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Formation of Clusters of Impurity Atoms of Nickel in Silicon and Controlling Their Parameters

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Abstract The paper studies the possibility of self-assembly of micro-particles of impurity atoms of nickel in silicon at certain thermodynamic conditions. It was shown that one could set the size, concentration and distribution of clusters of impurity atoms in the silicon lattice. Also, the effect of assembly of clusters was revealed.

Keywords Single crystalline silicon, Nanoscale structures, Self-organization of clusters of impurity atoms, Ni clusters

1. Introduction

The phenomenon of self-assembly of nanoclusters of impurity atoms interacting with lattice defects in the crystal lattice of semiconductor is likely to be one of the promising and novel methods for the development of nanoscale structures.

The possibility of building clusters of impurity atoms in semiconductors is currently being actively investigated by many researchers [1-5]. The interest in the assembly of nanoclusters is largely motivated by the desire to develop nanosize structures and control their parameters, which in turn will have given the possibility to develop novel elements for micro- and nanoelectronics. One can evidence that shaping of clusters of various nature significantly depends on solubility, diffusion parameters of impurity atoms and thermodynamic conditions as well. However, virtually no major research has been conducted over lately on the possibility to regulate the parameters of such clusters.

Clusters of impurity atoms in silicon were largely obtained by the technique of homo-epitaxial growth or molecular-beam epitaxial growth. The authors of the present research work propose the technology of obtaining of self-building impurity clusters by diffusion. The above method in contrast to the existing method of molecular-beam epitaxy has certain advantages and does not require complex and expensive equipment. The method allows:

- to produce nanoscale structures throughout the entire bulk of the crystal;
- to set easily the structure, composition, distribution and ordering of nanoscale clusters;
- to obtain magnetic nanoclusters with adjustable magnetic moment, i.e. a novel magnetic semiconductor material;
- to control the charging state of nanoclusters in the broad range (N+(-)n, где n>3), thus producing multiply charged centers in the semiconductor, which might serve as the basis for promising novel material for nanophotonics.

The possibility of building clusters of impurity atoms of Ni in silicon and controlling their parameters is currently investigated in the present research article. The choice of the impurity atom is preconditioned by the fact that firstly, its solubility and the diffusion coefficient in silicon is comparatively higher than that of other elements of the Fe group, and secondly, Ni will most probably turn out to be one of the most important metals for the semiconductor industry in the near future.

2. Main Body

2.1. Theoretical Analysis

Over the past few years, there has been a widespread interest among experts in the field of nanotechnology and nanoelectronics all over the world in the technology of self-organizing impurity clusters with manageable structures and magnetic properties. In this respect one can note some interesting results related to implanting Co and Ge ions on Si, ion implantation in other semiconductor materials [6-8]. As we have heard, the technology of self-organization of clusters of impurity atoms by using the diffusion technologies currently is not sufficiently studied. Diffusion technology for producing nanoscale structures is not only a

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more affordable and cheap technique, allowing large-scale production, but also gives the ability to synthesize nanoscale structures of various type (2D, 1D, 0D), as well as specify the distribution and density over the bulk of the crystal.

Therefore, the main objective of this research is to demonstrate that under certain doping conditions one can witness shaping of clusters which also gives us the opportunity to manage parameters of those clusters.

2.2. Experimental Part

Our group had developed a special technique for doping, the so-called "low-temperature doping" of semiconductors. This method of doping is based upon the diffusion process which is carried out in stages by gradually increasing temperature ranging from room temperature to the diffusion temperature [9].

The native sample and dopant respectively (pure metallic Nickel of certain weight (to be determined by ampoule volume)) are put into evacuated quartz ampoules (pressure ~10^{-6} mm.of mercury column), which are inserted into the diffusion furnace at T=300K. Shortly thereafter starting from T = 300 K the furnace temperature at the location of the ampoule is gradually increased at a rate of 5°C / min and preheated to the temperature on intermediate processing and this temperature is retained to a certain period, then the furnace temperature is increased quickly at the rate t = (15÷20) C/min. and so up to the diffusion temperature (1523°C) and the samples were retained under this temperature during 40 minutes. After having completed all the above stages the ampoules are removed from the furnace and cooled off. Such doping conditions ensure embedding maximum concentration of Ni in the Si lattice and uniform doping through the entire bulk of the crystal.

For the experiment we used single crystalline silicon of both n- and p-type with phosphorus concentration $N_P=10^{13}\div10^{17}cm^{-3}$ and the boron concentration $N_B=10^{14}\div10^{17}cm^{-3}$. The native samples had a minimal dislocations density which was $N_D<10^2$ cm^2.

Also, we used single crystalline silicon with various concentration of oxygen atoms $N_{O2}=10^{17}$ cm^{-3} and $N_D<10^{16}$ cm^{-3}

The choice of impurities of various concentrations was conditioned by the necessity to determine what role the type and concentration of impurity atoms do eventually play in building a cluster. The samples were doped in identical circumstances by using the above technology.

After diffusion the samples were subjected to low-temperature annealing, then polished for visual inspection by infrared microscope INFRA-N of the state and structure of impurity atoms of Ni in the lattice. The experimental studies have evidenced that under proper doping and annealing conditions (doping duration, cooling rate and annealing temperature) the experimental results appear to be 100% reproducible.

2.3. The Results and Discussion

Research of state of nickel atoms in the samples was carried out by infrared microscope at room temperature. The results of investigations showed that in samples doped at 1250°C no formation of clusters of Ni in the lattice of Si was documented and perhaps it is due to the limited resolution of the infrared microscope.

The doped samples were subjected to additional low-temperature annealing at T=600°C÷1100°C during t=1÷3 hours in air. Then, after removing about 50 microns from the surface, all samples were subjected to optical polishing for further infrared microscope investigation.

The results evidenced that in all samples that were subjected to low temperature annealing there occurs clustering of Ni atoms in the lattice of Si which allowed to reveal the following pattern:

1) The geometric dimensions of the clusters increase with decreasing annealing temperature and varies from 0,5÷0,2 microns to 2 ÷ 5 microns;

2) The density of clusters decreases as the temperature of annealing decreases;

3) Clusters are distributed fairly evenly over the entire surface;

4) The size, density and distribution of clusters does not depend on the type and concentration of initial impurity atoms.

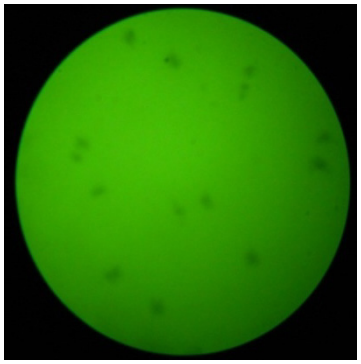
Meanwhile, it was important to find an answer to following question: How are they distributed in the bulk of the crystal? Do they exist only on surface?

To answer these questions, we conducted X-ray microprobe analysis on Jeol Super Probe YXA-8800R/RL analyzer. It had been revealed that the micro-particles consist of Ni atoms and Si at ratio ~60% to ~40%, respectively. It should be noted that nickel does not appear across the entire silicon surface but in some areas, i.e. in locations where the clusters are formed (determined by infrared microscope). This leads us to conclude that under certain conditions of low temperature annealing one can really witness self-organizational formation of nickel atoms into cluster.

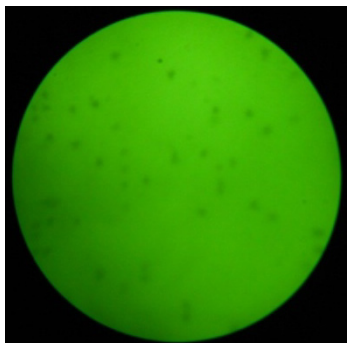
To examine the character of distribution of micro-particles in the bulk, the samples were subjected to surface polishing resulting in 15 ÷ 20 microns off each. After every step of grinding the samples were polished and studied under infra infrared microscope INFRA-N. The practice continued until the removal of more than a half of the thickness of the samples (about 350 ÷ 300 microns). As evidenced by the results of investigations, the size, density and distribution of micro-particles across the crystal is almost uniform, i.e., only coordinates of their location on the surface does changes. This implies that the formation and self-organization of clusters occurs over the entire volume of the crystal, and they are fairly uniformly distributed. The above data allowed us to calculate the concentration of clusters. The calculations show that their concentrations vary in the range of N=109 ÷1010 cm-3 to N=1012 ÷1013 cm-3. Having known the solubility of Ni in silicon and the temperature of annealing T=12500C (N_0) and at the temperature of annealing (NT), we have calculated number of atoms in clusters. In the meantime, we have assumed that all non-equilibrium

concentration of Ni atoms at temperature of additional annealing participate in the formation of clusters. Thus, number of atoms in clusters equals: $n = \frac{N_0 - N_T}{N}$, (where N is the concentration of clusters)

It was revealed that in clusters, depending on their size, one can assume the existence of several hundred to million atoms. The calculations lead us to conclude that in samples subjected to thermal annealing at T>1100°C, where we could not detect any clusters by using conventional IR microscope, one can still anticipate the availability of nanoclusters size 5÷100 nm. The results indicate that, under certain conditions of doping and heat treatment one can evidence self-organization of clusters of Ni impurity atoms throughout the entire volume of the crystal. There is the possibility to control their size, concentration and distribution (Fig. 1. a,b,c.).



a)



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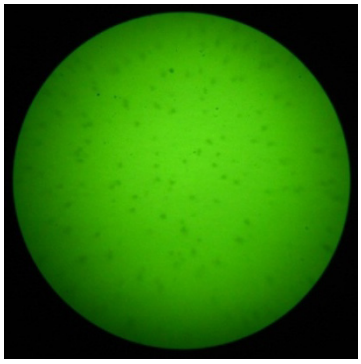
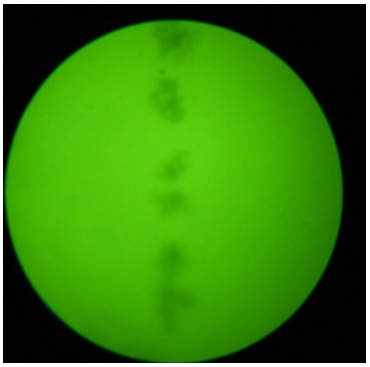


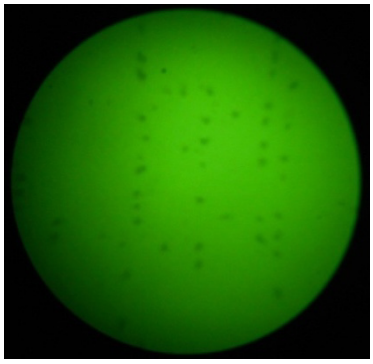
c)

Figure 1. Microphotograph of micro-particles of Ni atoms in Si. Magnification: x1000

Interesting results were obtained at additional low-temperature heat treatment at T=700°C. Thus, ordering of impurity clusters of nickel atoms in silicon occurs as a function of time of heat treatment (Fig. 2 a,b,c.).



a)



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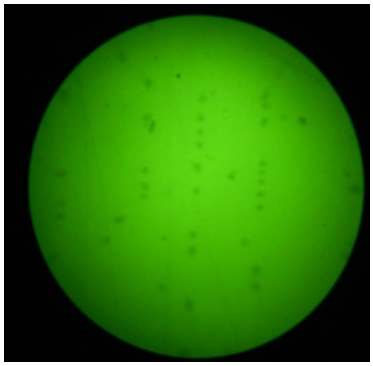

c)

Figure 2. The state of micro-particles of Ni atoms in Si after additional annealing at T = 700°C during 3 hours. Magnification: x1000

The results of investigations indicate that such ordering of clusters of impurity atoms occurs throughout the entire bulk of the crystal. So far, no physical mechanism is available to explain this phenomenon. However, the process of ordering of clusters leads us to conclude that there is the possibility of diffusion of clusters with relatively higher ration of diffusion. However to prove the above results one needs to carry out detailed thorough research and investigations. It was figured out that as the temperature of additional heat treatment increases the ordering of clusters collapses again and they become evenly distributed again. Preliminary investigations in I-V-curve of cluster – Si by microprobe have shown that

such a structure is characterized by almost ideal I-V-curve with very little reverse current and it starts to manifest sensitiveness to irradiation and temperature.

3. Conclusions

The experience evidenced that in silicon samples doped by Ni under the technology of staged diffusion and after additional low-temperature annealing, one can witness self-assembly of clusters of impurity atoms of Ni in the lattice of Si. By controlling the annealing temperature one can change the cluster sizes in the wide range d = 0,2÷5 microns, their distribution in the bulk, as well as their concentration whereas at T = 700°C a unique phenomenon, i.e. ordering of clusters occurs.

Formation of clusters and their parameters are practically independent of the type and concentration of the initial impurity atoms, which allows us to conclude that the initial impurity atoms do not play a role in the formation of clusters.

Physical mechanisms of the formation of clusters and their ordering, as well as the structure of clusters require more detailed research and investigation. Of great practical interest is the possibility to use such clusters as micro- and nano- Schottky barriers in the development of a novel class of microelectronic devices and integrated photocells.

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