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Surface Diffusion Controlled Formation of Nickel Silicides in Silicon Nanowires

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Nickel silicide/silicon contacts used in field-effect transistors (FET) based on silicon nanowires (SiNWs) can be formed by thermally activated axial intrusion of nickel silicides into the SiNW from prepatterned nickel reservoirs located at both ends of the NWs. This method seems promising for future electronic applications. Transformation of the longitudinal NW segments into single-crystalline nickel silicides throughout the entire NWs bulk has been interpreted as evidence of a volume diffusion control process. However, the volume diffusion coefficients of nickel in Ni_2Si at 300°C to 400°C are inconsistent with observable nickel silicide intrusion lengths. The experimental results published so far show a distinct dependence of nickel silicide intrusion length on the silicon NW diameter, which is indicative of a surface diffusion or a surface reaction controlled process. In this work, this problem was considered theoretically in the framework of a model of a diffusion-controlled phase formation. Diffusion growth of a wedge-like new phase in a cylindrical NW was described using a quasistationary approximation. The rate of longitudinal growth depends on the NW radius, R, and decreases with the radius increase as $\sim R^{-0.75}$. The dependence of R on annealing time, t, is close to $t^{0.5}$. The profile of the new phase was described for different combinations of two dimensionless parameters: R/δ and $D_{\gamma}/D_{s\gamma}$, where δ is the thickness of the high-diffusivity surface layer with diffusion coefficient $D_{s\gamma}$, and D_{γ} is the volume diffusion coefficient. After the formation of a continuous layer of a new phase, further growth is controlled exclusively by the interface diffusion of Ni along the nickel silicide surface and Si/Ni_2Si interface. The growth kinetics depends on the ratio of diffusion coefficients $D_{s\gamma}/D_b$, where D_b is the interface diffusion coefficient, and may be parabolic or linear. The calculated dependencies were compared with the published experimental results for nickel silicide formation in SiNWs. The analysis performed indicates that surface and interface diffusion of nickel play an important role in the formation of nickel silicides in NWs—a critical finding that should be considered in the design of SiNW FETs.

Key words: Semiconducting nanowires, field-effect transistor, nickel silicide/silicon contacts, surface and interface diffusion

INTRODUCTION

Quasi-one-dimensional semiconducting structures have recently been studied as potential candidates for electronic applications. Semiconducting nanowires (NW) have been implemented as the active channel of field-effect transistors (FETs) with linear and Schottky barrier (SB) source and drain contacts (SBFET).[1–5]

Silicon-NW (SiNW) FETs are of particular interest because they could allow the combination of one-dimensional transport and self-assembly techniques with well-established Si technology.

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Electrical contacts for these nanowires are of great importance. One plausible solution is nickel silicide contacts. Nickel silicide/silicon contacts were utilized in SiNW FETs using thermally activated axial intrusion of nickel silicides into the silicon NW from photo- or e-beam lithography prepatterned nickel reservoirs located at both ends of the NWs.[6–8] This seems to be a promising method for future electronic applications. However, such penetration of nickel silicide involves different thermally activated processes such as volume, surface, and interface diffusion of nickel, formation of substantial internal stresses in SiNWs, and detachment and even possible breakdown of SiO_2 films, influencing the surface diffusion and lateral growth of Ni silicide. The interplay between diffusion and internal stresses can significantly destabilize the FET fabrication process and affect electrical performance.

Formation of Ni silicides in SiNWs was studied recently in a number of works.[6–10] Transformation of the longitudinal NW segments into single-crystalline nickel silicides throughout the entire NWs bulk was interpreted as evidence of a volume diffusion controlled process. However, the volume diffusion coefficients of Ni in Ni_2Si at 300°C to 400°C measured in thin-film diffusion experiments[11] are inconsistent with observable nickel silicide propagation lengths.

Experimental results published until recently[6] demonstrated a distinct dependence of the intrusion length of nickel silicide on the diameter of the silicon NW. This can alternatively be considered as an indication of a surface diffusion controlled process or interface reaction controlled process. The kinetics of silicide growth controlled by volume, surface or interface diffusion (or interface reaction) should be different. Present understanding of the silicide formation mechanism seems to be insufficient because of contradiction between the volume character of epitaxial growth of nickel silicides and low rates of volume diffusion at the temperatures employed.

In the present work the problem of silicide growth in SiNWs was considered theoretically in the framework of the diffusion phase formation model.

MODEL

The model used in Refs. 12 and 13 considers diffusion-controlled formation of intermediate phases presented on the corresponding binary phase diagram. A nickel source is deposited on top of a SiNW at the wire end (depicted schematically in Fig. 1). After initial formation of a nickel silicide layer (in the immediate vicinity of the surface), diffusion occurs along the silicide surface and/or silicide/SiO_2 interface toward a Si/silicide interphase boundary (stage 1, Fig. 1a). Fast diffusion along the outer silicide interface is accompanied by leakage of nickel into the volume of the new phase and growth of silicide in the radial direction, causing the interphase boundary to move in the axial direction.

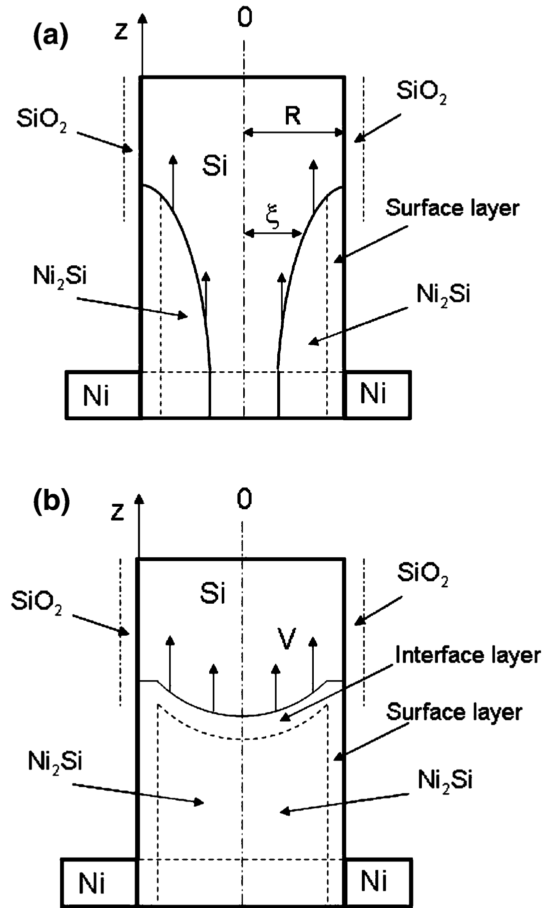

Fig. 1. Growth of nickel silicide along silicon NW (schematic): lateral growth, stage 1; (b) discontinuous growth, stage 2.

The problem of diffusion growth of a wedge-like new phase at stage 1 was solved using a quasistationary approximation.[14] This approximation is justified by the fact that volume diffusion is much slower than diffusion of Ni along the silicide surface. Assuming that only radial volume diffusion occurs, the steady-state distribution of nickel inside the new silicide phase, c, is described by the following equation:

$$\frac{\partial}{\partial \rho}\left(\rho \frac{\partial c}{\partial \rho}\right) = 0. \tag{1}$$

The boundary conditions at the cylinder surface and at the Si/silicide interphase boundary are:

$$c|_{r=R} = c_{\mathrm{s}} \text{ and } c|_{r=\xi} = c_{\gamma}, \tag{2}$$

where $\xi = R - \bar{\rho}$, and $\bar{\rho}$ is the thickness of the new phase. Equations 1–2 yield the following steady-state distribution of nickel inside the silicide phase:

$$c(r) = \frac{c_{\mathrm{s}} \ln(r/\xi) + c_{\gamma} \ln(R/r)}{\ln(R/\xi)}. \tag{3}$$

Let us now write down a Stefan condition at the moving interphase boundary:

$$(c_\gamma - c_\alpha)\frac{\partial \xi}{\partial t} = D_\gamma \left(\frac{\partial c}{\partial r}\right)\bigg|_{r=\xi}, \tag{4}$$

and a Fisher-like equation[14] for the nickel diffusion along the cylinder surface accompanied by leakage of nickel into the volume of the silicide:

$$D_{s\gamma}\frac{\partial^2 c_s}{\partial z^2} - \frac{1}{\delta}D_\gamma\left(\frac{\partial c}{\partial r}\right)\bigg|_{r=R} = 0, \tag{5}$$

where z is the longitudinal wire direction and δ is the thickness of the high-diffusivity surface layer. Combining Eqs. 4 and 5 with Eq. 3, one can obtain the differential equation which describes the time dependence of the wedge-like profile of the new phase, $z(\xi,t)$:

$$\frac{\partial}{\partial t}\left[\frac{\partial^2}{\partial z^2}\left(\frac{\xi^2}{2}\ln\frac{R\sqrt{e}}{\xi}\right) - \frac{D_\gamma}{\delta \cdot D_{s\gamma}}\frac{\xi^2}{R}\right] = 0. \tag{6}$$

Here c_α is the nickel concentration in silicon, and D_γ and $D_{s\gamma}$ are the diffusion coefficients in the volume and in the near-surface layer of silicide, respectively. This equation should be solved with the following boundary conditions:

$$z = 0: \quad \xi_0^2 \cdot \ln\frac{R\sqrt{e}}{\xi_0} = \frac{R^2}{2} - 2\alpha D_\gamma t, \tag{7a}$$

where $\alpha = \frac{c_0 - c_\gamma}{c_\gamma - c_\alpha}$, and at the wedge bottom

$$z = z_0: \quad \xi = R. \tag{7b}$$

The boundary condition (7a) follows from Eqs. 3 and 4. Integration of Eq. 6 with the boundary conditions (7) gives the profile of the new phase in the following form:

$$z(\xi) = \sqrt{2}R\int\limits_{\xi_0/R}^{\xi/R}\frac{x\cdot\ln(1/x)\mathrm{d}x}{[F(x) - F(\xi_0/R)]^{1/2}}, \tag{8}$$

where $F(x) = x^2[(r/p)x^2\ln(e^{1/4}/x) + A\ln(e^{1/2}/x)]$, $r = R/\delta$, and $p = D_{s\gamma}/D_\gamma$. This profile was calculated for different dimensionless parameters r and p and dimensionless time $\tau = 2\alpha D_\gamma t/\delta^2$. Typical profiles $z(\xi)|_{\tau=\mathrm{const}}$ and $z(\tau)|_{r=\mathrm{const}}$ are presented in Figs. 2–4.

Following these calculations one can find that the rate of longitudinal growth depends on the NW radius, R, and decreases with increasing radius as $\sim R^{-0.75}$ (Fig. 3).

The time dependence of the wedge length is close to parabolic, $z \sim t^{0.5}$. The maximal wedge length (at the cylinder surface) can be determined using Eq. 9

$$\frac{\mathrm{d}z_0}{\mathrm{d}t} = \frac{\alpha D_{s\gamma}\beta(t)}{\sinh[\beta(t)z_0]}, \tag{9}$$

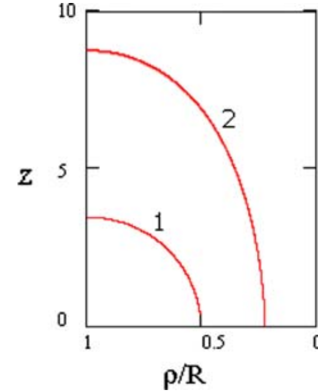

Fig. 2. Typical calculated profiles of a cylindrical wedge-like phase: $r = 50$, $\tau = 500$ (1) and $\tau = 10^3$ (2).

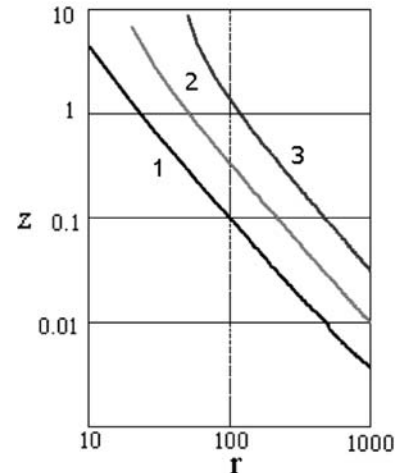

Fig. 3. Typical size dependence of the wedge length, $z(\xi = R) = Z/R \sim (1/r^{1.75})$ after different annealing times: 1 – $\tau = 10$; 2 – $\tau = 10^2$; 3 – $\tau = 10^3$.

which was derived using the steady-state nickel distribution in the silicide surface layer:

$$\frac{c_s - c_\gamma}{c_0 - c_\gamma} = \frac{\sinh[\beta(z_0 - z)]}{\sinh(\beta z_0)}, \tag{10}$$

where $\beta = \left[D_\gamma/D_{s\gamma}\delta \cdot R\ln(R/\xi_0)\right]^{1/2}$. Typical curves of $z_0(t)$ for different dimensionless radii R/δ are presented in Fig. 4a. The wedge length inside the cylinder (for example, for $\xi = R/2$) is described by the integral (Fig. 4b):

$$z(\tau, \xi = R/2) = \sqrt{2}R\int\limits_{\xi_0}^{0.5}\frac{x\cdot\ln(1/x)\mathrm{d}x}{[F(x) - F(\xi_0)]^{1/2}}. \tag{11}$$

The calculated curves presented in Figs. 2–4 can be well approximated by the following expressions:

$$\left(\frac{z(\xi)}{z_0}\right)^{1.493} = \frac{\xi - \xi_0}{1 - \xi_0}, \quad \frac{z(r)|_{\tau=\mathrm{const}}}{R} \approx \frac{b(\tau)}{r^{1.75}},$$
$$\frac{z(\tau)|_{r=\mathrm{const}}}{R} \approx c(r)\cdot\tau^{0.5}, \tag{12}$$

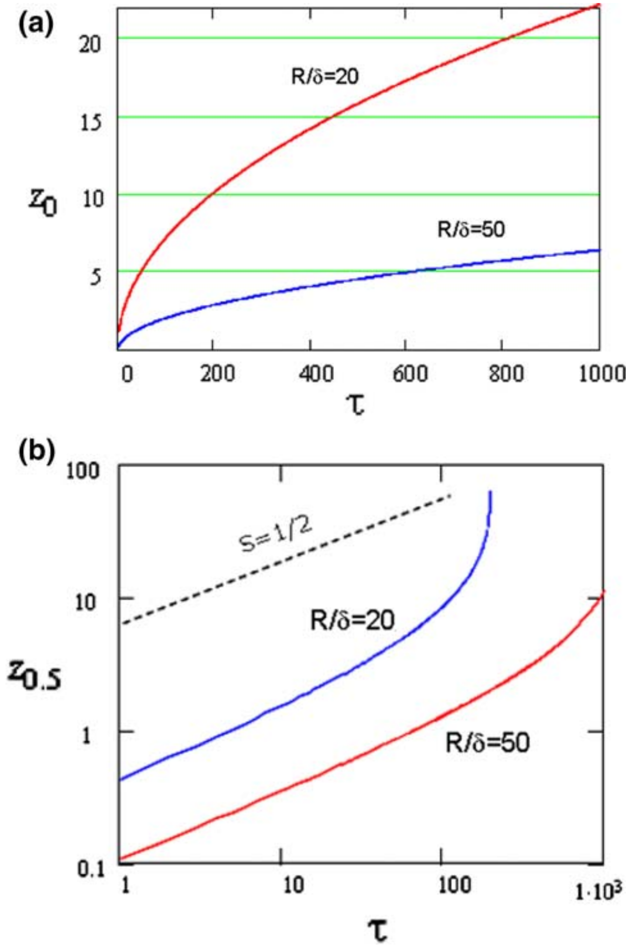

Fig. 4. Typical time dependences of the phase wedge length for $p = D_{s\gamma}/D_\gamma = 2.10^3$: (a) $\xi = R$, $z_0(\tau) \sim \tau^{0.5}$; (b) $\xi = R/2$, $z_{0.5}(\tau) \sim \tau^{0.5}$.

where the coefficients $b(\tau)$ and $c(r)$ depend on the ratio of diffusion coefficients, p.

For $\tau = r^2/2$ the profile becomes continuous, and the subsequent growth may occur as a result of interface diffusion of nickel along the nickel silicide surface and Si/silicide interface (stage 2, Fig. 1b). The corresponding growth kinetics depends on the ratio of interface diffusion coefficients $D_{s\gamma}/D_b$, where D_b is the diffusion coefficient along the Si/silicide interface. If $D_{s\gamma} \ll D_b$, the phase growth is parabolic: $z \approx 2\sqrt{(\delta/R) \cdot D_{s\gamma}t}$, with effective diffusion coefficient $D_{eff} = (\delta/R)D_{s\gamma}$. In the opposite case, $D_{s\gamma} \gg D_b$, phase growth is controlled by nickel diffusion along the Si/silicide interface and may proceed with a constant rate, V, in full analogy with the discontinuous precipitation reaction.[15] Diffusion along the interface boundary is driven by the gradient of nickel chemical potential, which depends on the nickel concentration and surface curvature gradients.

In the case of surface curvature gradient controlled diffusion, the mass balance equation can be written in the following form[16]:

$$\delta \cdot D_s \nabla^2 \left(\frac{\Omega}{k_B T} \gamma \mathrm{K}(\rho) \right) = V \cdot (c_\gamma - c_\alpha), \tag{13}$$

where γ is the interface energy, c_α is the nickel concentration in Si, Ω is the atomic volume, and K is the curvature of the Si/silicide interphase boundary. The steady-state curvature providing a constant rate V is determined as

$$\mathrm{K}(\rho) = \frac{\rho^2}{4\bar{R}^3}, \quad \bar{R} \equiv \left(\frac{\gamma \Omega \delta D_b}{k_B T V (c_\gamma - c_\alpha)} \right)^{1/3}. \tag{14}$$

The maximum growth rate provided by diffusion along the interphase boundary corresponds to a maximum curvature gradient, or to a maximum curvature at the wire surface:

$$\mathrm{K}(\rho = R) = \frac{R^2}{4\bar{R}^3} = \frac{2}{R}, \quad \bar{R} = R/2. \tag{15}$$

In this case the maximum growth rate is proportional to the diffusion coefficient D_b and interface energy γ, and inversely proportional to the cube of the NW radius:

$$V_{max}^{cur} = \frac{8\gamma \cdot \Omega \cdot \delta \cdot D_b}{k_B T (c_\gamma - c_\alpha)} \cdot \frac{1}{R^3}. \tag{16}$$

If the growth rate is controlled by the concentration gradient inside the interphase boundary, the maximal growth rate can be evaluated as[15]

$$V_{max}^{con} = \frac{2\delta \cdot s \cdot D_b \cdot \Delta c_\gamma}{(c_\gamma - c_\alpha)} \cdot \frac{1}{R^2}, \tag{17}$$

where Δc_γ is the maximum difference of nickel concentrations in the nickel silicide, and $s = c_b/c_\gamma \approx 0.5$. In this case the maximal growth rate is inversely proportional to the square of the NW radius. For typical values $D_b = 10^{-15}$ m^2/s to 10^{-14} m^2/s, $R = 20$ nm, $\delta = 0.5$ nm, $\Delta c_\gamma/(c_\gamma - c_\alpha) = 0.1$, Eq. 17 yields $V_{max} = 0.1$ nm/s to 1 nm/s. This rate is larger than the rate induced by the curvature gradient, provided that the NW radius is larger than a certain critical value, R^*:

$$R > R^* = \frac{4\gamma \cdot \Omega}{k_B T \cdot s \cdot \Delta c_\gamma}. \tag{18}$$

This result means that, for NWs with $R > R^*$, the Si/silicide interphase boundary will be flat. Using typical values of $\gamma = 0.1$ J/m^2, $\Delta c_\gamma = 0.05$, $\Omega = 5.9 \times 10^{-30}$ m^3, $T = 670$ K, and $s = 0.5$, one finds $R^* \approx 10$ nm.

DISCUSSION

Investigation of the size dependence of the kinetics of nickel silicide growth in SiNWs was performed recently in a number of works.[6–8] Appenzeller et al.[6] investigated Ni_2Si growth in SiNWs during annealing for 30 s and 300 s at the temperature of 280°C. They found that the silicide length depends on the NW diameter and presented their data as dependencies of silicide length versus

one over the NW diameter and diameter squared. They concluded that quadratic dependence describes the results better than the linear one because it can be extrapolated to zero length for very thick wires. From this fact they concluded that the silicide formation is a volume rather than a surface effect. At the same time, according to known diffusion coefficients in Ni_2Si[9], volume diffusion should be very slow at temperatures around 280°C.

According to our model these experimental results[6] can be discussed in terms of a surface diffusion controlled process, which leads to parabolic time dependence and reciprocal root dependence of silicide length on NW radius, $z \approx 2\sqrt{(\delta/R) \cdot D_{s\gamma} t}$. Evaluation of the surface diffusion coefficient from these results[6] gives reasonable values for the diffusion coefficient: $D_{s\gamma} = 5 \times 10^{-15}$ m²/s to 3×10^{-14} m²/s. The data[6] for silicidation length were replotted using coordinates of L and $(1/d)^{1/2}$ (Fig. 5). Extrapolation of the lines (which are a linear approximation to the experimental data[6]) to zero length ($L = 0$) highlights a threshold diameter of the NW, $d_{th} \approx 120$ nm (above which a continuous profile does not form), which can be considered as a diameter corresponding to the transition from stage 1 to 2 for a given annealing temperature and time. Volume diffusion coefficients evaluated from this threshold diameter value:

$$D_v = \frac{d_{th}^2}{8 \cdot \tau} = 5 \cdot 10^{-18} \div 3 \times 10^{-17} (m^2/s) \quad (19)$$

are still too large in comparison with volume diffusion coefficients found from thin-film experiments[11]: $D_{Ni}(Ni_2Si) = D_0 \exp(-Q/R_g T)$ with $D_0 = 3.54$ cm²/s and $Q = 239.8$ kJ/mol, which should be $\sim 10^{-26}$ m²/s for a temperature of 280°C. One can propose that this volume diffusion is substantially influenced by grain boundary (GB) diffusion in the silicide formed as in the case of solid-state reaction in thin films when the silicide formation is perpendicular to the Si substrate.[11]

Transformation of Si to nickel silicide at 470°C in SiNWs with diameters ranging from 7 nm to 40 nm was studied by Weber et al.[7] After annealing times ranging from 1 min to 10 min they observed a single-crystalline $NiSi_2$ throughout the entire NWs thickness with lengths of about 0.5 μm to 1 μm. The corresponding growth rate can be evaluated as ~1 nm/s. According to Eq. 17, when the growth is controlled by interphase boundary diffusion, the corresponding diffusion coefficient can be evaluated as $D_b \approx V \cdot R^2/(0.1\delta)$. For $R = 10$ nm and $\delta = 0.5$ nm, Eq. 17 yields $D_b = 2 \times 10^{-15}$ m²/s. This value can be compared with the grain boundary (GB) diffusion coefficient of nickel in Ni_2Si[11]: $D_{GB} = D_{GB}^0 \exp(-Q_{GB}/R_g T)$ with $D_{GB}^0 = 18.16$ cm²/s and $Q_{GB} = 165$ kJ/mol, which gives for the temperature of 470°C the value of 4×10^{-15} m²/s, in good agreement with Eq. 17.

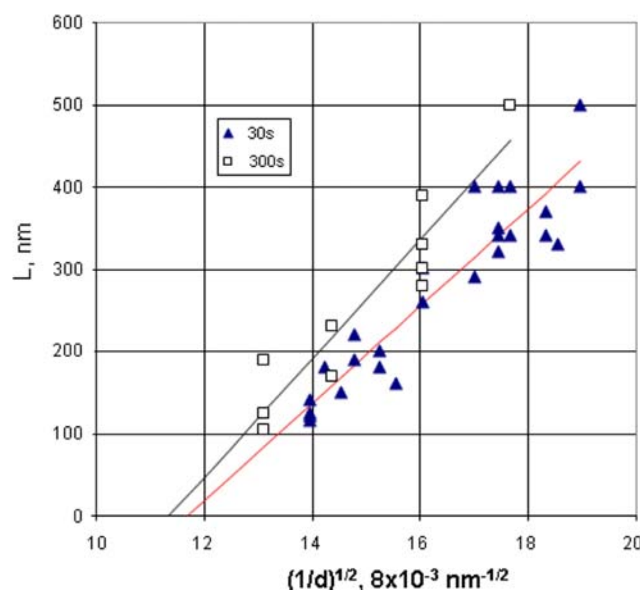

Fig. 5. Experimental results[6] illustrating the size dependence of Ni_2Si penetration length, L versus $(1/d)^{1/2}$, at 280°C; extrapolation of these dependences to $L = 0$ highlights a threshold thickness of the nanowire, $d \approx 120$ nm, above which a continuous profile does not form at the given annealing conditions.

Kinetic analysis of nickel silicide formation in SiNWs with diameters from 10 nm to 40 nm and in the temperature range from 500°C to 650°C was performed by Kuo-Chang Lu et al.[8] NiSi growth was the result of point contact reaction of nickel and SiNWs. The authors[8] found a linear time dependence of the silicide length for all temperatures investigated (Fig. 2a, *ibid*) with growth rates of 0.01 nm/s to 0.1 nm/s. The authors[8] extracted the activation energies from their data and obtained 1.25 eV (120.7 kJ/mol) for diffusion through the SiNW and 1.7 eV (164 kJ/mol) for diffusion through the nickel silicide phase. The latter is very close to the activation energy for GB diffusion mentioned above.[11] The authors[8] concluded that the observed epitaxial growth of NiSi may be an interface reaction controlled process accompanied by very fast volume diffusion of interstitial nickel atoms. We suggest an alternative explanation of the process as an interface diffusion controlled growth with a constant rate, V, given by Eqs. 13–17. Diffusion coefficients evaluated from Eq. 17, $D_b \approx 10^{-16}$ m²/s to 10^{-15} m²/s, are one or two orders smaller than the values estimated from data[7] and than the GB diffusion coefficient of nickel in Ni_2Si.[11] This difference can be associated with the different stoichiometry of the phases formed and with the different crystallographic orientations (⟨110⟩[10] and ⟨111⟩[8] SiNW directions), resulting in different crystallography of the Si/silicide interphase boundary. It was found[17] that the growth rate of Ni_2Si and the activation energy of the solid-state reaction in thin films depend on the Si substrate crystallinity

(single crystal or polycrystal) and orientation (⟨100⟩ or ⟨111⟩). For the growth direction normal to the substrate, typical values of diffusion coefficients are 10^{-18} m^2/s at 300°C and $Q = 1.6 \pm 0.2$ eV for films grown on ⟨111⟩ Si. Films grown on ⟨100⟩ Si or poly-Si exhibit higher formation rates and lower activation energies (1.5 ± 0.2 eV).[17] This growth is controlled by GB diffusion of Ni.[11] In lateral growth experiments the Ni_2Si is formed at 450°C to 630°C, and the rates are of the order of $10^{-14} m^2/s$, with an activation energy of 1.4 ± 0.1 eV.[11] The lateral growth of silicide is mainly controlled by surface and interface diffusion. By these means, the epitaxial silicide growth in SiNWs seems to be similar to lateral growth of silicides, with Si/silicide interface diffusion being a controlling factor.

CONCLUSIONS

Based on the diffusion model that was developed in this work and the analysis of previous experimental results published until now, the following conclusions can be drawn:

(1) Formation of nickel silicides in SiNWs in the temperature range of 280°C to 650°C is controlled by diffusion of nickel along the silicide surface and Si/silicide interface; the growth rate can also be controlled by the interfacial reaction at the Ni/Si contact pad;
(2) A certain preliminary nickel volume diffusion is required to form a continuous silicide layer. This volume diffusion may be enhanced by GB diffusion;
(3) A surface diffusion controlled process results in parabolic growth of the silicide segments and an inverse square root dependence of the silicide length on the NW radius, $L \sim 1/R^{1/2}$;
(4) Si/silicide interface diffusion controlled growth results in a linear time dependence of the silicide length and reciprocal dependence of the silicide length on the NW radius, R, according to $L \sim 1/R^3$ and $L \sim 1/R^2$ for the curvature controlled and concentration gradient controlled interface diffusion, respectively;
(5) Activation energies of nickel silicide growth in NWs (1.25 eV to 1.7 eV) are in reasonable agreement with those obtained from the kinetics of lateral growth of nickel silicides in thin-film solid-state reactions controlled by surface and interface diffusion of nickel.

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