coefficient found by Raaijmakers [6]. The presently studied layers have an extremely fine polycrystalline or cluster structure. The initial Si indiffusion will prefer the disordered regions and a monosilicide will predominantly grow in the disordered regions. This preferential growth will terminate by lack of disordered Ti, whereas the retarded indiffusion into the ordered regions (grains or clusters) sustains. This second process can be clearly observed in the absence of the fast diffusion. The Ti films studied by Raaijmakers are grown by sputtering, and have therefore a better micro-structure, and consequently, a lower diffusion coefficient for Si.

In our analysis of the experimental data, we have applied a simple planar layer model. The discrepancy between this model and the proposed diffusion mechanism will be obvious, and therefore it is required, to see whether or not we can distinguish optically the simple planar layer model from a more advanced model in better agreement with the proposed diffusion mechanism.

Once the Si diffusion along the grain boundaries has reached the Ti surface and all disordered Ti has been consumed, this growth mode terminates whereas the indiffusion of Si into the Ti grains or clusters sustains. Such a prolonged indiffusion then is observed as the second, slow growth mechanism. Since the grains near the interface are

longer exposed to the high Si concentration, they are further converted to TiSi as compared to the grains near the surface. Therefore, this final process of Si indiffusion into the grains is mainly observed near the surface of the sample and, if so, it looks like a mixing of Ti and TiSi above an already converted TiSi layer. We have illustrated this process in fig. 5. Such a process seems in better agreement with the proposed diffusion mechanism and can be optically modelled. We have assumed that initially a normal layer growth commences, but that the final 5 nm of Ti are not consumed planar from below but mix homogeneously with TiSi; the Ti:TiSi ratio diminishes from 1 to 0. Meanwhile, the volume expansion, or thickness, of this top region is increased linearly with the TiSi amount. The trajectories in the Δ - Ψ plane thus obtained for the final growth process are displayed by the dashed-dotted lines in fig. 1. From these results it is apparent that the trajectories are but slightly affected and that it is difficult to distinguish between an ordinary planar layer growth and the presently discussed terminal stage.

An alternative explanation is offered by the work of Raaijmakers on the growth kinetics of the a-Si/sputtered-Ti system [6]. He has observed that initially an amorphous monosilicide grows but that this growth terminates upon the formation of ~ 18 nm of amorphous TiSi. The interruption of the growth is tentatively ascribed to the formation of Kirkendall voids at the a-Si/TiSi interface. These voids then, can explain the second slow growth rate. However, Raaijmakers did not observe such voids for the case of a Ti layer on top of a c-Si substrate. Presently, we can also eliminate the formation of Kirkendall voids as a possible explanation for the second, slow diffusion process. Kirkendall voids are formed if there is a large transport of one component which is not balanced

Fig. 5. Schematic illustration of Ti-TiSi mixing in the top layer.

by outdiffusion of the vacancies created by the diffusing component. Fig. 3 shows that the largest Si transport is observed initially at $\sim 300^\circ\text{C}$. Therefore, Kirkendall voids are expected to form initially, whereas the slow diffusion process sets in later when the intermixing has almost ceased. Furthermore, if a large amount of micro-voids are formed at the Si–TiSi interface, this should greatly affect the optical properties of this voids-layer, an effect which should definitely be observed in the ellipsometrical data.

From the analysis of the experimental data it has been shown that the TiSi formation is a diffusion controlled process whereas there are no indications of a diffusion barrier at the Si interface. However, two growth rates are observed, which is attributed to the involvement of disordered regions or grain boundaries.

4. TiSi₂ formation kinetics

When the sample heating is proceeded at $T \approx 400\text{--}450^\circ\text{C}$ after the TiSi phase formation has ceased, a second enhanced indiffusion of Si is observed and a TiSi₂ layer is formed. As done previously for the TiSi formation, we should like to study the kinetics also. However, the initial Ti layers are better characterized and less complicated than the subsequently obtained TiSi layer. Hence, the study of the kinetics is harder to perform and acceptable analyses similar to those

for the monosilicide growth as shown in figs. 2–4 could not be obtained. However, detailed study of the growth kinetics of the reference sample also reveals that short-circuits for diffusion do interfere in the TiSi₂ formation in the case of thin films.

In fig. 6a we have plotted the trajectories as they are measured during the conversion of the monosilicide to the C49 disilicide. Together with the trajectories of the reference sample which we have analysed extensively in the first paper [1], we have plotted the trajectories for another, arbitrary sample, showing that they differ in detail whereas the global features are similar. The advantage of this particular sample is that the initial and final thickness and the dielectric functions are known exactly since these are derived from this particular sample. The initial and final situations are accurately known [1] and there can be only speculation on the question how the final state is obtained.

The crosses for $\lambda = 450$ and 550 nm denote the RT Δ – Ψ values. Upon heating the samples, these values are shifted to the start and end positions of the trajectories. We have calculated the trajectories for the hypothetical situation that the 19 nm thick TiSi layer is consumed by a planar TiSi₂ layer from below, which ultimately becomes 27 nm thick. These trajectories calculated from RT dielectric constants begin and end at the crosses, but we have shifted them allowing for a better comparison with the data points measured. The trajectory for $\lambda = 340$ nm has not been shifted. From the simulation it appears that, initially, this

Fig. 6. (a) Trajectories of the TiSi–TiSi₂ transition. Two experiments are displayed. The solid lines denote the planar layer growth mode. The crosses indicate the RT Δ and Ψ values for $\lambda = 450$ and 550 nm, which upon heating shift, reversibly, to the begin and end of the trajectory. The solid trajectories begin at the crosses, but are shifted. (b) Magnification of the 550 nm trajectory. The solid lines denote the planar layer growth trajectory whereas the dashed-dotted line corresponds to a remnant TiSi layer mixing. (See fig. 3.)

growth mode is followed but that after some time, upon a temperature increment, the growth mode deviates from this simple model. This is more clearly illustrated in fig. 6b. The planar layer growth is interrupted upon the formation of ~ 10.5 nm of TiSi_2 . The dashed-dotted trajectory is obtained if the subsequent process concerns an intermixing of the remnant TiSi layer with TiSi_2 , accompanied by the appropriate volume expansion. Fig. 5 illustrates this mechanism. It shows that a mixing instead of a proceeding planar layer growth provides a good explanation of the observed TiSi_2 formation. Initially, the evolution of the trajectories agree perfectly, whereas further on, after crossing the curve for the planar layer growth the measured data points and the mixing curve deviate. This deviation can be attributed to some surface roughening, which induces a shift of Δ to lower values.

Again, it shows that a planar layer growth mechanism cannot solely explain the observed trajectories; upon a temperature increment the planar layer growth is replaced by a mixing process, showing that short-circuits facilitating the diffusion are involved. This assumption is further supported by some scanning electron and scanning Auger microscopy pictures (SEM and SAM) made on a PHI 600 system. A quite thick Ti layer

(~ 20 nm) is annealed until the TiSi_2 formation had almost ceased in the centre of the sample. The sample temperature at the edges will be lower due to the sample support and one can expect to find the intermediate stages of the silicide formation on going from the middle to the edges. The SAM picture, fig. 7, shows a circle-shaped Si-rich area around a pit arisen in the layer. From the shape it appears that the Si has diffused out of the pit, clearly showing that there are short-circuits for Si diffusion and that Si is able to penetrate into the monosilicide layer from above the layer.

5. Conclusion

In this paper we have studied the growth kinetics of the low-temperature monosilicide growth and the subsequent disilicide (C49) growth.

The formation of the inhomogeneous monosilicide, requires Si diffusion through the previously formed monosilicide layer. This Si transport is diffusion-limited and a growth proportional to $\sqrt{t}$ is observed. The diffusion coefficient at 300°C is $5.0 \times 10^{-16} \text{ cm}^2/\text{s}$ and the activation energy is found to be 0.62 eV. This diffusion coefficient is about an order of magnitude larger as that reported by Raaijmakers [6]. This large discrepancy can be an indication that the Si transport is strongly affected by the Ti micro-structure.

Another peculiarity is the observed second, slow growth mechanism. The initial (fast) Si indiffusion is therefore tentatively ascribed to the Si diffusion into the disordered regions in the evaporated Ti layer whereas the final (slow) diffusion mechanism should correspond to the retarded diffusion of Si in the ordered regions (grains).

Once the formation of the inhomogeneous TiSi phase has terminated, a second Si indiffusion can be initiated by raising the temperature further and the homogeneous TiSi_2 (C49) phase is formed. The study of the growth of this disilicide proved to be more complicated. At low temperatures a planar layer growth is observed. However, increasing the temperature enhances Si diffusion along grain boundaries or other artifacts in the silicide layer to the surface, from where the Si can diffuse laterally over the layer.

Fig. 7. SAM picture of the silicide surface at an arbitrary moment during the transition from TiSi to TiSi_2 (C49). The light area is strongly Si-enriched as compared to the dark regions. Apparently there are short-circuits for Si diffusion.

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