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Interfacial reactions of Co/$Si_{0.76}Ge_{0.24}$ and Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ by pulsed KrF laser annealing

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Interfacial reactions of Co/$Si_{0.76}Ge_{0.24}$ and Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ by pulsed KrF laser annealing as a function of energy density and pulse number were studied. For the Co/$Si_{0.76}Ge_{0.24}$ samples annealed at an energy density of 0.2–0.6 J/cm^2, three germanosilicide layers, i.e., amorphous structure and/or nanocrystal, Co($Si_{1-x}Ge_x$), and Co($Si_{1-x}Ge_x$)$_2$, were successively formed along the film-depth direction. At 0.3 J/cm^2 Ge segregated to the underlying $Si_{0.76}Ge_{0.24}$ film, inducing strain relaxation in the residual $Si_{0.76}Ge_{0.24}$ film. At 0.8 J/cm^2 the reacted region was mostly transformed to a single layer of Co($Si_{1-x}Ge_x$)$_2$, whereas Ge further diffused to the Si substrate. At 1.0 J/cm^2, constitutional supercooling appeared. Even the Co($Si_{0.76}Ge_{0.24}$) film used as the starting material for laser annealing could not prevent the occurrence of constitutional supercooling at energy densities >1.6 J/cm^2. The energy densities at which Co($Si_{1-x}Ge_x$) transformation to Co($Si_{1-x}Ge_x$)$_2$, Ge segregation to the underlying Si, and constitutional supercooling occurred were higher for the Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ system than for the Co/$Si_{0.76}Ge_{0.24}$ system. Higher energy density and/or pulse number enhanced the growth of Co($Si_{1-x}Ge_x$)$_2$ film. In the present study, the Co/$Si_{0.76}Ge_{0.24}$ samples subjected to annealing at 0.2 J/cm^2 for 20 pulses produced a smooth Co($Si_{0.76}Ge_{0.24}$)$_2$ film without inducing Ge segregation out of the germanosilicide and strain relaxation in the unreacted $Si_{0.76}Ge_{0.24}$ film.

I. INTRODUCTION

$Si_{1-x}Ge_x$ material offers the promises of greater carrier mobility and band gap engineering and, hence, has potential applications in high-speed electronic and optoelectronic devices.[1,2] The formation of metal–$Si_{1-x}Ge_x$ ohmic or rectifying contacts is required for the device applications. Recently, the interfacial reactions of metals such as Ni,[3] Pt,[4,5] Pd,[5–7] Ti,[8–13] Co,[14–17] W,[18] Cr,[19] and Cu[20] with $Si_{1-x}Ge_x$ films by conventional furnace annealing have been studied. In these reactions, the formation of a ternary phase, e.g., M($Si_{1-x}Ge_x$)$_2$, Ge segregation out of the germanosilicide, strain relaxation of the unreacted $Si_{1-x}Ge_x$ film, and the occurrence of agglomeration structure at higher annealing temperatures were generally observed. These phenomena are presumably ascribed to the higher heat of formation for metal–Si than for metal–Ge.[21] Rapid thermal annealing[10,11] could shorten the annealing time and, hence, results in a reduction of Ge segregation and prevent the formation of agglomeration structure.

Pulsed laser annealing offers several advantages such as much shorter operational time, confinement of the heated area without causing changes in the preexisting structure, and reduction of contaminants. It has been extensively used in growing thin films of silicide,[22–25] germanosilicide,[26–28] $Si_{1-x}Ge_x$,[29] $Si_{1-x}C_x$,[30] and $Si_{1-x-y}Ge_xC_y$.[31] For Ni and Pd on $Si_{1-x}Ge_x$ pulsed KrF laser annealing has been found to effectively prevent Ge segregation out of the germanosilicide, the formation of agglomeration structure, and the occurrence of strain relaxation.[26–28] Strain relaxation is possibly induced by the chemical inhomogeneities and defects present in the interface between the germanosilicide and the unreacted $Si_{1-x}Ge_x$ film.[7]

In a salicide formation technique $CoSi_2$ has received much attention because it can significantly reduce the contact resistivity of Si devices and act as a solid diffusion source to form shallow junction. The impetus for

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studying the Co/Si system is easily transferred to the Co/$Si_{1-x}Ge_x$ system. In this study, pulsed KrF laser annealing as a function of energy density and pulse number was performed on the Co/$Si_{0.76}Ge_{0.24}$ system. The superior annealing condition was searched to effectively suppress or improve the phenomena, i.e., Ge segregation out of the germanosilicide, the formation of agglomeration structure, and the occurrence of strain relaxation in the unreacted $Si_{1-x}Ge_x$ film, which were generally observed on vacuum annealing.[3–20] In addition, to alleviate the constitutional supercooling phenomenon occurring during laser annealing, the Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ system was simultaneously studied because it was reported[23] that interfacial instability and cell formation can be suppressed by melting monosilicide or disilicide films.

II. EXPERIMENTAL PROCEDURES

Strained and partially relaxed $Si_{0.76}Ge_{0.24}$ films, about 100 and 150 nm thick, respectively, were grown at 550 °C in an ultrahigh vacuum chemical vapor deposition system. A Co layer, about 25 nm thick, was deposited onto the $Si_{0.76}Ge_{0.24}$ films at a rate of 0.1 nm/s in an electron gun deposition system. The base pressure was about 1.0×10^{-6} torr. Furnace annealing was conducted at 250 °C in a vacuum around 1.0×10^{-6} torr. Pulsed KrF laser annealing was performed at an energy density of 0.1–1.6 J/cm^2 in a vacuum around 2.0×10^{-2} torr. The pulse length was 14 ns. The laser beam was focused onto an area of 4×4 mm^2. For each laser annealing the sample was illuminated by a single shot unless otherwise specified. Samples with larger illuminated areas (10×10 mm^2) made of nine adjacent 4×4 mm^2 areas illuminated under identical conditions were prepared for x-ray diffraction (XRD) analysis to examine the annealing effect on the strain relaxation of the strained $Si_{0.76}Ge_{0.24}$ films. Phase formation and microstructures were observed by plan-view transmission electron microscopy (TEM) and cross-sectional TEM (XTEM). The depth profiles of the chemical elements in the films were observed by energy dispersive spectrometry (EDS) that was equipped with a field emission gun with a 1.2-nm electron probe.

III. RESULTS AND DISCUSSION

A. Co/$Si_{0.76}Ge_{0.24}$

On pulsed KrF laser annealing at an energy density of 0.2–0.6 J/cm^2 Co($Si_{1-x}Ge_x$)$_2$ appeared concurrently with Co($Si_{1-x}Ge_x$). An XTEM image of the sample annealed at 0.2 J/cm^2 is shown in Fig. 1(a), in which three distinct layers are present in the reacted region. From EDS/XTEM analysis the Co concentration decreased along the film depth as shown in Fig. 1(b). To clearly show the concentration changes of Co and Ge for those of $Si_{1-x}Ge_x$ and Si, respectively, in the Co($Si_{1-x}Ge_x$), Co($Si_{1-x}Ge_x$)$_2$, and $Si_{1-x}Ge_x$ layers, the Co and Ge concentration units in the figures, hereafter, are expressed as "atomic %" and "mole fraction (x)" respectively. From Fig. 1(b) it is evident that on annealing at 0.2 J/cm^2 the intermixing between Co and $Si_{0.76}Ge_{0.24}$ in the reacted region was not complete. Meanwhile some Ge segregated down to the bottom layer, indicating that Co tended to react preferably with Si. This result is presumably due to the higher heat of formation for Co–Si than for Co–Ge.[21] From microdiffraction analysis the top layer was of amorphous structure and/or nanocrystal, whereas the middle and bottom layers were of Co($Si_{1-x}Ge_x$) and Co($Si_{1-x}Ge_x$)$_2$ as shown in Fig. 1(c) and 1(d), respectively.

At 0.3 J/cm^2 Ge segregated out of the germanosilicide layers to the underlying $Si_{0.76}Ge_{0.24}$ film, forming a Ge-rich $Si_{1-x}Ge_x$ layer between the Co($Si_{1-x}Ge_x$)$_2$ layer and the residual $Si_{0.76}Ge_{0.24}$ film. Meanwhile, the XRD patterns in Fig. 2 show that after annealing at 0.3 J/cm^2 the c-axis lattice constant of the residual $Si_{0.76}Ge_{0.24}$ film was reduced compared with that of the as-grown $Si_{0.76}Ge_{0.24}$ film, revealing the occurrence of strain relaxation. Strain relaxation might be induced by the chemical inhomogeneities, e.g., Ge segregation, and defects present in the interface between the germanosilicide and the unreacted $Si_{1-x}Ge_x$ film.[7,17,26–28] At 0.6 J/cm^2 three germanosilicide layers were still present, and some Ge further diffused to the Si substrate, indicating that the melting depth exceeded the total thickness of the as-deposited Co layer (~25 nm) and the as-grown $Si_{0.76}Ge_{0.24}$ film (~150 nm). Meanwhile the inhomogeneous distribution of Co in the reacted region was significantly improved. At 0.8 J/cm^2 the reacted region was mostly transformed to a single layer of Co($Si_{1-x}Ge_x$)$_2$, but more Ge diffused into the Si substrate as shown in Fig. 3. It is apparent that annealing at higher energy densities not only allowed the Co layer to react with more of the underlying $Si_{0.76}Ge_{0.24}$ film, facilitating the growth of Co($Si_{1-x}Ge_x$)$_2$, but also enhanced Ge diffusion down to the Si substrate. At energy densities >1.0 J/cm^2, constitutional supercooling occurred, resulting in the cellular structures of Ge-deficient $Si_{1-x}Ge_x$ islands surrounded by Co germanosilicide.

The above results have shown that one pulse annealing at an energy density of 0.2–1.0 J/cm^2 can produce a three-layer germanosilicide film or a single-layer germanosilicide film with Ge diffusion down to the $Si_{0.76}Ge_{0.24}$ and Si substrates. Multiple pulse annealing has been reported to effectively homogenize the Pd concentration in the germanosilicide without inducing Ge segregation to the underlying $Si_{0.76}Ge_{0.24}$ film.[28] In the

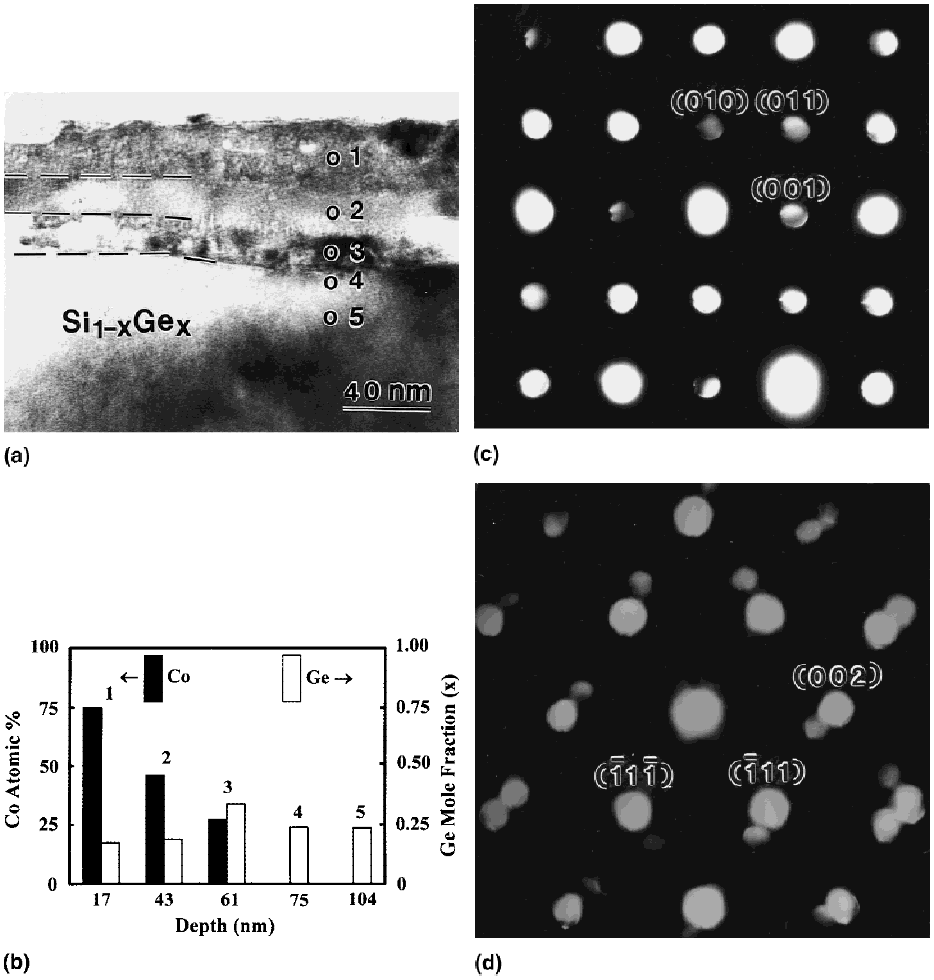

FIG. 1. (a) XTEM image and (b) the depth profiles of Co and Ge for the Co/$Si_{0.76}Ge_{0.24}$ sample annealed at 0.2 J/cm^2. Ge mole fraction (x) means the mole fraction of Ge in $Si_{1-x}Ge_x$ for Co($Si_{1-x}Ge_x$), Co($Si_{1-x}Ge_x$)$_2$, and $Si_{0.76}Ge_{0.24}$ films, respectively. The areas probed by electron beam are denoted as “O” and designated as 1, 2, 3, 4, and 5, respectively. (c) The [100] electron diffraction pattern (DP) of Co($Si_{1-x}Ge_x$) and (d) the [110] DP of Co($Si_{1-x}Ge_x$)$_2$ for the middle and bottom layers of the germanosilicide in (a), respectively.

present study, therefore, multiple pulse annealing at 0.2 J/cm^2 was performed on some samples. The decision to choose 0.2 J/cm^2 as the energy density for multiple pulse annealing is based on the result that on one pulse annealing at 0.2 J/cm^2 no Ge segregation out of the germanosilicide film occurred. For the sample annealed at 0.2 J/cm^2 for 5 pulses, two layers instead of three layers were formed in the reacted region. From electron diffraction analysis the two layers were of Co($Si_{1-x}Ge_x$) and Co($Si_{1-x}Ge_x$)$_2$, respectively. After annealing for 20 pulses the reacted region was mostly transformed to a smooth Co($Si_{1-x}Ge_x$)$_2$ layer as shown in Fig. 4(a). It is interesting to note that for a Co/$Si_{0.76}Ge_{0.24}$ sample annealed at 550 °C agglomeration occurred as shown in Fig. 5, in which Co($Si_{1-x}Ge_x$) islands were surrounded by Ge-rich $Si_{1-x}Ge_x$. The EDS/XTEM data in Fig. 4(b) show that no Ge atoms segregated out of the germanosilicide to the underlying $Si_{0.76}Ge_{0.24}$ film. Correspondingly, the XRD patterns in Fig. 2 reveal that no apparent strain relaxation appeared in the residual $Si_{0.76}Ge_{0.24}$ film after annealing at 0.2 J/cm^2 for 20 pulses. Similar results were observed in the Pd/$Si_{0.76}Ge_{0.24}$ system.[28] Those results imply that during the interfacial reactions of metal–$Si_{1-x}Ge_x$ the strain relaxation of the residual $Si_{1-x}Ge_x$ film can be suppressed by inhibiting Ge segregation out of the germanosilicide via pulsed KrF laser annealing. In the present study, we concluded that multiple pulse annealing at 0.2 J/cm^2 is an effective method to produce a smooth Co($Si_{0.76}Ge_{0.24}$)$_2$ film without Ge segregation out of the germanosilicide and inducing strain relaxation in the residual $Si_{0.76}Ge_{0.24}$ film.

In addition, the repetition rate, 1 Hz, used in the present study was low enough to exclude the cumulative heating effect.[32] Therefore, these results also prove that phase transformation and the diffusion of chemical species can be proceeded by each individual laser pulse for

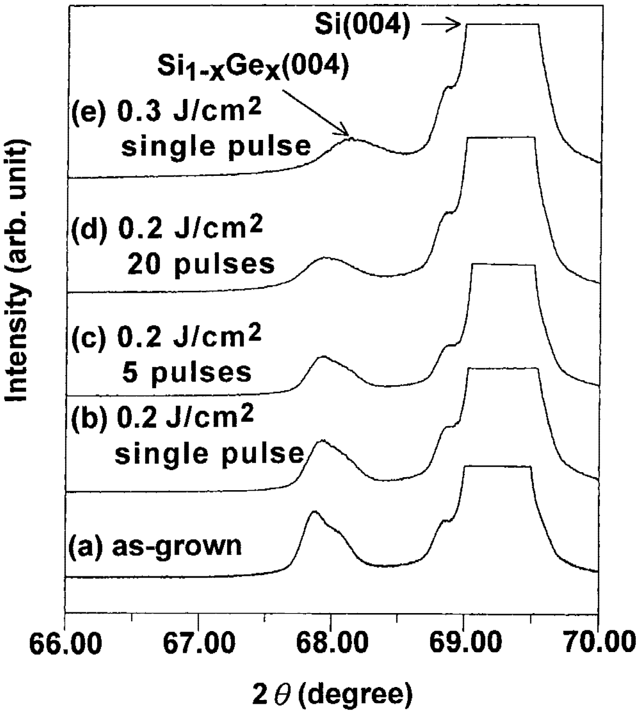

FIG. 2. XRD patterns of the Co/$Si_{0.76}Ge_{0.24}$ samples annealed at various conditions.

annealing at a low repetition rate. Similar results were found in the pulsed KrF laser annealing of Pd/$Si_{0.76}Ge_{0.24}$.[28]

B. Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$

It was reported[23] that constitutional supercooling can be suppressed by melting monosilicide or disilicide films; therefore, a simultaneous study of the Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ system by pulsed KrF laser annealing was also conducted. Figure 6 shows the XTEM image and EDS data of a Co($Si_{0.76}Ge_{0.24}$) film grown at 250 °C. Although some Ge tended to segregate to the surface of the Co($Si_{0.76}Ge_{0.24}$) film, both agglomeration and Ge segregation to the underlying $Si_{0.76}G_{0.24}$ film did not appear. Therefore, the Co($Si_{0.76}Ge_{0.24}$) films grown at 250 °C could be chosen as the precursors for subsequent growth of the continuous Co$(Si_{1-x}Ge_x)_2$ films by pulsed KrF laser annealing. On annealing at 0.2 J/cm^2 the Co($Si_{0.76}Ge_{0.24}$) layer remained inert without transforming to Co$(Si_{1-x}Ge_x)_2$. At an energy density of 0.4–0.8 J/cm^2 a Co$(Si_{1-x}Ge_x)_2$ layer was formed between the Co($Si_{1-x}Ge_x$) layer and the $Si_{0.76}Ge_{0.24}$ film. At 1.0 J/cm^2 a single-layer Co$(Si_{1-x}Ge_x)_2$ film was formed, but some Ge diffused to the Si substrate. It is worth noting that an energy densities >1.6 J/cm^2 even the monogermanosilicide, Co($Si_{0.76}Ge_{0.24}$), could not prevent the occurrence of constitutional supercooling.

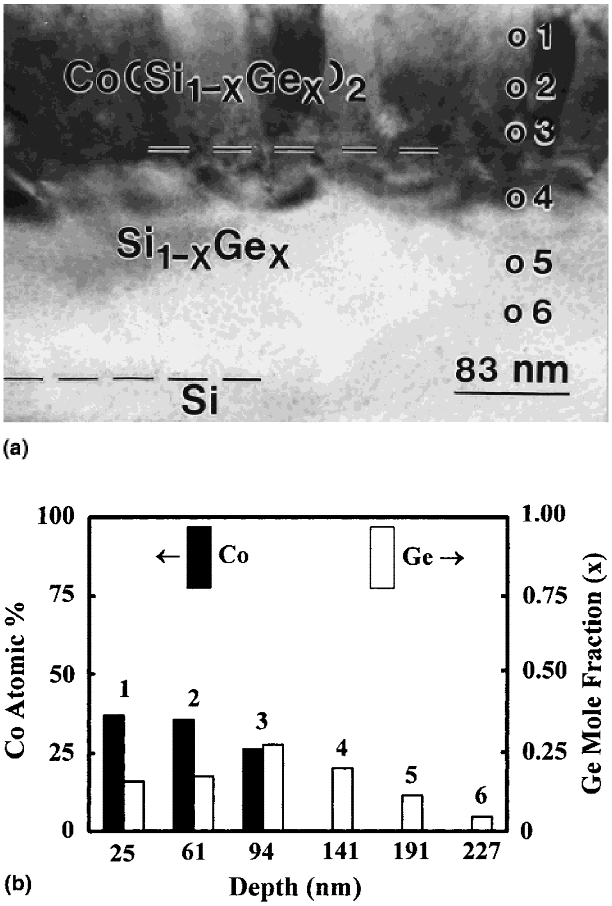

FIG. 3. (a) XTEM image and (b) the depth profiles of Co and Ge for the Co/$Si_{0.76}Ge_{0.24}$ sample annealed at 0.8 J/cm^2 showing that a single Co$(Si_{1-x}Ge_x)_2$ layer was formed concurrently with Ge diffusion to the underlying Si substrate. Ge mole fraction (x) means the mole fraction of Ge in $Si_{1-x}Ge_x$ for Co$(Si_{1-x}Ge_x)_2$ and $Si_{1-x}Ge_x$ films, respectively. The areas probed by electron beam are denoted as "O" and designated as 1, 2, 3, 4, 5, and 6, respectively.

In the present study the energy densities at which Co($Si_{1-x}Ge_x$) transformation to Co$(Si_{1-x}Ge_x)_2$, Ge diffusion to the underlying Si, and constitutional supercooling occurred were higher for the Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ system than for the Co/$Si_{0.76}Ge_{0.24}$ system. Similar results had been observed for Ni and Ni($Si_{0.76}Ge_{0.24}$) on $Si_{0.76}Ge_{0.24}$.[26,27] These phenomena may be explained in terms of the strong coupling of ultraviolet radiation with metals.[33]

IV. SUMMARY AND CONCLUSIONS

(1) For the Co/$Si_{0.76}Ge_{0.24}$ samples annealed at an energy density of 0.2–0.6 J/cm^2 three germanosilicide layers, i.e., amorphous structure and/or nanocrystal, Co($Si_{1-x}Ge_x$), and Co$(Si_{1-x}Ge_x)_2$, were formed in the reacted region along the film-depth direction. At 0.3 J/cm^2, Ge started to segregate to the underlying

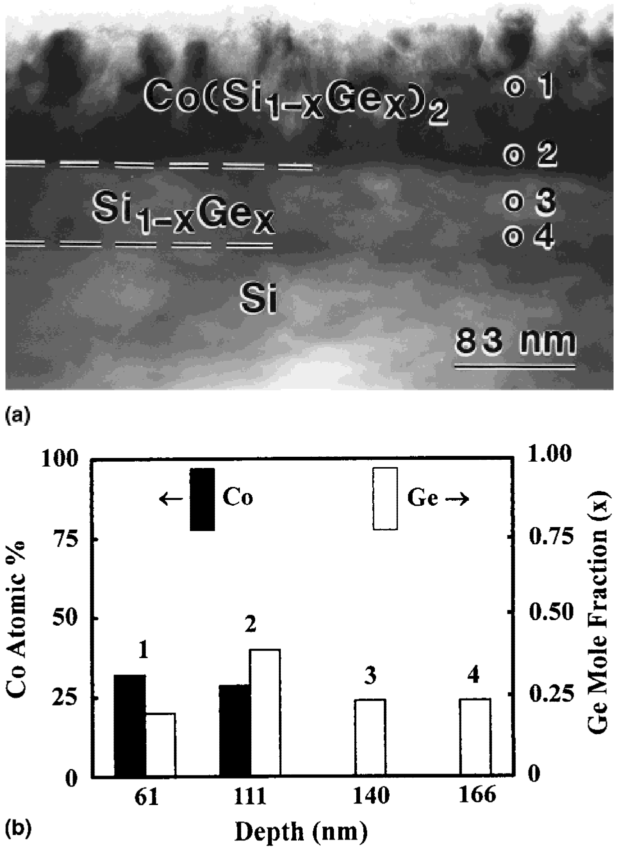

FIG. 4. (a) XTEM image and (b) the depth profiles of Co and Ge for the Co/$Si_{0.76}Ge_{0.24}$ sample annealed at 0.2 J/cm^2 for 20 pulses showing that a smooth Co($Si_{1-x}Ge_x$)$_2$ layer was formed without inducing Ge segregation. Ge mole fraction (x) means the mole fraction of Ge in $Si_{1-x}Ge_x$ for Co($Si_{1-x}Ge_x$)$_2$ and $Si_{0.76}Ge_{0.24}$ films, respectively. The areas probed by electron beam are denoted as "O" and designated as 1, 2, 3, and 4, respectively.

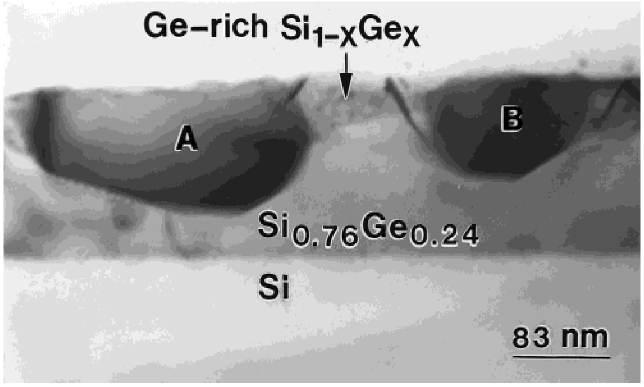

FIG. 5. XTEM image for the Co/$Si_{0.76}Ge_{0.24}$ sample annealed at 550 °C showing that the Co($Si_{1-x}Ge_x$) grains, A and B, are surrounded by Ge-rich $Si_{1-x}Ge_x$ film.

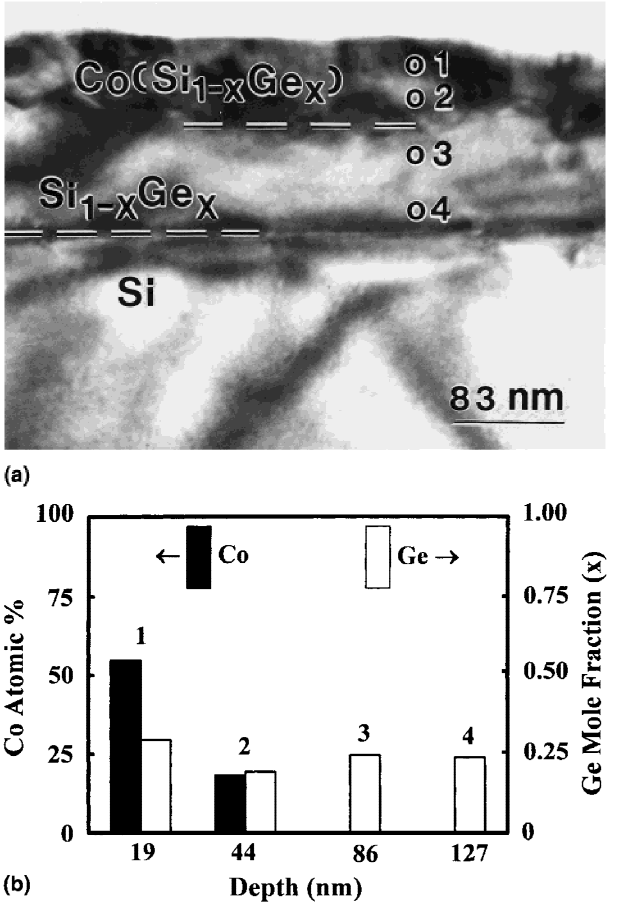

FIG. 6. (a) XTEM image and (b) the depth profiles of Co and Ge for the Co/$Si_{0.76}Ge_{0.24}$ sample annealed at 250 °C. Ge mole fraction (x) means the mole fraction of Ge in $Si_{1-x}Ge_x$ for Co($Si_{1-x}Ge_x$) and $Si_{0.76}Ge_{0.24}$ films, respectively. The areas probed by electron beam are denoted as "O" and designated as 1, 2, 3, and 4, respectively.

$Si_{0.76}Ge_{0.24}$ film, inducing strain relaxation in the residual $Si_{0.76}Ge_{0.24}$ film. At 0.8 J/cm^2, the reacted region was mostly transformed to a single layer of Co($Si_{1-x}Ge_x$)$_2$, whereas Ge diffused to the Si substrate. At 1.0 J/cm^2, constitutional supercooling appeared.

(2) For the Co/$Si_{0.76}Ge_{0.24}$ sample multiple pulse annealing at 0.2 J/cm^2 is a novel method to form a smooth Co($Si_{0.76}Ge_{0.24}$)$_2$ film without inducing Ge segregation out of the germanosilicide and apparent strain relaxation of the unreacted $Si_{0.76}Ge_{0.24}$ film.

(3) When the monogermanosilicide, Co($Si_{0.76}Ge_{0.24}$), was used as the starting material for laser annealing, the constitutional supercooling still occurred at energy densities >1.6 J/cm^2.

(4) The energy densities at which Co($Si_{1-x}Ge_x$) transformation to Co($Si_{1-x}Ge_x$)$_2$, Ge segregation to the underlying Si, and constitutional supercooling occurred were higher for the Co($Si_{0.76}Ge_{0.24}$)/$Si_{0.76}Ge_{0.24}$ system than for the Co/$Si_{0.76}Ge_{0.24}$ system.

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