Control of the Nucleation Density of Si Quantum Dots by Remote Hydrogen Plasma Treatment

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1. Research Target

The application of Si quantum dots as a floating gate to MOSFETs has been attracting much attention because it will lead us to new functionality such as multivalued memory operations even at room temperature [1]. The growth control of nanometer-scale silicon dots with an areal density as large as ~10^{12} cm^{-2} on an ultrathin SiO_2 layer is a crucial factor for the multivalued capability of the Si dots floating gate MOS devices. In our previous work, we demonstrated the fabrication of nanometer-scale Si dots on ultrathin SiO_2 layers by controlling the early stages of low-pressure chemical vapor deposition (LPCVD) using a SiH_4 gas [2]. Also we reported that the SiO_2 surface treatment with a dilute HF solution is very effective to obtain dot density above ~10^{11} cm^{-2} because Si-OH bonds created on the SiO_2 surface act as reactive sites to precursors such as SiH_2 during LPCVD. In addition, by spatially controlling OH- termination on the SiO_2 surface before LPCVD, the selective growth of Si dots has been demonstrated [3]. In fabricating multiply stacked structures of Si dots in SiO_2, it is very necessary to control Si-OH bonds on the SiO_2 surface by a dry process matching with subsequent LPCVD.

In this work, we demonstrate the feasibility of remote H_2-plasma pretreatment for controlling the areal density of Si dots.

2. Research Results

The SiO_2 surface was treated with a remote plasma of pure Ar and/or pure H_2. The plasma was generated by inductively-coupling between an external single –turn antenna attached to a 10 cmϕ quartz tube and a 60 MHz generator through a matching box. The substrate was placed on the susceptor at a distance of 32cm away from the position of the antenna. The RF power and the flow rate were kept constant at 200 W and 100sccm, respectively. The gas pressure was changed in the range of 0.1-1.0 Torr and the substrate temperature was varied from 27 to 540 ºC. The time of the remote plasma treatment is fixed for 5s to avoid the reduction of SiO_2 and to minimize plasma damages. The formation of Si dots on as-grown and plasma treated SiO_2 was performed by LPCVD using pure monosilane at 540 ºC. During the deposition, the gas pressure was maintained at 0.2 Torr. Figure 1 shows AFM images taken after Si dot formation on as-grown SiO_2 and remote plasma treated SiO_2. In the case on as-grown SiO_2, the Si dot density of 6×10^8 cm^{-2} was obtained. When the SiO_2 surface is treated with H_2 plasma prior to LPCVD, the Si dot density is markedly increased up to 7×10^{10} cm^{-2}. FT-IR-ATR spectra confirm the formation of Si-OH bonds by the H_2 plasma treatment. In the case of Ar plasma treatment in which ion bombardment may occur and induce some damages, the Si dot density is increased by a factor of 10. The results imply that an incidence of atomic hydrogen generated in H_2 plasma to the SiO_2 surface play an important role in the creation of the reactive sites such as OH bonds and hydrides for the Si dot formation. Note that the H_2 plasma treatment subsequent to the Ar plasma treatment provides a very uniform formation of Si dots with an areal density as high as ~1×10^{11}cm^{-2}. This is interpreted in terms of the improved coverage of OH bonds on the SiO_2 surface as confirmed by FT-IR-ATR measurements. It is likely that weaken bonds and dangling bonds created by Ar plasma exposure react efficiently with radicals, ions and excited molecules generated in H_2 plasma.

In conclusion, the combination of remote Ar plasma and subsequent H_2 plasma treatments is very effective to achieve a uniform size distribution of Si dots with an areal density of the order of 10^{11}cm^{-2}.

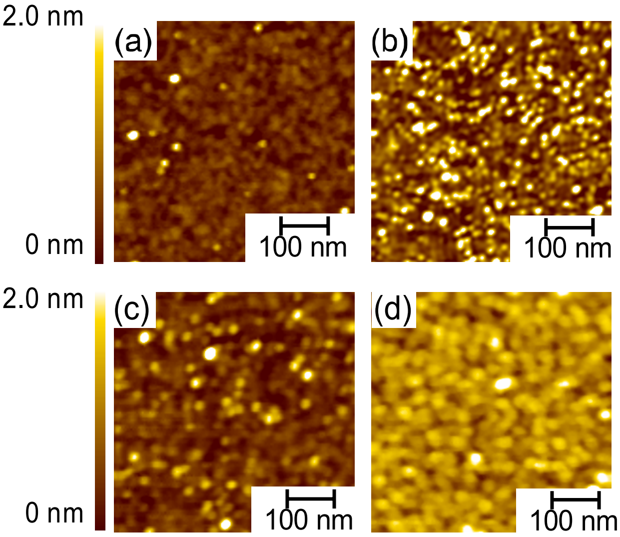

Fig. 1 AFM images of Si dots deposited on SiO_2 as-grown (a), remote H_2 plasma treated (b), Ar plasma treated (c), Ar plasma + H_2 plasma treated (d).

4. Conclusion

We demonstrated the control of the nucleation density of Si-QDs by remote H_2 and/or Ar plasma treatment. The density of Si dots was controlled from 6×10^9 to 7×10^{10} cm^{-2} by changing the substrate temperature and pressure at the remote H_2 plasma process. The combination of remote Ar plasma and subsequent H_2 plasma treatments is very effective to achieve a uniform size distribution of Si dots with an areal density of the order of 10^{11} cm^{-2}.

5. Relation of the COE program with the result

These results imply control of Si-QDs nucleation sites utilizing remote plasma treatment is very promising for fabrication of multiple stacked dot structure of Si-QDs.

6. References

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[2] S. Miyazaki et al., Thin Solid Films 369 (2000) 55.
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7. Published Papers and Patents

① Published Paper

1. K. Makihara, Y. Okamoto, H. Nakagawa, H. Murakami, S. Higashi and S. Miyazaki, "Electrical characterization of Ge microcrystallites by atomic force microscopy using a conducting probe": Thin Solid Films 457 (2004)103-108

② Proceedings

1. K. Makihara, Y. Okamoto, H. Nakagawa, H. Murakami, S. Higashi and S. Miyazaki, "Electrical Characterization of Ge Microcrystallites by Atomic Force Microscopy Using a Conducting Probe" SPSM-16. (2003.) B6-3, p115
2. K. Makihara, Y. Okamoto, H. Nakagawa, M. Ikeda, H. Murakami and S. Miyazaki "Local characterization of electronic transport in microcrystalline germanium thin films by atomic force microscopy using a conducting probe" AWAD2003. (2003). p37
3. K.Makihara, H.Deki, H. Murakami, S.Higasi and S.Miyazaki "Control of the Nucleation Density of Si Quantum Dots by Remote Hydrogen Plasma Treatment" ICSFS to be published.
4. K.Makihara, Y.Okamoto, H. Murakami, S.Higasi and S.Miyazaki "Characterization of germanium nanocrystallites grown on quartz by a conductive AFM probe technique" AWAD 2004 to be published.

③ Patents

1. K. Makihara, K. Takeuchi, M. Ikeda, H. Murakami and S. Miyazaki, The 20h Symposium on Plasma Processing p. 321.
2. K. Makihara, K. Takeuchi, M. Ikeda, H. Murakami and S. Miyazaki, The Japan Society of Applied Physics (The 50th spring meeting, 2003) No2, p975
3. K. Makihara, Y. Okamoto, H. Murakami, S. Higashi and S. Miyazaki, The Japan Society of Applied Physics (The 64th Autumn meeting, 2003) No2, p402
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