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## 主 論 文 の 要 旨

論文題目     **Study on Formation and Characterization of Fe-based Silicide Nanodots on Ultrathin SiO<sub>2</sub> for Functional Memories**  
(鉄系シリサイドナノドットの形成と機能性メモリデバイス応用に関する研究)

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## 論 文 内 容 の 要 旨

For the last several decades, microelectronics based on Si technology has been greatly developed in accordance with the development of advanced information society. The performance of Si ultralarge scale integration circuits (Si-ULSICs) has been growing exponentially by continuous downscaling of electronic devices for higher and higher levels of integration. As the integration level of circuits is more and more advanced, the physical problems and limitations become factors that hold back the development of microelectronics in many aspects. In addition, at the high integration level, the power consumption and the signal delay in the interconnect due to an increase in the resistance and capacitance of metal interconnection are also a stumbling block for evolution of ultralarge scale and high-performance chips, which are required for data processing and transfer via optical connections. Furthermore, the fundamental process for geometry scaling, photolithography is facing the most significant challenges.

Therefore, many constant efforts have been continued to overcome the obstacles to the continued growth of integration level. One of alternative promising approach is nanoelectronics which could provide novel unique advantages originating from unique physical properties and phenomena in nanometer-sized structures and materials, such as size effects, quantized energy level, Coulomb blockade phenomena. Some new devices such as single-electron transistor and dot floating gate memory have attracted considerable interest because these devices can lead to breakthrough of the scaling limitations. Therefore, the research on NDs as novel materials with unique optical, electrical, and magnetic properties has been extensively performed.

In this work, we focused on the formation of semiconducting and ferromagnetic metallic NDs further to improve the performance of Si-based nanoelectronic devices and to innovate the technologies for Si-ULSI. Considering compatibility with Si-based microelectronics technology, the development of Si-Based NDs is desired. Among Si-based materials, Fe-silicides have been studied extensively because of their various electronic and magnetic properties depending on their composition and phases. The purpose of this work is the formation of semiconducting and ferromagnetic Fe-silicide NDs by controlling the phase structures of Fe-silicide, and characterization of their physical properties.

In this thesis, the author described the formation of iron based silicides nanodots (NDs) for the new functional memory application. We demonstrated a novel technique for formation of Fe-NDs and Fe-silicide NDs (ferromagnetic  $\text{Fe}_3\text{Si}$  and semiconducting  $\beta\text{-FeSi}_2$  phase) on  $\text{SiO}_2$  with an areal density as high as  $\sim 10^{11} \text{ cm}^{-2}$ . These Fe based silicides NDs exhibit a potential advantage for the application to the magnetic NDs memory devices.

In chapter 1, the background and motivation of this work were introduced.

In chapter 2, the author introduced the formation method of metallic-NDs on SiO<sub>2</sub> by using H<sub>2</sub>-RP, and the physics behind the methodologies, subsequently, explained characterization methods of structural and physical properties of NDs.

In chapter 3, the author described successful formation of Fe-NDs, and discussed how effective remote hydrogen plasma treatment is on metal NDs formation. During the remote H<sub>2</sub> plasma exposure, the surface temperature rose up to ~500 °C because of the surface recombination of atomic hydrogen, which enhanced surface migration of Fe atoms and promoted self-assembling of NDs. Change in the surface potential after the charge injection from the conductive AFM tip was confirmed by using an AFM/Kelvin probe technique, indicating electrical separation among Fe-NDs. Furthermore, from results of magnetic characterization by using a magnetic force microscopy, we confirmed that Fe-NDs acted as active elements not only for the charge storage but also for the spin storage.

In chapter 4, the author demonstrated the formation of high-density Fe-silicide NDs on thermally grown SiO<sub>2</sub> by exposing a Fe/a-Si/Fe trilayer stack structures to the remote H<sub>2</sub> plasma without any external heating and characterized their silicidation state and crystalline phase. The silicidation reaction was induced by the remote H<sub>2</sub> plasma exposure, which was accompanied by the agglomeration of Fe and Si atoms on the SiO<sub>2</sub> surface. The formation of a crystalline β-FeSi<sub>2</sub> phase was confirmed. The electrical separation among the β-FeSi<sub>2</sub> NDs was confirmed from changes in the surface potential due to the charging of the dots. The surface potential of the NDs was changed in a stepwise manner with respect to the tip voltage applied for charge injection by using

AFM because of the multistep electron injection to and extraction from the semiconductor  $\beta$ -FeSi<sub>2</sub> NDs.

In chapter 5, the author described the method to control of the crystalline phase of the Fe-silicide NDs, in which he extended the results of Chapter 4 to the formation of high density Fe-silicide NDs on SiO<sub>2</sub> by controlling the ratio of initial film thickness of Fe/a-Si/Fe trilayer stacks and H<sub>2</sub> gas pressure. The influences of the H<sub>2</sub>-RP gas pressure on magnetic properties and the crystal phase of Fe-silicide NDs were investigated. With increasing H<sub>2</sub> pressure, namely the surface temperature of the sample, saturation magnetization, M<sub>s</sub>, was increased to reach its peak, and then decreased. From the results of XRD measurement, we can conclude that the reasons of the increase and decrease in M<sub>s</sub> are mainly explained by the growth of ferromagnetic Fe<sub>3</sub>Si phase and formation of nanocrystalline  $\epsilon$ -FeSi phase which is nonmagnetic at room temperature, respectively. These results suggest that, by controlling the thickness ratio of Fe/a-Si/Fe trilayer stack structure and gas pressure of H<sub>2</sub>-RP, the crystalline phase of Fe-silicide NDs can be controlled.

In chapter 6, as a summary, this technique is very effective for the low-temperature fabrication of various phases of Fe-silicide NDs. However, there remain several issues that could not be accomplished within the experimental work of this thesis.

In order to realize the application of the Fe-silicide NDs to the practical devices such as floating gate memory, light-emitting diode and Si-based spintronic devices, sophisticating the nanodots formation and the crystalline phase control processes is mandatory. Further examination of phase structure and physical properties of Fe-silicide NDs may lead to discovery of novel functions unique in Fe-silicide NDs. Throughout this work, we have found that the initial thickness of Fe films and

parameters of H<sub>2</sub>-RP are critical factors for control not only of dot density but also of phase structures. The optimization of these factors and the establishment of Fe-silicide NDs phase diagram are needed.

Since a Fe/a-Si/Fe trilayer stack structures were used and the agglomeration of Fe and Si atoms for the formation of NDs occurred in a three-dimensional direction at nonequilibrium state during H<sub>2</sub>-RP exposure, the diffusion and silicidation reaction mechanism in the interface region of the trilayer become very complicated, which needs more detailed investigation.

For the  $\beta$ -FeSi<sub>2</sub> NDs, which are expected to be applied to the light-emitting devices, the light emission characteristics should be evaluated by luminescence measurement. In the future, we will fabricate a three-dimensional stacked structure with the  $\beta$ -FeSi<sub>2</sub> NDs separated by ultrathin Si oxide interlayers to achieve the efficient light emission.

Although we have obtained the crystalline Fe<sub>3</sub>Si NDs, in which the value of saturation magnetization was very close to the DO<sub>3</sub> ordered Fe<sub>3</sub>Si thin film (50-nm-thick), we have to find a way to improve the ordering level of the DO<sub>3</sub> ordered Fe<sub>3</sub>Si NDs to improve the spin polarization ratio. For the application of Fe<sub>3</sub>Si NDs to memory devices such as spin dependent floating gate memory, electronic transport characteristics should be examined in magnetic tunnel junction with the NDs.

The author hopes that the research efforts could provide guideline for the formations of various phase metal-silicides or -germanides NDs based Si or Ge, which offers some opportunities in a next generation information technologies employing semiconductor or spintronic devices with innovative concepts.