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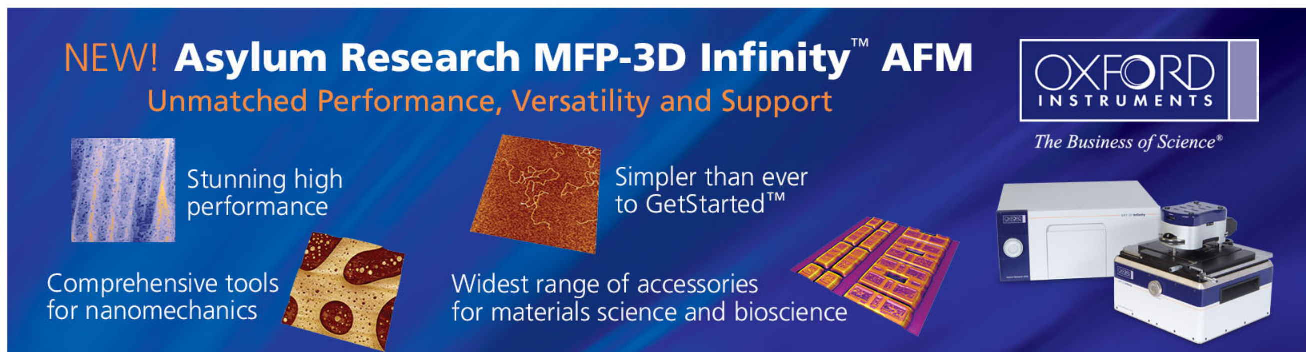

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Rapid self-assembly of Ni nanodots on Si substrate covered by a less-adhesive and heat-accumulated SiO_2 layers

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Rapid self-aggregation of Ni nanodots on Si substrate covered with a thin SiO_2 buffered layer is investigated. The Ni nanodots can hardly self-aggregate on highly heat-dissipated Si substrate with a thermal conductivity of 148 W/m K. Adding a 200-Å-thick SiO_2 buffer with an ultralow thermal conductivity of 1.35 W/m K prevents the formation of $NiSi_2$ compounds, enhances the heat accumulation, and releases the adhesion at Ni/Si interface, which greatly accelerates the self-assembly of Ni nanodots. Dense Ni nanodots with size and density of 30 nm and $7 \times 10^{10}\ cm^{-2}$, respectively, can be formatted after rapid thermal annealing at 850 °C for 22 s. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2336081]

Recently, the Al, Au, and Ag based metallic nanodot arrays[1–4] have been extensively used as either the nanoscale masks for fabricating quantum confined Si nanodots or the resonant surface-plasma-wave generator for biophotonic sensing applications. In particular, the self-assembly of Ni nanodots for potential-applications in field emission and biomagnetic sensing has also caused research interests. Previously, Lee *et al.*[3] have employed the Ni nanodot mask to fabricate Si nanorods on Si substrate with their diameter and height of about 41 and 472 nm, respectively. Nevertheless, the disadvantage of their process is that the self-aggregation of the Ni nanodots from the Ni film coated on the Si substrate by thermal annealing at 700 °C in N_2 ambient usually spends up to 10 min.[3] It was mentioned in previous experiment that the self-aggregation of the evaporated Ni film on pure Si wafer is hard to initiate without a long-term annealing process, while the density of the formatted Ni nanodots is too sparse for aforementioned applications. This is mainly attributed to the large thermal conductivity of the Si substrate that reduces the accumulation of heat at the interface between Ni and Si. In addition, the relatively strong adhesion between Ni and Si interfaces also restrains the self-aggregation of Ni nanodots. Such a long-term annealing inevitably leads to an unintentional doping or diffusion of the coated Ni atoms into Si substrate during the self-assembly procedure. On the other hand, it was noticed that a thin nitride layer such as Si_3N_4 (Ref. 4) or TiN (Refs. 5 and 6) could effectively prevent the formation of the $NiSi_2$ compound[7] and retain the heat within the nitride layer coated on Si substrate. In this letter, we propose a rapid process for Ni nanodots self-aggregating on Si substrate by depositing a thin SiO_2 buffered layer. The problems such as insufficient heat accumulation and strong adhesion at the interface of the coated Ni film and Si substrate can be solved, which essentially prevents the interdiffusion and greatly accelerates the precipitation of the evaporated Ni atoms.

In experiment, the RCA cleaning process is employed to completely remove the dusty particles and the native oxide from the *p*-type [100] Si wafer. Subsequently, a SiO_2 buffered layer with a thickness of 200 Å is deposited by using plasma enhanced chemical vapor deposition with a standard recipe. A 50-Å-thick Ni film is thermally evaporated on the SiO_2 covered Si substrate by using an e-beam coater with a biasing current of 70 mA and an evaporating rate of 0.1 Å/s. Afterwards, a rapid-thermal-annealing (RTA) process is performed at 850 °C under flowing N_2 ambient with a fluence of 5 SCCM (SCCM denotes cubic centimeter per minute at STP), which precipitates the Ni atoms into randomized Ni nanodots on the SiO_2 covered Si substrate. The morphology of Ni nanodots is analyzed by using a scanning electron microscope (SEM) (Hitachi FE-SEM S-5000) and an atomic force microscopy.

Figure 1 shows the SEM images of Ni nanodots self-

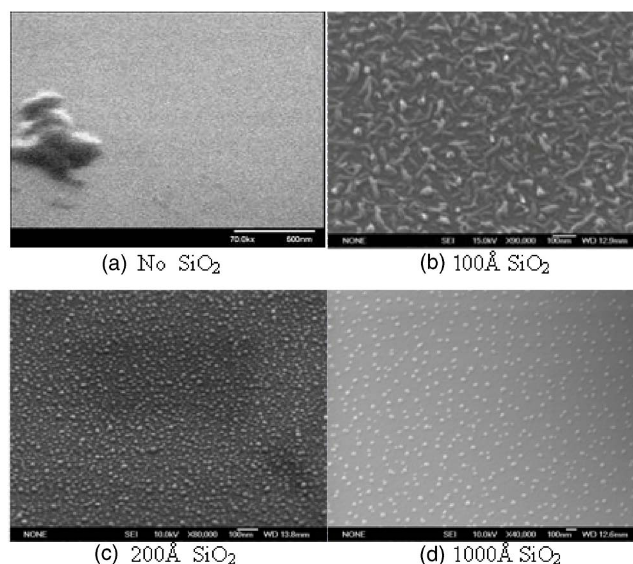

FIG. 1. Self-aggregation of Ni nanodots on Si substrates without or with SiO_2 buffered layer of different thicknesses.

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aggregating on SiO_2 covered Si substrate, the thickness of SiO_2 layer is varied, and the RTA condition of 850 °C for 120 s remains unchanged. As expected, Fig. 1(a) confirms that the Ni nanodots are unable to self-aggregate on Si wafer during such a short-term RTA process. In contrast, the self-assembly of Ni nanodots on all of the Si substrates covered with buffered SiO_2 of different thicknesses can apparently be observed. With a SiO_2 thickness of 100 Å, the evaporated Ni film initiates its self-aggregation to form a long strip during the RTA process, as shown in Fig. 1(b). However, the heat accumulation on such a thin SiO_2 layer is still insufficient to complete the reshaping of Ni nanodots during the RTA process. By increasing the thickness of SiO_2 layer from 100 to 200 Å, the coherent aggregation of Ni nanodots with a half-spheric morphology can be observed in Fig. 1(c). If the thickness of the SiO_2 layer enlarges to 1000 Å, the size of Ni nanodots becomes larger than those obtained on the thinner SiO_2 films, whereas the density of Ni nanodots greatly reduces. It is concluded from aforementioned results that the RTA induced self-aggregation of Ni nanodots can be achieved as the thickness of SiO_2 buffered layer increases to >200 Å, which is mainly attributed to the enhanced heat accumulation in thicker SiO_2 layer during RTA. Obviously, the self-aggregation of Ni nanodots on SiO_2 covered Si substrate is better than that on pure Si substrate due to the distinguishable difference in their thermal conductivities (148 W/m K for Si and 1.35 W/m K for SiO_2).[8] Therefore, the SiO_2 buffered layer can efficiently terminate the thermal dissipation from the evaporated Ni film to Si substrate such that the self-assembly of Ni nanodots can be finished at a shorter annealing period. In addition, we also observe that the diameter of Ni nanodots shrinks as the thickness of Ni film decreases.[2] At same RTA condition, the SEM images of the Ni nanodots self-aggregated from the evaporated Ni films with their thickness ranging from 30 to 200 Å are compared in Fig. 2. Although the larger Ni nanodots are obtained by evaporating a thicker Ni film, the size uniformity and the density of Ni nanodots are sacrificed simultaneously. It is concluded from experiments that the optimized thickness of the evaporated Ni film is around 50 Å.

In more detail, the size and distribution of the self-aggregated Ni nanodots on SiO_2 covered Si substrate are mainly determined by the thickness of the buffered oxide, the RTA condition, and the thickness of the evaporated Ni film. Although the accumulated thermal energy can be adjusted by changing either the annealing temperature or time, the best RTA temperature of 850 °C is observed for self-aggregating the Ni nanodots. Higher annealing temperature enhances the composition of the $NiSi_2$ compound[5] and degrades the uniformity in size distribution of the self-assembled Ni nanodots. In contrast, reducing the annealing temperature would inevitably lengthen the annealing time for self-aggregating the Ni nanodots. On the other hand, the effect of annealing time on size and density of the self-assembled Ni nanodots has also been characterized. The average size and density of Ni nanodots annealing at 850 °C for 30 s are 37 nm and 1×10^{10} cm^{-2}, respectively. However, the average size Ni nanodots annealing at 120 s significantly increase to 40 nm with a density decreasing to 7×10^9 cm^{-2}. This confirms the trend of enlarging size and decreasing density by lengthening the annealing time. Similar situation occurs in a thinner SiO_2 buffered sample. As an evidence, the SEM images of Ni nanodots precipitating on the 200-Å-thick SiO_2 buffered Si substrates after RTA at 850 °C from 20 to 26 s are shown in Fig. 3. The Ni nanodots cannot be formatted by RTA of shorter than 20 s. The Ni film initially breaks into large Ni strips and slowly approaches a hemispheric shape with increasing RTA time; however, these small Ni nanodots would eventually self-aggregate to enlarge their average diameter from 33 to 75 nm as the annealing time lengthens to >30 s. Figure 4 interprets that the optimized annealing time is 22 s

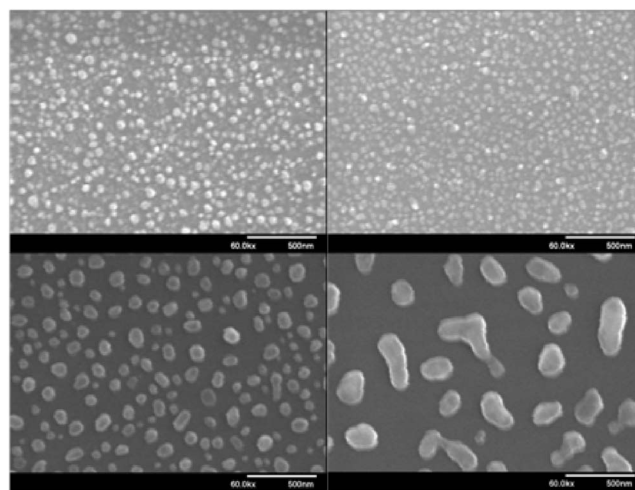

FIG. 2. SEM images of the samples with evaporated Ni film thickness of 30 Å (upper left), 50 Å (upper right), 100 Å (lower left), and 200 Å (lower right).

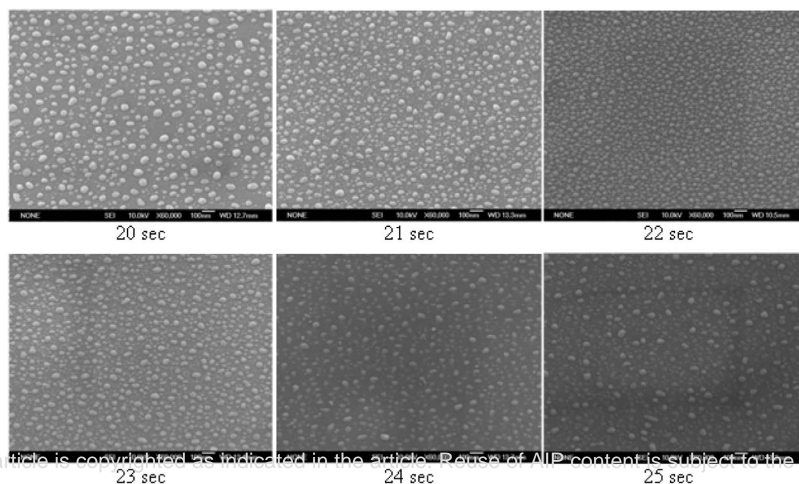

FIG. 3. SEM images of the self-aggregated Ni nanodots on 200-Å-thick SiO_2 covered Si substrates at different RTA durations.

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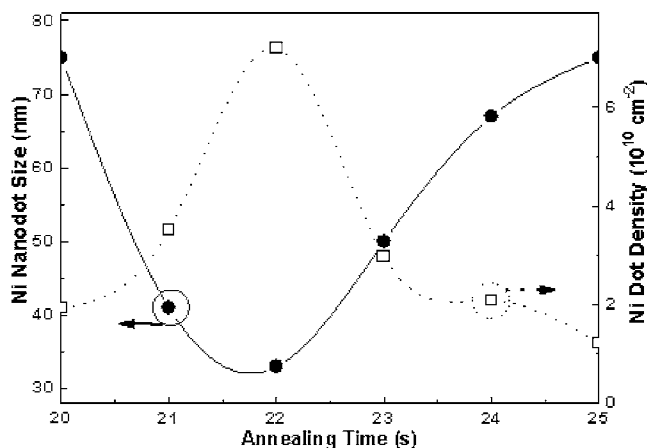

FIG. 4. Size and density of self-aggregated Ni nanodots as a function of RTA duration.

at 850 °C for self-assembling the Ni nanodots with a highest density of 7.2×10^{10} cm^{-2}, which are relatively comparable with the values ever reported under a far longer annealing duration.[1] The average size of 33 nm for the self-aggregated Ni nanodots is also much smaller than a previous record of 60 nm. The uniformity of Ni nanodots obtained by the RTA process is clearly better than previous works. Most important, the annealing time is greatly reduced from 10 min to 22 s with the aid of a SiO_2 buffered layer.

In conclusion, we demonstrate a rapid self-assembly of Ni nanodots on Si substrate by adding a highly heat-accumulated and less-adhesive SiO_2 layers. The Ni nanodots are relatively difficult to self-aggregate on Si substrate without a SiO_2 buffered layer due to the highly heat-dissipated feature of Si (with a thermal conductivity of 148 W/m K). The 200-Å-thick SiO_2 layer prevents the formation of $NiSi_2$ compound and facilitates the self-assembly of Ni nanodots, which accelerates the self-aggregation by retaining sufficient heat at Ni/SiO_2 interface due to the ultralow thermal conductivity of SiO_2 (1.35 W/m K). The annealing time required for precipitating dense Ni nanodots is greatly shortened to 30 s or less. The self-aggregated Ni nanodots with a highest density of 7.2×10^{10} cm^{-2} and a smallest size of 33 nm are obtained by RTA at 850 °C for 22 s. Raising the RTA temperature, lengthening the RTA time, or increasing the SiO_2 thickness inevitably causes enlarged size and dilute density of Ni nanodots.

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